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MOX Fuel Cycle Technologies for Medium and Long Term Deployment

INTERNATIONAL ATOMIC ENERGY AGENCY

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FOREWORD

An International Symposium on MOX Fuel Cycle Technologies for Medium and Long Term Deployment was held in Vienna from 17 to 21 May 1999. The symposium was organized by the International Atomic Energy Agency in co-operation with the OECD Nuclear Energy Agency. More than 150 participants from 28 countries and four international organizations took part.

The purpose of the symposium was to exchange information on MOX fuel cycle technologies worldwide with focus on how past experience has been or can be used to progress further, either for facing more demanding fabrication and utilization conditions or for extending into new processing or utilization domains. Present technologies of MOX fuel fabrication, fuel design, performance, testing, in-core fuel management, transportation, safety analysis, safeguards and MOX fuel cycle options, including back end, were covered by the invited overview papers describing the worldwide status of the topics. Contributed papers concentrated on the differences between MOX and UO_2 fuels and focused on the future, on the basis of today's perspectives and developments.

The place of plutonium recycle in the context of the whole nuclear fuel cycle activity under present conditions of a deregulated electricity market and in the future and its role in the reduction of separated civil and surplus ex-weapons plutonium stockpiles were examined further in the discussions by participants and a panel of experts.

The IAEA officer responsible for this publication was V. Onoufriev of the Division of Nuclear Fuel Cycle and Waste Technology.

EDITORIAL NOTE

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OPENING SESSION

OPENING ADDRESS

V.M. Mourogov
International Atomic Energy Agency

It is my pleasure to welcome you to this International Symposium on MOX Fuel Cycle Technologies for Medium and Long Term Deployment, which is organized by the IAEA in co-operation with the OECD's Nuclear Energy Agency.

Starting from the famous International Fuel Cycle Evaluation Study of 1976–1980 the different aspects of plutonium utilization in the form of MOX fuel have been discussed in the many international meetings during last two decades.

The great interest in the plutonium issue in the world is quite understandable. From one side a continuing accumulation of unused plutonium may present a future challenge to the world community because of an increasing risk of its use for non peaceful purposes. From the other side only with plutonium use may one involve practically unlimited resources of fertile uranium 238 in energy generation. G.T. Seaborg defined plutonium as a “key that unlocks the energy potential of uranium”.

This Symposium, is supposed to discuss the entire spectrum of issues related to the MOX fuel cycle, namely the reprocessing and fabrication technologies, utilization of civil and ex-weapon plutonium, performance, safety, safeguard and potential advanced cycle options.

Let me only mention some topics that you are going to elaborate during the meeting.

Interest to use of plutonium, accumulated in uranium fuel under irradiation in nuclear reactor, appeared simultaneously with commissioning of the first power reactors. Initially plutonium was sought to be used primarily in fast breeder reactors (FBRs), where its recycling increases the energy available 50 fold or more, making fission energy by far the largest energy resource available; indeed, a virtually inexhaustible one.

At the beginning plutonium use in the form of MOX-fuel in thermal reactors was taken only as an alternative back-end policy option. Plutonium use in thermal reactors also increases uranium efficiency, but only less than twice. However, today the plutonium recycling with LWRs has evolved to industrial level in some countries relying on the recycling policy. This happened mainly because of the delays in development of FBRs and a desire of a partial return on investment done in reprocessing facilities.

Today more than 30 BWRs and PWRs (in Belgium, France, Germany and Switzerland) have been loaded on a routine basis with MOX fuel. Their number is increasing and so Japan will start loading of MOX fuel in a BWR and in a PWR in 1999 or 2000. The maximum share of MOX fuel assemblies in cores of different LWRs is varying from 30 to 50%.

At present we can undoubtedly state that MOX fuel fabrication technology for LWR is now a proven technology. The total annual world capacity accounts for about 220 t of MOX fuel. In the year 2000 a worldwide increase of annual MOX fuel fabrication capacity up to about 430 t HM/a is expected essentially in France (extension of the MELOX plant) and the UK (start-up of the SMP plant). Further capacity increases can be expected from Japan and the Russian Federation

MOX fuel use in thermal reactors helped to accumulate efforts in mastering technologies of MOX fabrication and radioactive waste disposal. These achievements are of great importance for future nuclear power development. Reprocessing of spent fuel and extraction of plutonium leads to decrease of the amount of radioactive waste to be disposed of. These wastes that mainly consist of fission products could be disposed in vitrified form.

However, this scheme of utilization of MOX fuel only in thermal reactors will not resolve the issues related to accumulation of quantities of discharged spent fuel and separated Pu to be stored. In 1997 from 10.500 t of discharged spent fuel and 24 t of separated Pu, only 3.000 t of spent fuel were reprocessed and 9 t of Pu were used to fabricate MOX fuel. It means that in this year 7500 t of spent fuel and 15 t of Pu were transported to storage facilities.

It is expected that in the near future developments in the nuclear fuel cycle, such as the fuel burnup increase, introduction of "DUPIC" fuel cycle, and use of MOX fuel in CANDU reactors, or second recycling of MOX fuel in LWRs may additionally contribute to reduce these annual increments of separated Pu. Nevertheless, the imbalance between the separation and use of plutonium as MOX fuel is expected to increase.

In this respect, the IAEA continues activities on development of advanced reactor systems, including fast reactors, to keep the subject of burning MOX-fuel or Minor Actinides. Of course, decisions on reprocessing and MOX fuel fabrication scheme's contracts belong to industry and depend on both economics of specific fuel cycles and on the political situation and acceptance of MOX fuel activities the by public in respective countries. The IAEA could contribute into increase of transparency in the management of plutonium and openly published inventory information on plutonium in order to build public and international confidence.

The "availability" of additional dozens tones of ex-weapons plutonium surplus in the USA and in Russia in coming future sheds a different light on the MOX issue. Both countries decided to dispose part of this ex-weapon plutonium in the civil reactors. The rate of implementing this disposition option depends on mutual position of both countries and the readiness to use MOX fuel and, to some extent, on efforts in other countries, such as of France, Belgium Germany, experienced in MOX fuel fabrication technology. The IAEA is active in discussion of safeguard's aspects of ex-weapons surplus Pu, but could be also a forum to discuss other technical issues.

Upon advice of a Standing Advisory Group on Spent Fuel Management the Agency has conducted activities on spent fuel from power and research reactors, including spent fuel arising, spent fuel reprocessing and conditioning technologies, and transportation of irradiated MOX fuel. Development in MOX fuel design, technology, performance and utilization has been the focus of the Agency's activities upon the recommendations by the International Working Group on Water Reactor Fuel Performance and Technology.

The IAEA's Safeguards Department is actively pursuing a trilateral arrangement for safeguarding ex-defense surplus Plutonium. In 1998, the Agency established the International Working Group on Nuclear Fuel Cycle Options to provide a forum for discussion on the disposition options of separated plutonium inventories, from both civil and ex-defense sources. At this Symposium, issues associated with disposition of ex-defense plutonium will be presented by experts from Russia and the United States.

The IAEA Department of Nuclear Safety jointly with the Department of Nuclear Energy conducts a Study on fuel safety criteria for innovative WWER fuel designs including the use of MOX fuel.

Approaching to the end, I would like to remind you what efforts in the area of separated Pu and MOX fuel management were formulated by the International Symposium on Nuclear Fuel Cycle and Reactor Strategy: Adjusting to New Realities in 1997 in order adjust to "new realities" in the nuclear energy sector:

- Major international co-operative programmes to use or dispose of excess weapons plutonium;
- Addition of new capacities to fabricate and utilize MOX fuel, to correct plutonium imbalances;
- Increased transparency in the management of plutonium and openly published inventory information on plutonium, to build public and international confidence;
- Continued R&D on fast reactors;

- Continued research on advanced fuel cycle approaches, and pursuit of increased fuel burnups for both uranium and MOX fuels, to reduce spent fuel arising and fuel cycle costs; and
- Continued development of improved environmental management and safeguards technologies, to ensure safety and security in whatever fuel cycles are implemented.

I think that this Symposium will provide us opportunity to contribute to the effort which major directions I has just mentioned.

More than 150 participants from 28 countries and 3 international organizations are attending this Symposium. In total, 47 oral papers and 21 posters reported on the current status of the technology and some of the major issues encountered for further development. This reflects the worldwide interest in this important topic.

I would like to thank the group of MOX fuel specialists organized by International Working Group on Water Reactor Fuel Performance and Technology and chaired by Mr. Hubert Bairiot from Belgium which contributed a lot into preparation of the Symposium. As I mentioned above, this Symposium has been prepared in close co-operation with OECD/NEA and the IAEA Department of Safeguards and Department of Nuclear Safety.

OPENING REMARKS

P. Wilmer
OECD Nuclear Energy Agency

I would like to add my support to Mr. Mourogov's welcome address to this International Symposium on MOX Fuel Cycle Technologies for Medium and Long Term Deployment. It is very pleasing that the IAEA and the OECD/NEA are working jointly to develop this very interesting, challenging and fast moving facet of nuclear development to ensure sharing of views in an efficient and co-operative way.

Mr. Philippe Savelli, who had been expecting to be here today, sends his apologies for not being able to attend the meeting and to speak to you personally but nevertheless joins with me in expressing the appreciation of OECD/NEA for your support in participating in this Symposium. A special "thank you" goes to those people who have developed and organized the meeting and to those who will be presenting papers.

This is a technical meeting reflecting the current state of the practice of recycling plutonium into MOX and it is appropriate that I reflect a little on the past and present activities of NEA in this area. Early work, of course, was concerned with fast reactor studies from which current, successful LWR MOX technology emerged once it became evident that quantities of stored plutonium from thermal reactor would increase over a considerable time period, decades at least, whilst fast reactor technologies were not being widely deployed.

The strategic options for plutonium management and the logistical issues involved have been addressed by expert groups reporting in 1989 and in 1997, "Plutonium Fuel: An Assessment and "Management of Separated Plutonium: The Technical Options" respectively. The economic dimensions, entirely appropriate for an Agency within the OECD family, was addressed within the broad ranging study of "The Economics of the Fuel Cycle" in 1994. I also envisage that MOX fuel will comprise an important element in our new project "An Integrated Assessment of the Nuclear Fuel Cycle" just starting. In addition, some aspects of MOX usage will be addressed by our PARCOM project which is currently assessing the radiological impact of a variety of fuel cycle options and is expected to report early next year.

There are various facets to our work at the scientific level. Speaking just briefly, we have contributed basic nuclear data on the various isotopes of plutonium of interest and the most recent data are being incorporated in our JEFF-3 library by our Data Bank team for the benefit of its users. Modelling has been an extensive activity which has taken the form of international benchmarking studies to address the calculational and computational methods available for specific reactor configurations, addressing the real issues for today. These include fuel pin cell calculations with a variety of plutonium vectors, reactivity void effects in PWRs and physics limitations of multiple recycling in various scenarios. The contributions
of 70 experts have been incorporated and the work has resulted in a six volume publication, "Physics of Plutonium Recycling".

Basic phenomena studies have been undertaken. Key amongst those have been thermal performance modelling, fission gas release, swelling and pellet-clad interaction effects as fuel irradiation levels have been increased. This work has led to joint seminars with the IAIEA. Finally, criticality safety has been specifically studied.

I would like to turn now to some more general comments. MOX fuel use is becoming a mature technology and it is interesting to reflect on the environment within which it resides. In this sense, it is an alternative to the more established, more widely deployed and more advanced uranium technology. There is undoubtedly opportunity to harmonize performance parameters between the two to bring wider and more flexible operational choices for plant operators. Another positive feature is the

increasing quantities of plutonium which is coming about by the release of military material to the civilian cycle. This may lead to the involvement of more plants and more countries in the development and the implementation of MOX fuel technology.

There are concerns and prospective changes in the picture. The concerns for the technology relate to the inevitable and intimate linkage of MOX fuel deployment to reprocessing activities on a commercial scale and the unavoidable transport of plutonium in separated form or as fresh fuel. Some countries have previously made the choice not to participate in this and others now seek not to, having earlier decided otherwise. This latter action subjects the deployment of MOX technology to particular scrutiny and has the potential to have repercussions for the entire nuclear industry. The technology that we are developing
with MOX is part of the broader nuclear industry activity on which judgments will continue to be made. It is important that plutonium recycle is done well and widely recognized as valuable and trusted as part of establishing an acceptable legacy for the current generation of plants alongside safety, decommissioning and radioactive waste disposal.

Finally, the change dimension. I would like to touch upon the evolution of the electricity supply industry into a new deregulated and competitive world. We see the nuclear power industry being increasingly exposed to new pressures and obligations on a commercial front whilst the regulatory constraints are held firm or strengthened. Strategic decisions taken in the past, sometimes decades ago, will come to be revisited and the financial pressures, those are economic and others, will be more firmly applied alongside the more explicit control of business risk. The use of MOX fuel in our plants today will not be exempt from these changes and the technology must be robust to such external developments. It is a challenge for all of us to ensure that it is.

May I wish you a very stimulating and productive time at the Symposium, both within the confines of the meeting and with your colleagues having similar interests from around the world.

Invited Paper

MOX FUEL USE AS A BACK-END OPTION: TRENDS, MAIN ISSUES AND IMPACTS ON FUEL CYCLE MANAGEMENT

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Abstract

In the past decades while the FBIULWR fuel cycle concept was zealously being developed, MOX-fuel use in thermal reactors was taken as an alternative back-end policy option. However, the plutonium recycling with LWRs has evolved to industrial level, gaining high maturity through the incubative period while FBR deployment was envisaged. Today, MOX-fuel use in LWRs makes integral part of the fuel cycle for those countries relying on the recycling policy. Developments to improve the fuel cycle performance, including the minimisation of remaining wastes, and the reactor engineering aspects owing to MOX-fuel use, are continued. This paper jointly presented by IAEA and OECD/NEA brings an integrated overview on MOX use as a back-end policy, covering MOX fuel utilisation, fuel performance and technology, economics, licensing, MOX fuel trends in the coming decades.

1. INTRODUCTION

Early in the development of nuclear power in the 1960s the closed fuel cycle scheme was perceived as the best option for nuclear energy development. With anticipation of a rapid growth of nuclear power capacity and the deployment of Fast Breeder Reactors (FBRs) in the decades to come, commitments were made to construct and operate reprocessing facilities. Today, however, the lower than expected growth rate of nuclear power capacity and the delays in the development of FBRs create a significant imbalance between the rate of the plutonium separation in reprocessing and the rate of plutonium utilization as nuclear fuels. With inventories of separated plutonium accumulating, the recycle of plutonium in MOX fuel for LWRs was developed as a partial return on investment in reprocessing.

The use of MOX-fuel in LWRs, has been experimented since the early start of nuclear power production in the 1960s. Before 1980, some western European countries and the USA have tested several hundred of MOX fuel assemblies in LWRs and test reactors. The substantial use of MOX fuel started middle of the 1980s. MOX-use is today a mature business and continuous improvement is in matching two main aspects, i.e. fuel cycle management and fuel performance as interface with the nuclear steam supply system.

Both agencies have in the past years developed complementary activities in the field of MOX-fuel use. The OECD-NEA addresses the fuel cycle strategies under it's Nuclear Development Committee's activities and Nuclear Science Committee. The IAEA tackles the MOX fuel technical issues under the International Working Group on Water Cooled Reactor Fuel Performance and Technology, covering all of the countries with the nuclear programs. This paper is aimed to bring a comprehensive overview of the current state and future trends of MOX-use as an integral part of the back-end policy in the whole fuel cycle.

2. MOX FUEL UTILISATION

Plutonium has been successfully recycled in the form of MOX fuel in thermal reactors for more than thirty years. Today, more than 30 thermal reactors use MOX-fuel complying with a partial core loading pattern. Table I lists the current status of MOX fuel utilization in thermal reactors world-wide. The commercial application of MOX fuel in LWRs has been started in the mid 1980s when some modification or withdrawal of fast reactor programmes was enforced. The developed technologies of recycling and fuel fabrication were applied for Pu-recycling in LWR-fuel, in the mean time focusing on stabilisation of the separated plutonium inventory.

Table I Status of large scale MOX fuel utilisation in thermal reactors[a], as of yearend 1998.

	Number of Thermal Reactors			
	Operating [l]	Licensed to use MOX FAs	Loaded with MOX FAs	Applied for MOX license
Belgium	7	2	2	
France	57	20	17	8[b]
Germany	19	12	10	4
Japan	52	3	1	1
Switzerland	5	3	3	
Total	130	40	33	13

[a] There are a number of reactors, notably in Europe and India, not included in this Table, which are licensed to use MOX fuel on an experimental basis;

[b] Technically capable reactors planned to be licensed.

Currently, the use of MOX fuel has been established on an industrial scale in a number of countries. In Belgium, France, Germany, Japan and Switzerland, a considerable number of thermal power reactors (PWRs and BWRs) are either licensed (i.e. 40 licensed reactors of which 33 have MOX fuel loaded) or have applied for a license (about 13) to use MOX fuel at levels of up to 30% of the reactor core (see Table I).

Reprocessing capacity today amounts some 5000 t HM/y (whereof 1500 t HM/y for Magnox-fuel) in the OECD Member countries [l], essentially commercial, and some 620 t HM/y in non-OECD countries (Russia and India), especially as non-commercial plants (Chelyabinsk RT-1, Russia; Tarapur PREFRE-1 and Trombay, India). It is notable that China would join the reprocessing community, where India, Japan and Russia may remarkably expand their reprocessing capacities by commissioning new facilities in the coming decade. Meanwhile, the BR205 Magnox plant in the UK, with a capacity of 1.500 t HM/y would be shut-down sometime in the coming decades. Forecasts of world-wide reprocessing capacity after 2015 account for some 6.000 t HM/y. This reprocessing capacity is sufficient according to current projections of requirements.

About 10.500 tHM of spent fuel were discharged from nuclear power reactors and about 3,000 t HM of spent fuel were reprocessed in 1997, which corresponds to about 30 % of the total. About 24 t of plutonium were separated in reprocessing plants and 9 t of plutonium were used mainly as MOX fuel in LWRs. The imbalance between the separation and use of plutonium as MOX fuel had resulted in an accumulated inventory of separated civil plutonium of about 170 t at the end of 1997, increasing to about 180 tonnes at the end of 1998.

Currently, six plants for MOX fuel fabrication are in operation in Belgium, France, Japan, UK and India (see Table II). World-wide MOX fuel fabrication capacity at the end of 1998 amounts about 220 tHM per year. In the UK, a large scale MOX fabrication plant (THORP) has been constructed and is awaiting consent to start operation. In Russia, the first pilot plant (with a capacity of 10 t HM/y) for fabricating MOX fuel is under construction inside the RT-1 plant. A new MOX plant (Complex 300) is planned to commence operation in 2010 [2,3]. There are plans for the construction of a new MOX plant

in Japan and of a demonstration facility in China. The available MOX fabrication capacities world-wide are projected to be over 600 tHM/y in the coming decade owing to deployment of new facilities and expansion of capacities of existing facilities, where it is anticipated that some 25 to 30 t of plutonium will be recycled per year.

Table II. MOX fuel fabrication capacity (tHM/y), as of yearend 1998.

Country	Site	*Plant*	1998	2000	2005	2010
Belgium	Dessel	PO	35	40	40	40
France	Cadarache	CFC	35	40	40	40
	Marcoule	MELOX	120	200"	200"	250"
India	Tarapur	AFFF	5	10	10	10
Japan	Tokai	PFPF	15[b]	15[b]	5"	5"
	Rokkasho-mura	MOX FFF			100	100
Russian Fed.	Chelyabinsk	inside RT- 1			10	10
	Chelyabinsk	Mayak, Complex 300				40
UK	Sellafield	MDF	8	8	8	8
	Sellafield	SMP		120	120	120
Total			218	433	533	623

[a] date not fixed
[b] for ATR Fugen and FBR Monju
[c] for FBR Monju

More efficient use of Pu will be made in FBRs, where multiple recycling is possible, and has been demonstrated. In Russia, it is intended to recycle plutonium in commercial FBRs and there are plans to construct three such reactors.

3. MOX FUEL PERFORMANCE AND TECHNOLOGY

3.1. **MOX Fuel Performance**

Application of plutonium as MOX fuel in LWRs was an alternative option for effective use of plutonium separated by reprocessing in the nuclear fuel cycle, since FBR development and deployment is delayed. MOX use in existing LWRs requires objective evidence ensuring that its performance in pile is not significantly different from that of UO_2 fuel. Therefore, the performance has been examined in a variety of in-pile and out-pile tests in the past decades, focusing on differences with that of UO_2 fuel.

In the weak of a number of comparative tests on reliability of MOX fuel use in LWR, no significant difference has been found, at least at present burnup level. Following are notable characteristics of the MOX fuel on irradiation performance.

- Fission gas release: By integrating currently available data on the fission gas release with a function of burnup [4-8], the releases from the PWR and BWR MOX fuels are not significantly different from those of UO_2 fuels (see Fig. 1). The intensive releases from MOX and UO_2 fuels in PWR obviously resulted by high power irradiation experiments. In the early phase, higher fission gas release from MOX fuel of PWR, due to higher fuel temperatures of MOX than that of UO_2, and less homogeneity of the Pu distribution were observed. Such high release would not occur currently by improvement of MOX fuel fabrication technology. The fission gas release from MOX fuel against the rod power is comparative to that from UO_2 fuel [6].

- Fuel rod growth: Growth of MOX fuel rods is fairly the same as those of UO_2 fuel rods [9].
- Creep behaviour of MOX pellets: Creep rate is deeply related to PCI behaviour of fuel rods. Out-of-pile creep rate of MOX pellets is pretty larger than that of UO_2 pellets which seems to lead to much mitigation of PCI comparing to UO_2 [lo]. On the other hand, it was found by a comprehensive review [11] of creep rates of irradiated fuel pellets that radiation-enhanced creep rates of both oxide pellets were not so significantly different from each other at the temperature of operation.
- Transient behaviour: Transient tests of high burnup MOX fuel rods (up to 50 GWdlt HM) indicated that integrity of the fuel rods was maintained without defects up to power levels of 4 16 - 474 W/cm with a power ramp rate of 100 W/cm.min. This result would imply better transient performance of the MOX fuel rods than the UO_2 rods [12]. The fission gas release from MOX pellets in transient conditions is clearly compatible with those from UO_2 pellets [9, 13].
- Plutonium homogenisation: Coarse MOX agglomerates cause local high burnup in pellets which leads to enhanced fission gas release from the agglomerates due to local high temperature. MOX fabrication technology has been improved to homogenise plutonium distribution by adoption of micronization or co-milling process in the dry route.

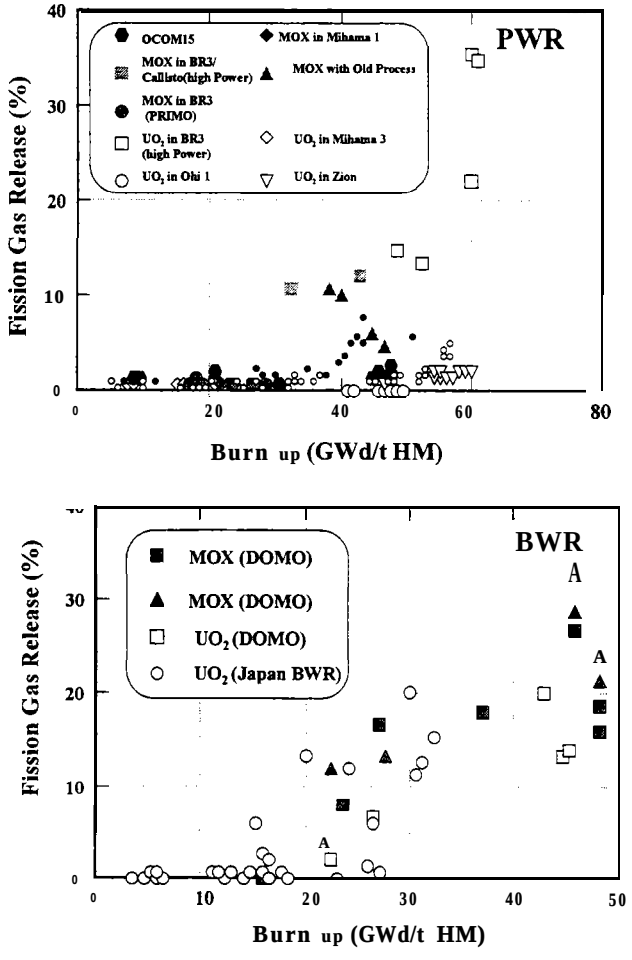

Fig. 1 Fission gas releases from PWR and BWR MOX fuels with a function of burnup [4-8].

Note that the high fission gas releases in PWR are caused by specific conditions such as the high power and old production technology. Except for the specific conditions particularly in PWP, the fission gas releases are mostly the same as those from ordinal UO_2 which are less than 10 % at burnup up to 50 GWd/t. In the case of BWR MOX use, the fission gas releases are about 20-30 % at burnup of 50 GWd/t, which are also comparable with those from UO_2.

MOX performance in pile has been well ensured for the safe use in thermal reactors. In the past decades, a particular concern was placed on the fission gas release from MOX fuel which might influence on fuel rod safety under irradiation. However, it was proved that the release from MOX fuel fabricated by the advanced technology was not significantly different from that from UO_2 as far as the present burnup level is concerned. The other MOX fuel behaviour would be the same as those of UO_2. For increasing reliability on MOX fuel use, further efforts are required to accumulate the data, particularly focusing on safety related behaviour such as RIA conditions. The database of out-of-pile characteristics of MOX fuel supporting better understanding of in-pile behaviour are rather concentrated in the region of plutonium concentration higher than 20 wt % which corresponds to FBR fuel. It would be necessary to accumulate the MOX data in the region of lower plutonium content.

3.2. **MOX-Fuel Benchmarks**

The Working Parties on the Physics of Plutonium Recycling and Innovative Fuel Cycles addressed the question of MOX multi-recycling in LWRs [14-21]. The essential nuclear data evaluation for MOX-fuel have been covered by the Working Party on International Nuclear Data Measurement Activities.

The OECD/NEA Nuclear Science Committee has commissioned a study and a series of benchmarks through its Working Party on Physics of Plutonium Recycling and Innovative Fuel Cycles (WPPR) [16-22]. The benchmarks cover PWR MOX pin cells of a variety of isotopic plutonium vectors, reactivity void effects in PWRs, physics limitations in multiple recycling of plutonium in standard and highly moderated advanced PWRs, and different scenarios of fast plutonium burner reactors. Benchmark studies were undertaken in areas considered to be of particular interest. The first set investigated particular issues related to mixed oxide (MOX) usage in PWRs with plutonium, both of typical and poor isotopic quality. This latter type of plutonium is expected to become available for recycling early in the next century when multiple recycling of plutonium, such as PWR MOX, is expected to be implemented. The benchmarks were designed to question whether present nuclear data and lattice codes are likely to require further development and validation to be able to satisfactorily calculate the core physics performance of such plutonium. A second set of benchmarks examined fast reactor systems to determine the level of agreement of the rates at which plutonium can be burnt and minor actinides can be fissioned, in particular for the purpose of reducing the source of potential radiotoxicity.

For the two benchmarks the agreement between the various solutions was not completely satisfactory indicating a further need for improvement in both methods, e.g. self shielding treatment of ^{242}Pu, and basic data for higher plutonium isotopes and minor actinides. Further experimental validation would also be needed, in particular for integral parameters, e.g. reactivity coefficients, in the case of degraded plutonium isotopic composition.

The analysis of the intermediate steps of five recycle generations in standard and highly moderated PWRs was made [16] with the aim of determining where the nuclear codes and data libraries would start losing their applicability. The observed spread of results are now consistent with the uncertainties in the underlying nuclear data and, therefore, enable multiple recycling scenarios to be correctly assessed, at least in the near term for a first generation recycle. Multirecycling of plutonium in PWRs of current design beyond a second recycling can have intrinsic limitations and the related physics issues have been considered, in particular the plutonium content limitation to avoid positive void effects and the minimisation of minor actinide production during multirecycling.

3.3. Burnup Credit

The motivation for using burnup credit for spent UO_2 fuel has been dictated in many countries by different needs (e.g. introduction of higher enriched fuel in existing storage, reprocessing or

transport systems; increasing storage capacities; and increasing transport cask capacities to reduce the number of shipments needed). The same arguments are valid for spent MOX fuel.

However, if burnup credit is used for the design of a spent fuel management system, it should be noted that spent MOX fuel characteristics are different then those of spent UO_2 fuel.

- For spent UO_2 fuel, provided the fuel does not have any initial burnable absorber content, the reactivity decreases monotonically with increasing average discharge burnup. For a MOX fuel assembly, however, this decrease in reactivity is significantly less;
- Compared to uranium dioxide fuel, employment of MOX fuel leads to a neutron spectrum hardening which affects the rates of fissile depletion and plutonium and fission product buildup, and may affect the distribution of burnup within a fuel assembly;
- The reactivity change of MOX fuel with increasing burnup may be dependent on the initial Pu quality isotopic vector of the MOX fuel of interest. Accordingly, improvements in depletion code validations require chemical assay data to determine concentrations of actinides and fission products of interest as a function of initial Pu quality isotopic vectors and irradiation histories;
- Evaluation of the reactivity effect of MOX PWR axial burnup shapes on burnup credit requires collection of data on such shapes. As it has been the case for uranium dioxide systems, treatment of MOX BWR fuel is more complex than examination of MOX PWR fuel.

Today, a MOX burn-up credit benchmark is in progress covering various combinations of initial MOX fuel composition, burnup, cooling and spent fuel representation [23].

4. ECONOMICS OF MOX FUEL

Competitiveness is a key issue for the deployment of MOX fuel utilisation world-wide even if other factors such as natural resource management and waste minimisation play an important role in the choice of fuel cycle options.

The economics of MOX fuel utilisation is determined mainly by the costs of reprocessing, MOX fuel fabrication and spent fuel disposal, and by the price of fresh uranium. A number of studies on economic aspects of the fuel cycle, including the NEA analysis published in 1994 [14], have pointed out uncertainties on future trends in uranium and fuel cycle service costs and prices. However, the accumulated experience in spent fuel reprocessing and LWR MOX fuel fabrication and use provide some insights on economic aspects of the closed LWR fuel cycle.

With present technical, industrial and economic conditions, MOX fuel is barely competitive. When the 1994 NEA report was prepared, calculations in the reference case showed a difference of around 10 per cent in favour of the direct disposal: 5.46 USmills/kWh versus 6.23 USmills/kWh for the once-through and closed lifetime levelised fuel cycle costs respectively, at 5 per cent discount rate (see Fig. 2). As pointed out above, the uncertainties on cost elements, such as spent fuel disposal cost, are large enough to make the difference insignificant.

Historical trends show that uranium prices mainly depend on supply and demand balance and have not been significantly affected by production costs. In the light of the present uranium market trends, prices should remain rather stable. World-wide, nuclear capacity growth is expected to remain modest in the coming decades; technology improvements, allowing higher burn-up for UO_2 and MOX fuels, will decrease specific uranium consumption (tU/kWh); and significant amounts of ex-military fissile materials will become available on civil markets thereby reducing the demand for fresh uranium. In this context, MOX fuel will have to compete with UO_2 fuel at current uranium prices.

At present MOX fuel fabrication is three to four times more expensive than UO_2 fuel fabrication. This is partly due to the specific safety and radiation protection measures required in MOX fuel fabrication plants for the safe handling of plutonium. However, the main reasons for the high cost of MOX fuel fabrication are the small size of the plants and the relatively early stage of

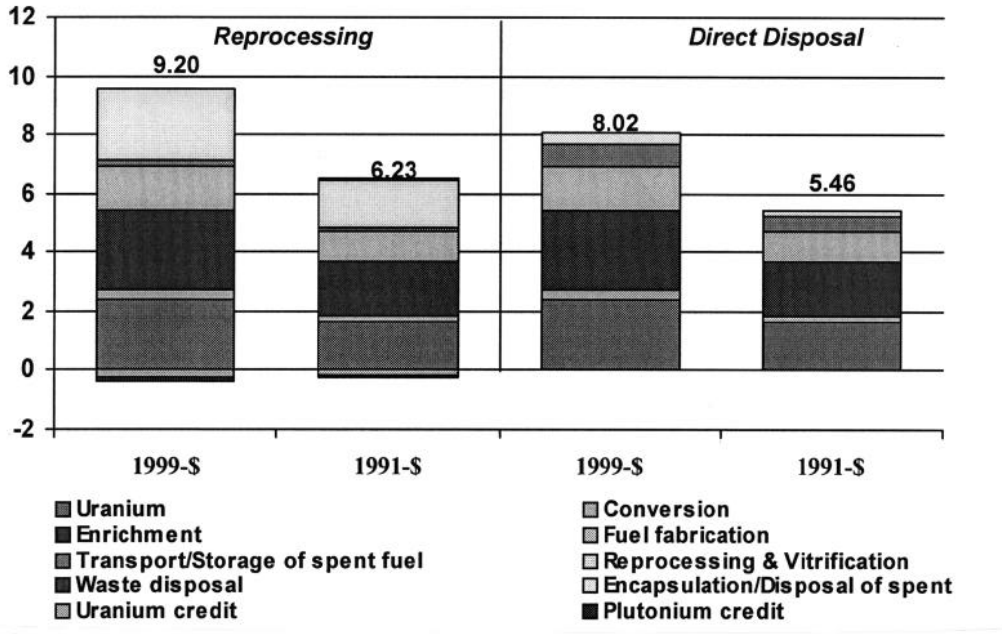

Fig. 2 Results of the OECD/NEA Economics of the Nuclear Fuel Cycle (1994) [14]

Note that 1999-$ shows the monetary value change from the value in 1991.

development of recycling industry. It is expected that feed-back from experience (learning process) and economies of scale will reduce significantly the cost of MOX fuel fabrication as industry matures and outputs of the plants increase.

Another important parameter is the burn-up of MOX fuel. As industrial experience accumulates, it is expected that MOX fuel will be licensed for burn-up as high as those authorised for UO_2 fuel.

The costs of reprocessing services, which have an impact on the overall economics of MOX, are likely to decrease owing to amortisation of the existing plants in the short term and industrial maturity in the longer term. For example, in France, costs are expected to drop by 50 per cent once the La Hague plant investments will be amortised. Also, the industry is considering an integrated approach through offering back-end of the fuel cycle services including reprocessing, MOX fuel fabrication and high level waste conditioning. Such an approach aims at enhanced effectiveness and reduced costs for the entire back-end.

Finally, it should be noted that internalisation of external costs could make MOX more economically attractive. The merits of recycling, well recognised in most industries, are significant in the case of nuclear power since the use of plutonium allows to reduce the amount and activity of radioactive waste and to increase the energy extracted from natural uranium.

5. MOX FUEL LICENSING

Licensing is a key part of MOX fuel use as a back-end option. The licensing process of a MOX fuel fabrication plant and the reactors which use MOX fuel varies from country to country It usually involves licensing reviews of safety analysis reports by regulatory authorities, some countries may include reviews by standing groups of experts, public inquiries, and/or other stakeholders' involvement . The outcome of the licensing effort often depends on the country's back-end fuel cycle policy and its acceptance to MOX-use in reactor.

5.1. Licensing for MOX Fabrication Plants

MOX fuel fabrication plants are in operation in Belgium, France, India, Japan, Russia and UK. Table III shows the general characteristics of the currently-operating and licensed fabrication plants. It shows the start of licensed operation of the plants, the fabrication processes used, the anticipated (licensed) capacity and the fabrication products, and the licensing limits, expressed in terms of the maximum ^{241}Am content, and minimum ^{240}Pu content in the plutonium. The content of ^{241}Am is regulated because it affects the radiation exposure to the worker. Most of the operating fabrication plants are designed and operated to limit the maximum worker exposure to about 10% of the radiation exposure limit (50 mSv/y) stated in the ICRP. The content of ^{240}Pu is regulated because of the concern for criticality safety in unit processes of the fabrication plants. The shapes and sizes of the equipment must be designed and the amount of plutonium processed must be regulated to avoid accidental criticality. Other licensing concerns include the recycling of scraps to minimize the loss of plutonium to wastes and to the release to the environment.

Table III Licensed MOX fabrication plants

Country	Licensed plant	Operating since(year)	Fabrication process	Capacity[a] (tHM/y)	for	Licensing Limits[b], % max ^{241}Am	min ^{240}Pu
Belgium	BN-Dessel	1973	MIMAS	40	LWR	1.7	5
France	CFCa	1962	COCA	10	FBR	--	--
		1989	MIMAS	35	PWR	1.5	17
	MELOX	1995	A-MIMAS	100	PWR	3.0	17
India	AFFF	'90s	conventional	10	BWR	--	--
Japan	PFFF, Tokai	1972	MH/MB [1]	10	ATR	1.0	--
	PFPF, Tokai	1988	MH/MB[c]	5	FBR	3.0	10
Russia	PAKET	'80s	GRANAT	0.3	FBR	--	--
	RIAR	'80s	vibro-pack	1	FBR	--	--
UK	MDF	1993	SBR	8	PWR	3.0	17

a. These are anticipated capacity of the plants

b. Depicted from Ref. "Management of separated plutonium, The technical options," OECD/NEA, 1997

c The Tokai plant uses $(U\text{-}PU)O_2$ feed obtained by micro-wave heating of (U-Pu) nitric solution (MH) from Tokai Reprocessing Plant, followed by mechanical blending (MB).

5.2. Licensing Examples of MOX Fabrication Plants

In Germany, a MOX fabrication plant was operated by Siemens in Hanau from 1972 to 199 1. It was shutdown because of a contamination incident, and is now decommissioned. A larger plant was built and ready to start, but because of the change of local politics, it was never licensed and is now abandoned [15].

In France, the Melox Fabrication Plant was successfully licensed in the early '90s. License application was sent to the French Ministry of Environment and Ministry of Industry in May 1990. The application comprised of the preliminary safety analysis report and the public inquires. Reviews of the safety analysis report and recommendations by a standing group of experts, together with the results of the public inquires formed the basis for a Plant Authorization Decree. After examination of safety documents by the Safety Authority, an operating license was granted in August 1990. The Melox Plant was started up in 1995 after the granting of the Liquid and Gaseous Effluent Release License and the approval of a final safety analysis report [24].

In UK, the finished SMP Plant was ready to start since Spring of 1998. However, the plant is still awaiting its operating license from the government:s Department of the Environment, Transport and the Regions. The delay in the operation, even for a uranium-commission, has resulted in economic penalty to the operator (BNFL) [25].

In the US, pilot MOX fabrication facilities were operated in the '70s and MOX fuel was recycled in several LWRs. When commercial reprocessing was terminated in the late '70s because of proliferation concerns, these pilot facilities were also shutdown and decommissioned. Currently, the US is considering a MOX option to disposition its ex-weapons plutonium. The USDOE signed a contract with a consortium led by Cogema to design and build a MOX fabrication plant [26]. License application would have to be submitted to the US Nuclear Regulatory Commission (NRC). The last attempt at licensing a MOX fabrication facility in the US was in 1974. That licensing process was stopped because of concerns on nuclear proliferation. The current effort is intended to reduce the separated weapons-grade plutonium inventory, the proliferation issue should not prelude a licensing basis for a MOX fabrication facility.

5.3. **Licensing for Reactors using MOX Fuels**

MOX fuels are currently used as replacement fuels in LWRs. They are partially loaded in the reactor core (see Table V for % allowed in countries). The MOX fuel assemblies (FA) design is basically the same as that of the UO_2 FAs to preserve the thermal-mechanical integrity of the reactor. The plutonium contents (total or fissile) and the burnup for the MOX FAs are limited such that when they are loaded into the core, they would not compromise the safety margins established as the licensing bases for the reactor.

Licensing requirements and procedures vary from country to country. In general, the license applicant should conduct safety analyses to demonstrate to the country's regulatory body that the fuel related safety criteria associated with MOX-use meet the licensing bases established for that reactor. The fuel related safety criteria relevant to MOX-use, as listed in Reference [27] for PWR fuel are shown in Table IV.

Table IV Fuel safety related criteria

Fuel Safety Related Criteria	**Applicable Conditions**	
• reactivity coefficient	B, C	A: normal operation
• shutdown margin	A, B, C	B: anticipated transients
• strain level	A, B	C : accidents
• oxidation	A	
• hydride concentration	A	
• internal gas pressure	A	
• pellet cladding interaction	A, B	
• fuel fragmentation	C	
• fuel failure	C	
• cladding embrittlement	C	
• coolant activity	C	
• gap activity	C	
• source term	C	

The safety analyses should address the impacts on these relevant fuel safety related criteria due to MOX-use in the reactor, especially related to concerns on MOX fuel behaviours, such as:

- fission gas release,
- helium gas accumulation,

- swelling,
- fuel centerline temperature

In addition, a spectrum of operating transients (e.g., control rod ejection or drop) and design basis accidents (e.g., loss of coolant accidents (LOCAs), reactivity-insertion accident (RIA), etc) should also be included in the safety analysis to demonstrate that the differences in neutronic and thermal properties of MOX FAs would not create the possibility of new types of transients and accidents, and the probability or consequence of any previously analyzed accidents would remain within the safety envelope on which the operating license of the reactor was based.

5.4. Experiences with MOX-Use in Thermal Reactors

Under the licensed conditions, the accumulated experience in MOX utilization in European thermal reactors demonstrates that the MOX fuel performance is excellent and comparable to that of UO_2 fuel. Table V shows the experience with MOX-use in these reactors, it includes the licensing limits, expressed in terms of maximum MOX loading in the core, and the maximum concentration of plutonium in the MOX fuel. Japan is included since it is expected to use MOX in year 2000.

Table V Experiences with MOX-Use in LWRs

Country	Operating reactors	Reactor licensed to use MOX	"Moxified" LWRs	First MOX loading date"	Licensing limits, % max in core	max Pu, conc
Belgium	7	2	2	1995	33	7
France	57	20	17	1987	30	5.3
Germany	21	11	10	1972	50	--
Japan	52	3	0	2000[b]	33	13
Switzerland	5	4	3	1984	40	--

a From Booklet: "Cogema: Reprocess to recycle," Feb. 1999.
b Anticipated date.

To reduce the uncertainties associated with higher burnup or larger core loading of MOX fuel in the future, more experiments and R&D efforts would be needed to demonstrate that the reactivity insertion accident (RIA) with high MOX burnup fuel would not be a limiting condition, or the beyond-design-basis LOCA involving MOX fuel would not cause additional impacts on the environment.

6. MOX FUEL TRENDS

Trends in MOX fuel use will vary from country to country according to national policies on spent fuel reprocessing and MOX fuel fabrication. In addition, it is expected that some limited quantities of ex-military plutonium become available on civil markets. Therefore, there are large uncertainties on the future evolution of plutonium use and inventories. In order to illustrate alternative paths and investigate related issues, a computer code developed by the IAEA was used to calculate spent fuel arisings and plutonium utilisation in different nuclear growth variants and fuel cycle strategies.

MOX fuel trends were forecast by the computer code, VISTA [28] newly developed by the IAEA. The VISTA was designed for calculating spent fuel arisings, actinide generation, plutonium separation and its utilisation, as well as other information related to the back-end. The VISTA is a scenario based tool which can be used to estimate trends for the closed cycle where recycle of separated fuel material is taken into account. In the course of its development, in the last few years, the VISTA was used for investigating the new realities [31] the nuclear power and climate change

[30] and for a number of front-end estimations such as natural uranium, conversion and enrichment service requirements [29].

In order to analyze MOX fuel trends, the model estimates, for eight reactor types, average MOX fuel requirements corresponding to electricity generation capacities and fuel cycle scenarios. Closing the fuel cycle is made by calculating two sets of fuel loads and unloads. One is for reactors using only uranium fuel and the other is for reactors using uranium and MOX fuels. A special scenario file defines, among others, the fuel cycle scenario to be investigated with some fractions of total amount of discharged fuel (spent fuel arisings) which are reprocessed. The contents of plutonium (and other actinides) in the spent fuels are calculated using IAEA model CAIN [32]. The final step involves separating plutonium and fabricating MOX fuel which is then loaded as a part of the core of some LWRs. This method enables the simulation of a closed nuclear fuel cycle.

6.1. **Main Assumptions**

The VISTA calculation is based on a given scenario portraying the future of the nuclear power and its fuel cycle. In line with objectives of this paper, two nuclear capacity variants, high growth (HV) and low growth (LV) were selected, coupled with three reprocessing-recycling strategies as described below. According to IAEA database, 437 nuclear power reactor units were operated in 32 countries, with a total electric generating capacity of 35 1 GWe worldwide as of December 1997 [40]. Future trends are based on IAEA projections as shown in Table VI [33]. The table lists the world nuclear power capacity together with capacities of LWR group (PWR, BWR and WWER) and non-LWR group (PHWR, RBMK, AGR and GCR).

TABLE VI World Nuclear Power Capacity Variants (GWe)

	2000	2010	2020
High variant - HV	352	432	567
LWR	309	383	519
Non-LWR	43	49	48
Low variant - **LV**	**348**	**374**	**322**
LWR	**307**	**336**	**295**
Non-LWR	41	38	27

The fuel cycle scenarios, shown in Table VII, are characterised by difference in the reprocessing ratio and number of cycles to be applied. The Sl scenario was followed to earlier investigations [28 - 3 1]. It assumes that half of the spent fuel arisings is offered for reprocessing (50% reprocessing ratio) and that plutonium extracted from UO_2 spent fuel can be recycled twice in LWRs. This scenario is used as a maximal-reference option. The scenarios S2 and S3 assumes one cycle only, without the reprocessing of spent MOX fuel, and with end-of-period reprocessing ratio of 50% and 35% respectively. These scenarios are considered to be the more probable for future reprocessing and recycling of spent fuels.

TABLE VII. Fuel Cycle Strategies

	Reprocessing Ratio (%)	Number of Cycles
Maximal-reference strategy - **S1**	**50**	**2**
High realistic strategy - S2	50	1
Low realistic strategy - S3	35	1

6.2. Spent Fuel Arisings and Storage

Annual spent fuel arisings estimated by the VISTA indicated a peak of about 11300 t HM/y in 1990 and reduced slightly to about 10200 t HM/y in 1998. This trend is reflected by the higher burnups causing the reduction of the amount of spent fuel to be discharged [34, 351. However, change of spent fuel arisings for future are compensated by diametrical factors of extended burnup and nuclear power capacity growth. Estimation of annual fuel arisings in the years up to 2020, indicates either reducing or increasing which depend on the world nuclear power capacity variants. The scenario with high variant (HV) in Table VI predicts the annual spent fuel arisings to increase steadily up to 12000 t HM/y, and LV scenario, less than 8000 t HM/y in 2020.

A comparison between spent fuel storage capacities and total amounts of spent fuel stored designated as requirements for storage, is shown in Fig. 3. The storage requirements which is the amounts deducted by the amount reprocessed, were calculated with the scenarios mentioned above. The results predict that storage capacities will have about 100000 t HM surplus over the total storage requirements until 2015. It is noticed that the spent fuel storage capacity is almost comparable with accumulated spent fuel arisings.

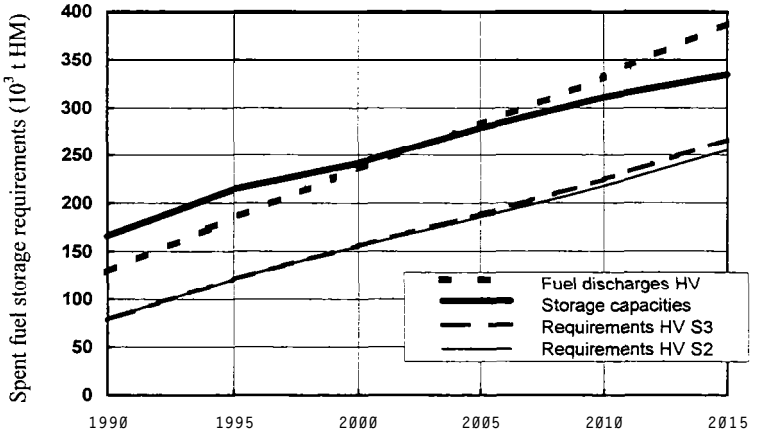

Fig. 3 Comparison between spent fuel storage requirements and capacities worldwide (10^3 t HM)

Significant in this results is that continous efforts to expand the storage capacities and to keep a current level of spent fuel volume for reprocessing are required to address potential shortage issue on the storage capacities for future. The spent fuel storage capacity projected worldwide which was estimated from information collected by the IAEA, could be sufficiently large for growing amounts of spent fuel to be stored, if reprocessing continues with the present level in the time period until 20 15. However, it should be reminded that the situation of storage capacity can be to a great extent different individually from country to country, for instance, those in Western Europe, have enough capacity, while others do not.

6.3. Reprocessing

Capacities and requirements of reprocessing worldwide up to the year of 2020 was predicted using information on the projected reprocessing capacities of which information was collected by IAEA and amounts of spent fuel required for reprocessing which were calculated by the VISTA code. Fig. 4 shows a comparison of both factors. The total projected reprocessing capacity worldwide increases over the period 1998-2010 due to the deployment of new plants in Japan and India, but after 2010, a further change is anticipated due to likely closure of the Sellafield (B205) plant and commissioning of a second plant in China.

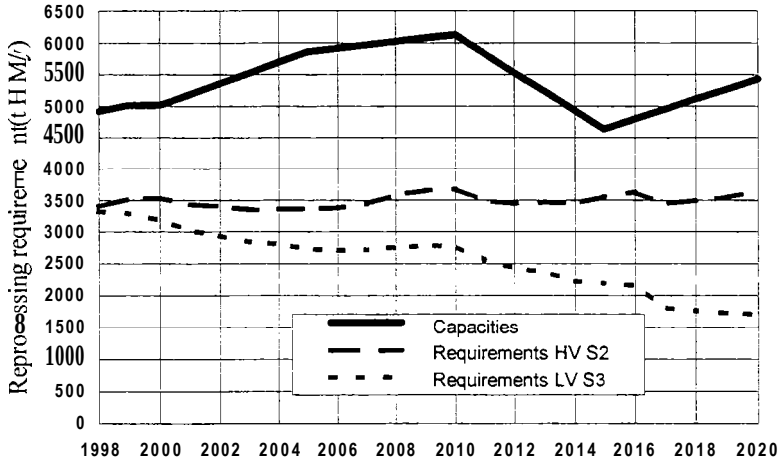

Fig. 4 Comparison between worldwide reprocessing capacities and anticipated reprocessing requirements calculated by the VISTA code (t HM/y)

The worldwide reprocessing requirements calculated by the VISTA code are substantially a total of amounts of spent fuels from the LWR and the non-LWR to be reprocessed. In the scenarios mentioned former, the LWR nuclear power capacity is likely to increase or almost steady during the time frame of this work. Since reprocessing requirements change following the trend of annual spent fuel discharges, the VISTA calculation predicts that reprocessing requirements for LWR fuels would come to around 2000 t HM in 2000, followed by a small reduction to around 1,700 t HM in 2005 and again increase to 2000 t HM in 2015. This calculation was made including the reprocessing of non-LWR fuels. The VISTA calculation projects a constant reduction in reprocessing requirements of these fuels, especially due to the end of the GCR Magnox program in the United Kingdom sometime within the next 10 to 15 years. It should be noted here that reprocessing requirements are very sensitive to political environment in each country and may change dramatically in the coming years.

6.4. Plutonium Utilization

Plutonium is generated (and is partly burned) during the operation of uranium-fuelled nuclear reactors and forms part of their spent fuels. The IAEA estimates that about 76 tonnes of plutonium were contained in discharged spent fuels worldwide in 1998. It is foreseen that the annual plutonium production, as presumed from spent fuel arisings in Fig. 3, will remain more or less the same until 20 15. The cumulative amount of plutonium in spent fuels from power reactors worldwide is predicted to reach 2,000 tonnes in 2015. Some of the plutonium contained in reactor spent fuel has been separated and a part of it (approximately one third) has, up to now, been used to manufacture MOX fuel for LWRs and experimental and prototype FBRs, but the major part of the separated plutonium is currently stored, mainly at the British, French and Russian reprocessing sites.

For taking an outlook of MOX fuel trends, the VISTA calculation was made to estimate fabrication amounts of MOX fuel to be used in LWRs. In 199.5, about 8 tonnes of plutonium as MOX fuel were used in LWRs and fast reactor development programs. The VISTA calculation predicted that some 25 tonnes of plutonium would be consumed annually to fabricate MOX fuel until 2010, if assuming that MOX fuel will have a share of 30% in the core of LWRs worldwide [36].

The calculation is based on the assumption that the separated plutonium is used to fabricate MOX fuel which is loaded only in LWRs. The procedure used to calculate these estimation involves using of the separated plutonium for the required MOX fabrication.

A comparison of MOX fuel production capacities and predicted MOX fuel requirements to 20 10 is given in Fig. 5.

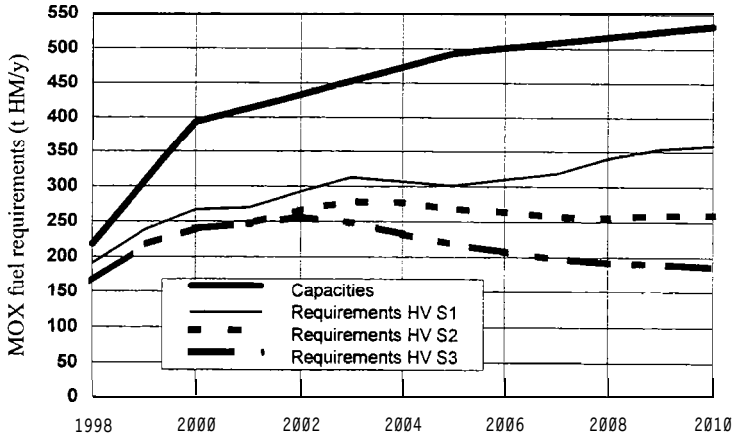

Fig. 5 Comparison between worldwide MOX fuel production capacities and anticipated annual MOX fuel requirements calculated by the VISTA code (t HM/y)

MOX fuel requirements in the last year were close to the full production capacities. However, the relation between requirements and capacities is being improved quickly by commissioning new fabrication facilities and expanding capacities of existing facilities (see TABLE II). The results obtained from the VISTA calculation in any scenarios indicate a trend that MOX fuel requirements increase in the years to come, followed by slight reduction. This trend is caused by reduction of annual spent fuel arisings in these years, which reflect smaller amounts of spent fuels to be sent to reprocessing plants. This trend may change depending on the scenarios which adopt either multi-cycle or a single cycle of plutonium. The trend may also change if the share of MOX fuel in the core will increase over the 30% value used in this investigation. As can be seen in Fig. 3, in all scenarios adopted, the situation of the excessive capacity over requirement would continue up to 20 10.

Projection of world inventory variation of separated civil plutonium calculated by the VISTA code is presented in Fig. 6. The calculation estimates about 180 tonnes of inventory at the end of 1998. This corresponds to earlier projections [36] and coincides precisely with the real world inventory in this year.

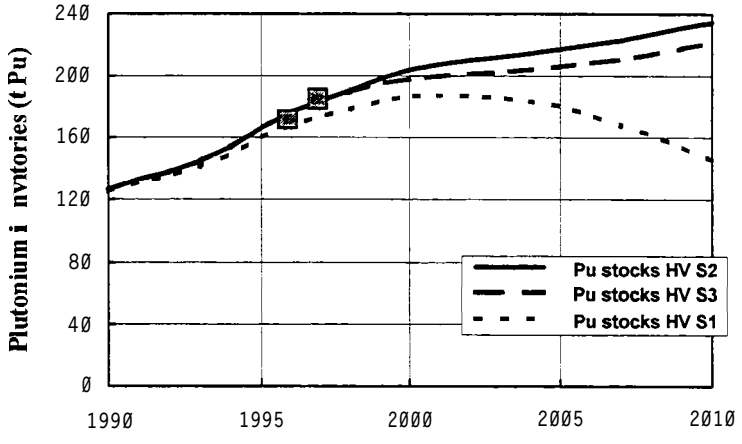

Fig. 6 Estimation of separated civil plutonium inventories worldwide (t Pu).
Note: Open dots indicate the real world civil inventory

On the basis of the scenarios adopting the single recycle strategies (S2 with HV and S3 with HV), the VISTA calculation predicts that the stockpiles of separated civil plutonium will continuously rise in the next decade. This trend may change if multiple recycling of plutonium will take place, as is demonstrated under the assumption using the S1 with HV, or in the case of using plutonium from stocks to fabricate MOX fuel. Eventually, the results of calculations demonstrate a trend that the separated civil plutonium inventories reduces under the multiple recycling scenario, Sl. The calculation with this scenario estimates that the fabrication of MOX fuel to feed 30% of LWR cores requires larger quantities of plutonium than those by reprocessing of spent fuels. This additional quantities are compensated by draw-down of the plutonium stockpiles and consequently such operations lead directly to reduction in separated civil plutonium inventories.

Forecasts of separated plutonium inventory for future have significantly associated with uncertainties reflected by policies of individual countries on MOX fuel fabrication and spent fuel reprocessing. Situation of plutonium stocks and fuel cycle plans are different from country to country. In some countries MOX programmes are being actively implemented, but in others recycling of separated plutonium is not expected to take place in the near future. In addition, limited quantities of military stocks of ex-weapon plutonium may soon be put into the civilian sector.

7. CHALLENGES FOR THE FUTURE

In France, the MOX operating license already allow a burn-up of 39 GWd/t for a third core fuel reload cycle and in the near future, MOX fuel performance will be brought to the levels currently achieved with UO_2 fuels which have an average burn-up of 45 GWd/t. Developments currently pursued focus smoothing fuel cycle management by increase of burn-up of these MOX-fuels, the possibility of load-following with MOX-fuelled reactors (already demonstrated and licensed in France) and the possibility of increased MOX contents (100% MOX-cores) resulting in a net reduction of the Pu-inventory (-15 kg/tHM in a 900 MWe 100% MOX core).

A short-term economical issue is that the MOX-use also reflects on the final back-end waste management. The increased accumulation of minor actinides (especially ^{241}Am and ^{244}Cm) in the fuel indicates that a proper waste management policy should be set-out. The multi-recycling of MOX-fuel is a possible option which is predicted to reduce the inventory of separated plutonium shown in Fig. 6. but at the same time, to increase a Ii-action of minor actinides in the discharged fuel [15, 22]. Improvements in this balance can be made by the use of overmoderated MOX-cores such that the ratio of net minor actinide production over plutonium reduction becomes minimal and remains small after successive recycling. These improvements demand however changes to the core and as such could only be envisaged as possibilities in advanced reactor concepts in the next decades. The overmoderated MOX-cores also seems to pose more difficulties for present nuclear data libraries and codes, as evidenced by the large number of discrepant results seen in OECD/NEA benchmarks.

Open questions to be addressed in the longer term are LWR@R complimentary programmes. Several scenarios could be considered, i.e.: multiple plutonium recycling via MOX-fuel in standard LWRs until maximum depletion of the fissile material is reached and where minor actinides are treated as wastes, providing economic advantages; single recycle through existing LWRs, followed by reprocessing and burning in a fast reactor which also can incinerate minor actinides; and thirdly taking advantage of existing facilities by optimising the number of LWR plutonium recycles and as such reducing the number of burners required. The second scenario seems highly probable for the next decade based on the secured back-end fuel cycle flexibility.

Disposition of the ex-weapons plutonium is a matter of concern in the MOX fuel sector. The International Experts Meeting on Safe and Effective Management of Weapons Fissionable Materials designated as No-longer Required for Defence Purposes, held in Paris, October 1996 after the 1996 Moscow Nuclear Safety and Security Summit, submitted recommendations that MOX fuel for burning the ex-weapons plutonium in existing reactors is a top priority. In response to the recommendations, several processes for conversion, MOX fabrication and MOX burning are proposed under international co-operations. The USA is adopting the dual track approach for the disposition that are MOX use and immobilisation [37]. The USA recently announced to use the solvent extraction processes for conversion for feed preparation [38]. It is envisaged that 91,000 MWe LWRs with l/3 MOX core or 3 reactors with full MOX-core are required to complete the mission for destroying 50 t Pu [38]. It is planed that the irradiation starts by the year 2007 and complete by 2022 [38]. MOX fuel fabrication capacity is estimated to be sufficient about 75 metric tonnes of MOX per year [37]. In addition, the USA is seeking an alternative for CANDU reactors. Russia pursues the trilateral co-operation with France and Germany. The joint French-Russian studies under the AIDA-MOX Program [39] concluded to adopt a process for MOX fuel fabrication as a initial measure that the COCA and MIMUS processes are used after plutonium conversion with HNO_3-HF into PuO_2 through Pu oxalate. This process would be modified latter. France, Germany and Russia jointly proposed the construction of a pilot scale MOX plant in Russia with capacity of 1.3 t Pu (approximately 30 metric tonnes of MOX per year), providing enough MOX fuel for a hybrid core BN-600 fast reactor and 4 WWER-1000 reactors at the Balakovo site [39]. Japan is to co-operate with Russia for upgrade of the vibro-packed fuel fabrication facility and the conversion of BN-600 to the hybrid core.

CONCLUSIONS

The IAEA and OECD-NEA jointly reviewed versatility of MOX fuel use in LWRs at the time when it is reaching the maturity level in countries with a plutonium recycling policy. The review brings following conclusions.

- Currently, more than 30 thermal reactors world-wide are operated with MOX fuel loading without major modification of the reactor core design and the number of the reactors to use MOX fuel are certainly increasing. Six MOX fuel fabrication plants with a total fabrication capacity of 218 t HM/y are operated currently and it is anticipated that the capacity will expand sharply to approximately 600 t HM/y in the next decade.
- MOX fuel has exhibited excellent in-pile performance which is comparable with that of UO_2. The fission gas release from MOX fuel fabricated by the previous technology, was barely higher than that of UO_2, particularly, in PWR. Today, the fission gas releases from both oxide fuels are not significantly different, owing to improvement of MOX fuel fabrication technology. The other behaviours of MOX fuel are the same as those of UO_2. It is advisable to expand the database of MOX with plutonium content lower than 20 wt %.
- MOX fuel benchmarks were studied covering PWR MOX pin cell of a variety of isotopic plutonium vectors, reactivity void effect in PWRs, physics limitations in multiple recycling of plutonium in standard and highly moderated advanced PWRs. A reference was made in fast plutonium burner reactors. It is concluded that further improvement of reactivity benchmark method is required considering, self shielding treatment of ^{242}Pu and basic data for higher plutonium isotopes and minor actinides. Multi-recycling of plutonium in PWRs of current design over second cycling can have intrinsic limitation.
- For the burnup credit application of spent MOX fuel, difference in reactivity, distribution of burnup along the assembly axial length between spent UO_2 and MOX, plutonium quality in MOX fuel, collection of reactivity effect on axial burnup shape are of importance to be considered.
- The OECD-NEA study showed that the fuel cycle economics with direct disposal and reprocessing was 10 % in favour of the direct disposal option and pointed uncertainties that

render the difference insignificant. Moreover, recent changes of the fuel cycle related factors such as uranium price, matured experience and economic scale of MOX fabrication, amortisation of the existing plats may reduce overall cost of the fuel cycle with reprocessing.

- For the licensing of the MOX fabrication plants, plutonium quality is commonly regulated world-wide, although a variety of items for the plant licensing is assessed which is different from country to country. ^{241}Am content in plutonium to be dealt with in plants, is limited from 1.7 to 3.0 % world-wide from the viewpoint of radiation protection. ^{240}Pu in plutonium is limited to be from 5 to 17 % world-wide for critical safety. For the licensing a number of safety criteria is recommended by the OECD-NEA. Maximum MOX fuel loading in the LWR is licensed from 33 to 50 % in the core world-wide with maximum plutonium content in MOX fuel from 5.3 to 13 wt %.
- It is anticipated that world-wide capacity of spent fuel storage would be large enough for discharged volume in the next decade, if the current reprocessing option is continued. In spite of this view, however, the situation of storage would not be optimistic in some countries due to difference in policy of nuclear fuel cycle. Capacity of reprocessing is also enough large for reprocessing requirement in the coming decades. Capacity of MOX fuel fabrication is rather close to requirement up to date, but the situation between capacity and requirement would be mitigated due to commissioning of new plants or expansion of the existing plant capacity. Inventory of separated civil plutonium would increase, if the single recycling of plutonium is continuously applied. Employment of the two recycling option would reduce it.
- As challenges in MOX fuel use in the future, extension of MOX fuel burnup, full-core loading of MOX fuel, overmodelated MOX-core to minimise minor actinide generation, re-assessment of LWR/FR programmes and disposition of ex-weapons plutonium as MOX are reviewed.

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OVERVIEW OF THE CURRENT STATUS OF AND PROSPECTS FOR Pu MANAGEMENT AND MOX UTILIZATION

(Session I)

Invited Paper

MOX FUEL FABRICATION AND UTILISATION IN LWRs WORLDWIDE

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Abstract

Early in the development of the nuclear programme, a large part of the countries using nuclear energy has studied the reprocessing and recycling option in order to develop a safe conditioning of fission products and to recycle fissile materials in reactors. In the sixties, the feasibility of recycling plutonium in LWRs has been successfully demonstrated by several experimentations of MOX rod irradiations in different countries.

Based on the background of the MOX behaviour collected during the seventies and on the results of the important MOX experimentation program implemented during this period, a large part of the European utilities decided at the beginning of the eighties to use MOX fuel in LWRs on an industrial scale. The main goals of the utilities were to use as a fuel an available fissile material and to control the stockpile of separated plutonium.

Today, the understanding of the behaviour of plutonium fuel has grown significantly since the launch of the first R&D programmes on LWR and FR MOX fuels. Plutonium oxide physical and neutron behaviour is well known, its modelling is now available as well as experimentally validated. Up to now, more than 750 tHM MOX fuel (more than 2000 FAs) have been loaded in 29 PWRs and in 2 BWRs in Europe, corresponding to the recycling of about 35 t of plutonium.

Reprocessing/recycling technology has reached maturity in the main nuclear industry countries. Spent fuel reprocessing and recycling of the separated fissile materials remains the main option for the back-end cycle. Today, the operation of MOX-recycling LWRs is considered satisfactory. Experience feedback shows that, in global terms, MOX cores behaviour is equivalent to that of U02 cores in terms of operation and safety.

1. THE ORIGINS OF PLUTONIUM RECYCLING IN LWRS

Early in the development of the nuclear programme, a large part of the countries using nuclear energy has studied the reprocessing and recycling option in order to develop a safe conditioning of fission products and to recycle fissile materials in reactors.

In the sixties, the feasibility of recycling plutonium in LWRs has been successfully demonstrated by several experimentations of MOX rod irradiations in different countries like :

- ⇨ in 1963, in BR3 reactor (PWR), at Mol (Belgium),
- ⇨ in 1964, in Saxton reactor (PWR) in USA,
- ⇨ in 1966, in VAK reactor (BWR) at Kahl (Germany),
- ⇨ in 1968, in Garigliano reactor (BWR) in Italy.

From the beginning of the sixties, an important development programme has also been implemented regarding MOX fuel for the FBRs in different European countries (France, Germany, UK, Belgium) but also in the USA and in Japan.

During the seventies, MOX recycling was implemented on a larger scale in several LWRs by the loading of batches of MOX FAs. The main experience of MOX recycling during this period has been obtained in Europe, in industrial scale nuclear power plants :

- ⇨ inBWR
  - VAK (Germany),
  - Gundremmingen A (Germany),
- ⇨ inPWR
  - Obrigheim (Germany)?
  - Chooz A (France).

At the beginning of the eighties, the development of FRs was delayed, and later stopped in France, and in Germany. Important stocks of plutonium, separated in the seventies for the FRs were available for the LWRs.

Based on the background of the MOX behaviour collected during the seventies and on the results of the important MOX experimentation program implemented during this period, a large part of the European utilities decided to use MOX fuel in LWRs on an industrial scale. The main goals of the utilities were to use as a fuel an available fissile material and to control the stockpile of separated plutonium.

Experience feedback of the seventies, based on reactor irradiations and R&D programmes in hot cells on the initial MOX design leaded the fuel manufacturers to improve the fuel design particularly to enhance the solubility of the MOX fuel for the reprocessing. The fuel manufacturing procedures had to be changed accordingly.

Today, the understanding of the behaviour of plutonium fuel has grown significantly since the launch of the first R&D programmes on LWR and FR MOX fuels. Plutonium oxide physical and neutron behaviour is well known, its modelling is now available as well as experimentally validated.

2. MOX FUEL FABRICATION

2.1. Fabrication Process

Today, only two processes of LWRs MOX fuel fabrication are used world-wide : the MIMAS process (Micronized MASter blend) developed by Belgonucléaire and the SBR process (Short Binderless Route) introduced by BNFL. These current processes satisfy the requirements of the fuel designers and the utilities regarding pellet specifications, fuel quality and fuel dissolution properties.

The MIMAS powder is obtained through two successive blending steps : the primary blend at high plutonium content obtained by ball milling so-called micronization step and the secondary blend

to reach the required plutonium content. The main advantages of this process is the reduced powder volume implemented during the micronization step, and the flexibility due to the possible intermediate storage of the master blend which can be used for isotopic homogenisation.

The SBR powder is obtained by only one blending step to prepare a granular feed suitable for pressing. The main advantages of this process is the homogeneity of the pellet and the lack of large Pu-rich areas.

The Siemens process used during the eighties was mainly the OCOM process (Optimised CO-Milling), quite similar to the MTMAS process with two steps : a master blending obtained by co-milling and a secondary blending to adjust the plutonium content.

2.2. Fabrication Plants (table 1)

The MIMAS process is implemented in 3 MOX fabrication plants : the PO plant (35tHlvVy) at Dessel (Belgium) operated by BN, the CFCa plant (30tHM/y) at Cadarache (France) operated by Cogema and the MELOX plant (lOOtHM/y) at Marcoule (France) also operated by Cogema.

The SBR process is used by BNFL in 2 MOX fabrication plants at Sellafield (UK) : the MDF plant (StHM/y for demonstration purpose) in operation and the SMP plant (120tHIWy) to be started in 1999.

Germany had MOX fabrication capabilities since the early seventies. The Siemens MOX fabrication plant at Hanau had a capacity of 30 tHIWyear (so called “old plant”). After a contamination event the plant was stopped in 1991 and Siemens decided in 1994 to close it because of the blockage of the local government. At that time the “new plant” (120 tHM/y) was about 95 % completed. For the same political reasons this new plant never went into operation and in 1995 Siemens decided to close down the total Hanau fabrication plant.

In Japan MOX fuels for core physics and irradiation testing have been manufactured in PNC plant since 1967. PNC MOX manufacturing plant (10 tMOX/y) produced nearly 135 ton of MOX fuel for thermal reactor utilisation, equivalent to about 1.6tHM of plutonium by 1998. Most of MOX fuel was for advanced thermal reactor Fugen. Micro-wave heating technique was developed by PNC to obtain a homogeneous plutonium-uranium powder directly from a reprocessed product of Tokai Reprocessing Plant. In the near future, when Rolckasho reprocessing facilities will go in operation, a commercial MOX fuel fabrication plant will be built in Japan with a capacity of about 100tHWy.

3. THE CURRENT SITUATION OF MOX UTILISATION

Today, 40 LWRs are licensed for MOX recycling world-wide, in 5 countries (20 in France, 12 in Germany, 4 in Switzerland, 2 in Belgium and 2 in Japan). In these countries, the main option for the back-end of the fuel cycle is currently reprocessing and plutonium recycling (table 3).
By the beginning of 1999, four European countries burn MOX fuel in LWRs (Germany, France, Belgium and Switzerland), and before the end of 1999, a fifth country : Japan will start using MOX in two LWRs (1 BWR and 1 PWR).

The main part of the NPP using MOX are 29 PWRs (17 in France, 7 in Germany, 2 in Belgium and 3 in Switzerland), but 2 BWRs use also MOX (in Germany).

3.1. Reactor Adaptation

The larger amount of plutonium in the core shifts the neutron spectrum towards higher energy levels, thereby reducing the efficiency of the reactivity control systems. The reduced neutron absorber worth needed in some cases to improve the control rod pattern, to increase boron concentration of the

boron make-up storage tank and to increase boron concentration of the refuelling water storage tank. In some specific cases, use of enriched boron is needed.

Taking into account these neutronic considerations, the number of MOX assemblies is limited on each reactor (MOX recycling rate), the limit depending upon the reactor initial design and its capability of evolution.

In Belgium the low recycling rate of 20% used by the utility is compatible with the reactor operation without any modification of the reactivity control systems.

In France, some light modifications of the reactor (4 RCCA added, increased boron concentration in the refuelling storage tank) have enabled to increase the recycling rate to 30%.
In Germany and in Switzerland, the recycling rate depends upon the type of reactor and is limited by each plant licence between 10% to 50% and yield normally no problems complying with the allowed limits for core-wide parameters as reactivity coefficients or shutdown margins without any changes of the plants. For high Pu-content, corresponding to high uranium enrichment, it could be necessary for PWRs to switch over to enriched B 10 to yield enough margins for a flexible operation.

3.2. Fuel Design and Core Management

MOX rods, together with all components of the MOX assembly, are designed to meet the same mechanical and thermohydraulic specifications as those set for uranium assemblies. The MOX skeleton is generally identical to the skeleton of a typical uranium assembly.
A MOX assembly contains only mixed oxide rods. The plutonium oxide is mixed with natural or depleted uranium oxide (tails from 0.15% to 0.3% U235) to maximise the quantity of plutonium per assembly.

To achieve better balanced rod power distribution in the assemblies, several plutonium contents (three for PWRs) are used in the MOX assembly (zoning). The low plutonium content zone is located at the periphery of the MOX assembly in order to compensate for local power peak in the interface with the uranium assemblies induced by the large increase of the fission and absoption cross-sections of the MOX compared to the U02. In Germany, a specific MOX assembly design uses also four water rods near the assembly centre to flatten the power distribution. Due to the relatively flat reactivity shape versus burn-up there is no urgent need for Gd rods in the PWR MOX FAs.

The BWR MOX fuel assemblies are even more complex with up to 6 MOX fuel rod types to compensate the spectrum changes between the fuel channels. As for all BWR fuel assemblies also the MOX FAs usually have some Gd poisoned rods. These Gd rods don’t have any plutonium in the uranium matrix.

In order to meet the energy equivalence with enriched uranium fuel, the average plutonium content is adjusted using an equivalence formula that takes account of the isotopic composition of the plutonium. As the main part of the reprocessed fuel originates from the PWR reactors, the total plutonium contains between 60% and 70% of fissile isotopes of plutonium : 239 and 241 (table 2).

Regarding core management, the goal of each utility is to obtain the equivalence between MOX and U02 in terms of fuel performance and reactor operation.
In France, the MOX FAs are managed in annual cycles but with 3 cycles for MOX (equivalent to U02 3.25%) and 4 cycles for U02 3.7%. MOX limited irradiation performance is due to the physical behaviour of MOX pellets with respect to that of U02 pellets (higher temperature and greater fission gas release induced by higher power histories).

French utility target is to lift MOX fuel at the level of the uranium in four-batch core management by the beginning of the next century. MOX fuel will be equivalent to uranium fuel enriched to 3.70% and will reach 50 GWd/t.

In Germany, Switzerland and Belgium, the MOX FAs are used in short and long cycles and there is no special MOX measures in the normal fuel management. MOX and U02 FAs are still equivalent in terms of energy.

4. USING MOX : THE GLOBAL EXPERIENCE

4.1. Usable Feedback (table 4)

Up to now, more than 750 tHM MOX fuel (more than 2000 FAs) have been loaded in 29 PWRs and in 2 BWRs in Europe, corresponding to the recycling of about 35 t of plutonium.
In France, since 1987 about 1000 MOX assemblies have been loaded and 24 t of plutonium recycled in 17 PWRs (900 MWe - 17x17).

In Germany, since 1985, about 800 MOX FAs have been loaded in 9 PWRs (16x16 and 18x18) and 2 BWRs : Gundremmingen B and C (1344 MWe-9x9).
In Switzerland, about 160 MOX FAs have been loaded in PWRs : since 1978 in Benau 1 and 2 (PWR 350MWe - 14x14) and recently in Gosgen (PWR 1OOOMWe - 15x15).

In Belgium, since 1995 about 70 MOX FAs have been loaded in Doe1 3 and Tihange 2 (PWR 1OOOMWe - 17x17).

In Japan, KEPCO is scheduled to load 9 MOX fuel assemblies in Takahama Unit-4 in 1999 and 9 MOX assemblies in Unit-3 in 2000 and an increased number during the following cycles in order to reach the maximum MOX recycling rate of a quarter of the core. TEPCO also expects to start reloading BWR Fukushima I Unit-3 (784 MWe) in 1999 and Kashiwazaki-Kariwa Unit-3 (1100 MWe) in 2000 with MOX fuel up to a third of the core after around four successive refuelling campaigns.

In terms of performance, the MOX discharge exposures are currently lower but close to those of uo2.

In Germany, mean MOX FA discharge exposure is about 40 GWd/t, maximum MOX FA exposure is about 48 GWd/t in commercial operation. The trend in this field is increasing and now many of the MOX FAs under irradiation will reach mean discharge exposure around 48 GWd/t.

In France, the average burn-up rate per MOX reload is maintained at around 37 GWd/t and the maximum assembly burn-up rate at 41 GWd/t, compared with the 50 GWd/t reached by UO2 assemblies obtained in annual cycle (3 cycles for MOX and 4 cycles for U02). By the beginning of the next century, the implementation of an optimised core management will increase MOX burn-up rate close to 50 GWdlt.

4.2. Reactor Operation

Reactor operation in base load and load follow mode has not raised any problems to operators. The main effect to be taken into account by operators is the improved axial neutronic stability due to the hardness of the neutron spectrum. One benefit observed was a cut in the volume of liquid waste due to the weaker influence of xenon : the volume of liquid waste generated during load follow decreases by about 30%. In France, where more than 75% of electricity is nuclear generated, load follow has been authorised and applied to all reactors recycling plutonium since 1994.

The loading patterns take account of the recommendations concerning MOX fuel induced by the physical properties of plutonium. One example is the limited number of MOX assemblies at the core periphery to prevent, if necessary, a build up of fluence in the reactor vessel, or under the RCCA to prevent the decrease of their efficiency. Loading pattern optimisation with these constraints is achieved without any difficulty.

Improvements of computer programmes relating to core behaviour supervision have resulted in greater consistency between predicted values and measurements taken during start-up tests and operation (neutronic flux maps). Computer models to predict the behaviour of MOX rods have also been validated by physical measurements taken during operation as well as by post-irradiation inspection.

Regarding the behaviour of MOX rods during transients, power ramp tests have been implemented at different burn-up rates, they have shown that MOX fuel behaves equal or better than UO2 fuel in terms of pellet/cladding interaction.
To justify the acceptable behaviour of the MOX during RCCA ejection transient (RIA), 3 MOX rod tests have been carried out in the Cabri reactor (Cadarache - France), the results show sufficient safety margins between the maximum values of stored enthalpy observed during the transient and the physical limit.

Globally, the operation of MOX-recycling LWRs is considered as satisfactory as the operation of pure uranium cores.

4.3. In-core fuel behaviour

Today, some 2000 MOX FAs have been loaded into LWRs, and more than 500 of them have been unloaded with an average discharge exposure reaching 35 to 40 GWd/t. So far, the behaviour of these MOX assemblies has been satisfactory.

The MOX rod failure statistics are within the range of the uranium rod failure statistics and the small failure frequency is not dependent on burn-up. As expected no MOX specific failure type appeared up to now. The MOX fuel in the LWRs has the same good reliability as the corresponding uranium fuel.

As an example, in France, after eleven years of reactor operation with MOX assemblies, corresponding to more than 70 reactor-cycles, only 2 MOX rod failures have been detected (due to debris), constituting an adequate demonstration of the outstanding performance of this fuel, which compares with UO2 for reliability.

5. MOX R&D

A number of experiments (irradiated materials, physical properties) and studies (core management, safety analysis) are under way with a view to achieve higher MOX discharge burn-up rates (50-60 GWd/t) and more economical core management, to reach the parity between MOX and **uo2.**

In France, out of pile studies, analytical experiments in test reactors and hot cell examinations of power reactor fuel rods have been conducted on the current MIMAS product up to 55GWj/t. The results confirm the good behaviour of this fuel in nominal operating conditions as well as in off-normal situations (cladding failure, class 2 incident transients, RIA).The main difference between U02 and MOX is a larger gas release enhanced by a higher linear heat rate at high burn-up : comprehensive investigation and modelling of the related phenomena are under way. Irradiation of lead fuel rods for a fifth cycle in a power reactor is in progress to reach a 60GWd/t target burn-up. In a more long term perspective (2010), an extensive RetD programme has been launched to support a 70GWd/t target burn-up, based on improving fission gas retention by developing optimized fuel microstructure.

In Germany, to cover the next burn-up step for MOX assemblies with average burnups over 60 GWd/t, utilities participate in the ARIANE program (also supported by Belgian, Swiss and Japanese utilities) to verify the data basis of actinides for depletion codes. Further on, Germany plans to participate in new RIA experiments to cover high enthalpy input conditions beside the operational experience. even if such conditions are extremely imurobable in German LWRs. The German

In Japan, the effect of plutonium spot, helium accumulation, and fuel failure were investigated up to high burn-up for MOX fuel rod. MOX fuel manufactured with MOX powder obtained by microwave heating technique tends to include a small size of plutonium spot. Its plutonium content in the spot, which is controlled with Pu-U ratio in a conversion process, is much lower than in MOX fuel manufactured by a direct mechanical mixing of PuO2 and U02 powders. Irradiation tests shows that above properties of PNC MOX fuel suppress a FP gas release, almost comparable to U02 fuel rod. Amount of helium released seems larger than in a U02 rod. This effect is included in the MOX fuel design to calculate an internal pressure.
PIE results of 60 GWd/t irradiated rod showed that there is still no significant difference between MOX fuel behaviour and U02 fuel. Power transient in BR-2 tests up to 600 W/cm demonstrated no breach of MOX fuel rod. Maximum linear heat of 440 W/cm in the present BWR is guaranteed by this results.

In order to verify the MOX fuel integrity which provides enhanced operational flexibility and the fuel safety tolerance, fuel segments pre-irradiated in Fugen since March 1987 were supplied for RIA tests to JAERI. Failure limit of MOX fuel under off-normal condition is currently investigated.

6. CONCLUSIONS

Reprocessing/recycling technology has reached maturity in the main nuclear industry countries. Spent fuel reprocessing and recycling of the separated fissile materials remains the main option for the back-end cycle.

Today, the operation of MOX-recycling LWRs is considered satisfactory. Experience feedback shows that, in global terms, MOX cores behaviour is equivalent to that of U02 cores in terms of operation and safety. The reliability of MOX fuel in LWRs has been confirmed by the experimental results as well as the operating experience to be equivalent to U02 fuel.

MOX fuel developments are in progress so as to achieve higher burn-up in order to manage MOX and U02 fuel with similar performances in terms of safety, reliability, flexibility. All R&D results show no unexpected behavior up to now and we have faith in further equivalent U02 and MOX operation under operating conditions of the future.

Table 1 : LWRs MOX fuel fabrication plants currently in operation

Manufacturing process	plant	capacity (tHM/Y)	location country	operator
MIMAS	P0	35	Dessel-Belgium	BN
MIMAS	CFCa	30	Cadarache-France	COGEMA
MIMAS	MELOX	100	Marcoule-France	COGEMA
SBR	MDF	8	Sellafield-UK	BNFL
SBR	SMP start-up	120	Sellafield-UK	BNFL

Table 2 : Plutonium in MOX fuel isotopic composition

Plutonium origin	Fissile P u (%)	Pu238 (%)	Pu 239 (%)	Pu240 (%)	Pu 241 (%)	Pu 242 (%)	Am 241 %	Pu content (%)	U02 enrichment equivalent (%)
U02 3,25 % 33GWd/t 5 years cooling 2 years storage	69,0	1,8	57,9	22,5	11,1	5,6	1,1	5,3	3,25
U02 3,7 % 45 GWd/t 9 years cooling 3 yearsstorage	63,l	2,5	54,5	25,2	8,6	7,9	1,3	7,l	3,25
U02 3,7 % 45 GWd/t 9 years cooling 3 years storage	63,l	2,5	54,5	25,2	8,6	7,9	1,3	8,6	3,7

Table 3 : MOX utilisation world-wide
the current situation

Countries	Reactors in operation		Reactors licensed for MOX use		Reactors loaded with MOX	
Reactor type	PWR	BWR	PWR	BWR	PWR	BWR
France	57	0	20	0	17	0
Germany	14	6	10	2	9	2
Belgium	7	0	2	0	2	0
Switzerland	3	2	3	1	3	0
Japan	23	2s	1	1	0	0
Total	104	36	36	4	31	2
LWRs totai	140		40		33	

Table 4 : Using MOX in LWRs
the global experience

Country	MOX loaded		Plutonium loaded in tHM (estimated)	reactors number
	in tHM (estimated)	in FAs number		
FRANCE	450	1000	23	17 PWR
GERMANY	210	800	12	9PWR 2BWR
SWITZERLAND	60	160	3	3PWR
BELGIUM	30	70	2	2 PWR
TOTAL	750	2030	40	33

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Invited Paper

OVERVIEW OF SAFETY ANALYSIS, LICENSING AND EXPERIMENTAL BACKGROUND OF MOX FUELS IN LWRs

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Abstract

Current practices of safety analysis and licensing of MOX fuel utilization in LWRs are overviewed. MOX fuels are used as reload fuels in the existing LWR cores without changing any core or fuel rod design. Loading ratio, Pu content and maximum burnup are limited to meet the safety requirements for these current practices. General concerns on safety analysis are reviewed and national practices in major MOX using countries are introduced. Major requirements for experimental support for advanced utilization of MOX, such as burnup extension, are discussed.

1. INTRODUCTION

MOX fuel utilization in LWR started in USA, Italy and other countries in 1960's as an attempt of burning recycled plutonium in thermal reactors. Currently MOX fuel is used in LWRs in France, Germany, Switzerland and Belgium, and the utilization will be started shortly in Japan.

As these practices of MOX fuel utilization are the partial loading in existing LWR cores as replacement fuels, fuel rod design is 'basically the same as that for the U02 rods, and MOXAJ02 loading ratio, Pu content, burnup limit and other fuel/core parameters are limited to satisfy the conditions that MOX loading should not demand any modification on existing plant systems and equipment, and no modification of the existing plant fuel management strategy should be required.

The safety analysis and licensing reflect these current practices of MOX utilization. MOX/U02 loading ratio is limited to about l/3 (50% at maximum in Germany), and Pu content is limited to about 13% considering the different nuclear characteristics, physical properties and fuel behavior. Fuel burnup is also limited to a little lower level than UO_2 fuel due to limited experiences.

The burnup limit of MOX fuel use in LWR will be increased in accordance with the burnup increase of U02 fuel in future. However, there are some safety concerns about the fuel behavior of MOX fuel at high burnup. They are, for example, FGR (fission gas release), helium release and so on which are

possibly correlated with plutonium content and microstructure. Recent pulse irradiation tests at CABRI suggested that the MOX fuel might behave differently from UOz in accidental conditions in terms of a RIA (reactivity initiated accident).

2 . SAFETY RELATED CHARACTERISTICS OF MOX FUEL

For the safety related concerns, the following physical properties relating to thermal property are important.

- Melting temperature
- Thermal conductivity
- Thermal expansion
- Thermal creep

However, the plutonium concentration of MOX fuel for LWR is low, and these differences from UO_2 fuel are relatively small.

Concerning the MOX fuel performance in LWRs, it is recognized that the MOX fuel rod behavior is very similar to that of standard UO2 fuel rod and holds no additional problems with the possible exception of high gas release and hence rod internal pressure increase at high burnup, because MOX keeps relatively higher linear power than UO2 in high burnup conditions. The main concerns on the MOX fuel rod behavior are as follows.

- Fission gas release
- Helium gas accumulation and release
- Swelling
- Fuel centerline temperature
- PCMI (pellet cladding mechanical interaction) under power ramping

Plutonium in reprocessed fuel is composed of four principal isotopes: ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu. Thermal neutron absorption cross-sections of plutonium isotopes, especially those in ^{239}Pu and ^{241}Pu are much larger than that in ^{235}U. This causes hardening of neutron spectrum. ^{240}Pu has a very large resonance absorption cross-section near the thermal energy region. Fraction of delayed neutrons in the fission of ^{239}Pu and ^{241}Pu is smaller than that in the fission of ^{235}U. ^{240}Pu is converted to fissile ^{241}Pu by absorbing a neutron. This process gives higher conversion factor.

The above-mentioned characteristics of plutonium isotopes bring the following concerns when MOX fuels are introduced in the LWR cores;

(1) The hardening of the neutron spectrum reduces the reactivity worth of control rods and soluble boron, and hence reduces shutdown margin of LWR cores.
(2) Moderator temperature and void coefficients shift to more negative values. This causes larger difference in reactivity between cold shutdown and hot full power conditions.
(3) Fuel temperature Doppler coefficient becomes larger negative value.
(4) &becomes smaller. This makes a reactivity transient faster.
(5) Power of a MOX fuel adjacent to water hall or UO_2 fuel where neutron spectrum is softer becomes higher giving high local power peaking.
(6) Prompt neutron lifetime becomes smaller due to large neutron absorption cross-section in plutonium isotopes.
(7) The reactivity of MOX fuel decreases more slowly with burnup due to larger conversion factor.
(8) Larger neutron absorption cross-section gives larger radial peaking factor within a MOX fuel.
(9) MOX fuel produces larger amount of helium gas.

These features of MOX fuel in LWR core should be properly addressed in the safety evaluation.

3. SAFETY ANALYSIS AND LICENSING

3.1. General Trend

Current practice of MOX fuel utilization in LWRs is the partial loading in existing LWR cores as reload fuels. Therefore, fuel rod design is basically the same as that for the UO_2 rods, and MOX/U02 loading ratio, Pu content, burnup limit and other fuel/core parameters are limited to satisfy the conditions that

(1) MOX loading should not demand any modification on existing plant systems and equipment, and
(2) No modification of the existing plant fuel management strategy should be required.

Table 1 summarizes the limiting conditions of MOX fuel loading.

In the licensing procedures; it is required that safety analysis has to be conducted considering the effects of the MOX fuels regarding the physical parameters and irradiation behavior. Larger uncertainties specifically for power distribution are considered in some countries due to limited experimental background. Specific safety standards or guidelines relevant to MOX fuel loading exist in some countries, while the same standards for UO_2 are applied in the other countries. However, the safety requirements for MOX fuel are basically not different from the UO_2 fuels, and there exists no additional item bounding the safety requirements.

General observations obtained through the experiences of licensing procedures and operation so far with MOX fuels are that no significant effects were identified to indicate less safe than the UO_2 cores up to a limited percentage of MOX loading in a normal LWR core (max 50% in German reactor). For example, fission gas release data are shown in Fig.1. The data indicate a somewhat higher fractional gas release after two irradiation cycles (about 0.7 %) than UO_2 rods irradiated in similar conditions (about 0.2 %). After three cycles, the fractional release ranges from 1 to 7%. That difference mainly results from the quite high power levels of some MOX rods (ranging from about 150 to 220 W/cm) during the last irradiation cycle [I].

One of the safety concerns for future is for high burnup MOX fuels. Concerning MOX fuel behavior during steady state and ramp test conditions in LWR, some international projects such as DOMO, PRIMO, FIGARO etc. have been established and have performed irradiation tests at test reactors. The Halden project also has performed MOX irradiation tests at HBWR.

TABLE 1. CONDITIONS FOR MOX FUEL ASSEMBLY LICENSES IN LWR

Name of country	Maximum ratio of MOX in core	Max. concentration of fissile Pu (%)	Max. assembly burnup (GWd/t)
France	30%	5.3 (Total Pu)	40
Germany	50%	4.65	48
Belgium	1/5	7.7 (Total Pu)	3
Switzerland	40%	5.5	50
Japan	1/3	8 (Total Pu: 13%)	45

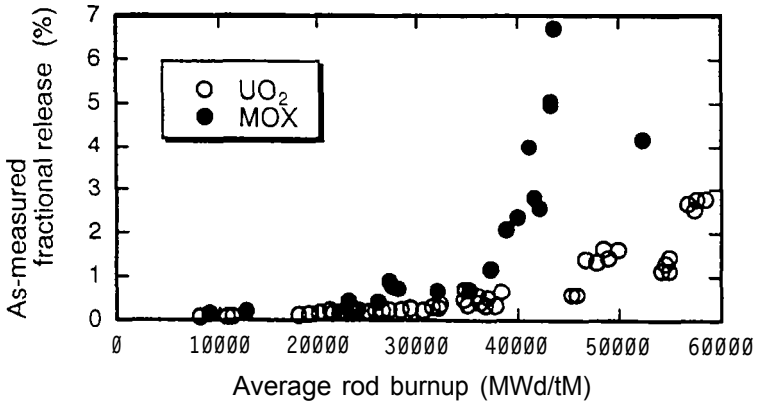

FIG.1. Fission gas release of MOX fuel and UO_2 fuel [I]

Concerning MOX fuel behavior under accidental conditions, experimental results of the NSRR and CABRI RIA tests indicated the large effects of fuel burnup for over 50 GWd/t. Some concerns for LOCA (loss of coolant accident) are also cited due to quite limited experimental background.

Regarding RIA, new regulatory guide for high burnup fuels (up to 55 GWd/t) has recently been issued in Japan. Regulatory criteria for higher burnups are still under discussion worldwide including implementation of international experimental cooperation for RIA.

3.2. National Practices

3.2.1 Japan

Utilization of MOX fuel in Japan started in ATR (Advanced Thermal Reactor) FUGEN in 1978, and over 680 MOX fuel bundles have been loaded to now [2]. This number is unique as a results of a single plant, and this experience provides important information for MOX fuel utilization in LWRs. Application of MOX fuel in LWRs was initiated as lead use of limited bundles in PWR (MIHAMA #l) in 1986, and BWR (TSURUGA #l) in 1988 [3]. Batch size utilization is to be started shortly. Currently, NSC (Nuclear Safety Commission)? safety review for l/4 MOX fuel loading in TAKAHAMA#3 and #4 (PWRs) is finished, and that for l/3 loading in FUKUSHIMA #3 (BWR) is underway.

A regulatory guide for MOX fuel utilization in LWRs was issued in 1995 by NSC [4]. The application of this new regulatory guide is limited to the MOX fuel loading of less than l/3 of the core, MOX fuel burnup of below 45 GWd/t bundle average and Pu fissile content of 8% at maximum. The guide admits the use of the current safety analysis methodology/codes for UO_2 to MOX core within the limitation mentioned above, but requires to perform safety analysis reflecting the properties of MOX fuel to the analysis model and inputs, as the functions of Pu content, fuel loading ratio and other loading conditions.

In the RIA analysis, the guide requests to consider the effects of lower melting temperature of MOX when applying the safety limit defined for UO_2 [5]. The effects of MOX fuel loading were also examined related to the reactor siting guideline [6]. In this examination, confirmatory dose calculation was performed for MOX fuel loading up to l/3 in every existing plants assuming 1% release of Pu inventory from the core to the containment as aerosol in a similar assumption done in the site evaluation of FBRs in Japan. It was decided based on this examination that no additional siting evaluation for Pu release should be needed for the currently planned MOX fuel loading in LWRs.

In the NSRR at JAERI, RIA test with ATR fuel is progressing [7]. Up to peak fuel enthalpy of 140 cal/g, fuel failure did not occur in the four ATR tests. In the ATR fuel up to burnup of 20 GWd/t with pre-pulse pellet/cladding gap of about 80 pm, PCMI occurred at peak fuel enthalpy of 110 cal/g or higher. It corresponds to residual hoop strain of the cladding tube, and the maximum residual hoop strain

exceeded 4%. Cladding deformation in the irradiated MOX fuel is much larger than that in the irradiated BWR fuel. During the base-irradiation, fission gas release of the ATR test fuel remained only 0.2% or lower. However, in the pulse irradiation, significant fission gas release occurred. Higher fuel enthalpy correlates with the higher fission gas release during the pulse irradiation. The fission gas release during the pulse irradiation in the MOX fuel with the burnup of 20 GWd/t is higher than that in the irradiated UO_2 fuels at the same burnup level. Up to the currently studied burnup range, 20 GWd/t, the MOX-effect did not show significantly different fuel behavior in comparison with UO_2 fuels. However, MOX effect, e. g. larger power distribution in pellet, appeared as higher fission gas release and larger fuel swelling. These suggest that decreased failure threshold in high burnup region.

Current utilization of MOX fuel is limited to l/3 loading and maximum burnup of 45 GWd/t. This is due to the existing needs for just decreasing excess Pu. But it is quite preferable to extend fuel burnup from the viewpoint of effective use of Pu and the decrease of number of spent fuels, making use of the good nuclear characteristics of Pu fuel. Full core loading might be another future task in order to rationalize the handling of MOX fuels by centralizing the MOX burning plants. Construction of a fully MOX-loaded ABWR plant is now being planned in OHMA in Japan.

R&D are underway to furnish fuel performance database for burnup extension of MOX, for example, in the Halden project, and producing nuclear database is also underway.

3.2.2. France [8,9, 10]

Since the first batch of 16 MOX fuel assemblies loaded in Saint Laurent Bl plant, in 1987, about 960 MOX fuel elements (which amounts to approx. 23 t of Pu) were recycled in EDF PWRs.

The status of MOX utilization in France is as follows.

- 20 reactors among the twenty-eight 900 MWe PWRs (3 loops CPl - CP2 series) are licensed for plutonium recycling up to 30 % of the core. 16 reactors are actually loaded with MOX.
- The number of reactors involved in plutonium recycling depends on the production capacity of the MELOX plant and the availability of some extra recycling capacity. Thus, EDF is planning the progressive introduction of MOX fuel into the 8 others 900 MWe PWRs (3 loops CPl - CP2 series). Public inquires will be commissioned.

As regards in core fuel management, actual loading pattern optimization is performed with a view to prevent a fluence build up in the reactor vessel. This is achieved by limiting the number of fresh MOX assemblies at the core periphery. Load follow operation was also performed without any specific difficulty. As a matter of fact, thanks to the lower influence of Xe within MOX cores, a cut (by approx. 30 %) was observed in the volume of liquid wastes. No major specific discrepancy was observed between predicted and experimental values produced for start-up tests after refueling outages. The behavior of MOX fuel elements has been satisfactory. Two clad failures have been observed probably due to the presence of debris.

As regards the assembly design and more specifically the fuel design, the average Pu content is adjusted to reach energy equivalence with 3.25 % enriched uranium fuel. The maximum average Pu content authorized is 5.3 %. In the short term, this limit should be extended to about 7 % to maintain such equivalence, taking into account the evolution of plutonium composition. In terms of in core fuel management, the so called “hybrid MOX” scheme is implemented, where UOX fuel elements (3.7 % enriched) perform 4 cycles and MOX elements 3 cycles in assembly burnup rate at 40 GWd/t. Design studies are underway to reach the objective of “MOX parity” that is, to have MOX fuel elements equivalent to 3.7 % enriched UOX elements. In that case MOX fuel elements would perform 4 batches fuel management and reach 50 GWd/t.

The higher quantity of plutonium in a MOX core induces a neutron spectrum shift toward higher energy levels which tends to a reduction of control systems efficiency (soluble boron and AIC control rods). The impact of such reduction has been assessed and minor modifications of reactor control

systems were necessary in order to maintain safety margins. Since PWR protection and safeguard systems designs are mainly governed by a limited number of accidental situations, transient calculations were performed in order to check accidental situations which have to be taken into account in the safety demonstration. The major reactor adaptations were the following:

- 4 shutdown RCCA added in order to increase antireactivity margin (8 excess locations were actually available in 900 MWe PWRs),
- boron concentration of the refueling storage tank was increased from 2,000 ppm to 2,500 ppm.

As regards accidental situations, the major impact of MOX elements is related to LOCA and breaks on secondary loops and more specifically steam line break.

As for UOX cores, the safety analysis for MOX cores licensing has to mainly focus on such accidents as RIA and LOCA. The CABRI REP Na program was launched on CEA/IPSN CABRI experimental reactor, in order to assess and validate the behavior of irradiated fuel rods during a RIA. Figure 2 illustrates the results obtained. The main characteristics or results of this program are the following.

- 7 tests on UOX fuel elements up to 64 GWd/t
- 3 tests on MOX fuel elements up to 55 GWd/t
- The failure threshold mostly depends on the initial cladding state (corrosion, hydriding level) as well as on the energy pulse width.
- As regards MOX fuel, the maximum enthalpy assessment in the case of a rod ejection accident leads to approximately 60 cal/g (at 45 GWd/t, local burnup), where the CABRI results showed no failure at equivalent conditions.

Experiments and studies are under way aimed at improving in core fuel management scheme using MOX elements, based upon higher discharge burnup rates. Such studies mostly rely on the following issues.

- MOX fuel performances assessment, and understanding of fuel rod behavior
- Fuel management studies (actual four batches fuel management schemes, extended cycle length.. . .)
- Optimizing fuel rod design to accommodate higher fission gas release rates
- Reactor/core safety studies using enhanced methodologies based upon detailed calculations

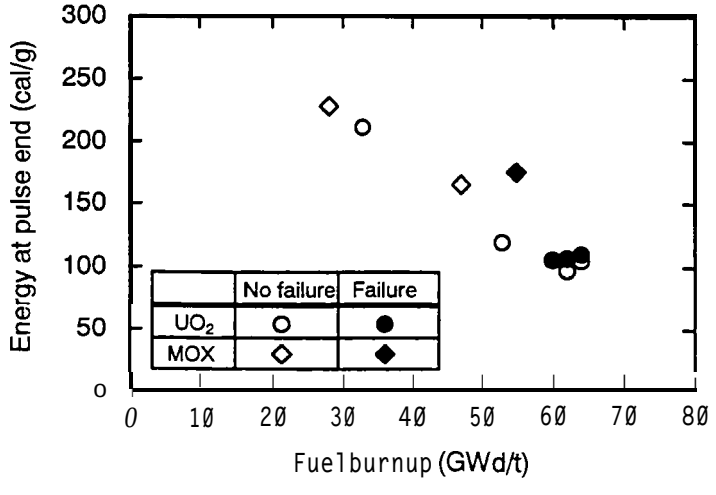

FIG.2. Results of CABRI experiments

3.2.3. Germany

The technology of MOX fuel has been developed in Germany since 1966. The MOX programs at the PWR Obrigheim and the first BWR at Gundremmingen were the basis for testing and validation of the methods of analysis. The results have been used to support the licensing of MOX fuel in other German NPPs [1 1].

Up to now the German experience with MOX fuel assemblies is based on the insertion of 768 MOX fuel assemblies with more than 106,000 MOX fuel rods in 18 LWRs and in 1 pressurized heavy water reactor, covering different Pu compositions.

Significant experience has been gained from evaluating the cycle length, power distribution, reactivity coefficients, and control rod worth of cores containing changing numbers of MOX fuel assemblies in different positions [12].

The results of post-irradiation investigations of MOX fuel rods range from 6 to 47 GWd/t and comprise poolside inspections on fuel assemblies and measurements of individual fuel rods [13].

Any design of MOX fuel assemblies and MOX containing cores must fulfill the same safety requirements as in case of cores with UO_2 fuel. The relevant rules and regulations for Germany are formulated as "Safety Standards of the Nuclear Safety Standards Commission (KTA)" for the "Design of Reactor Cores of Pressurized Water and Boiling Water Reactors" (KTA 3 10 1) and valid for all types of new fuel assemblies [1 1].

The technical feasibility is examined on the basis of realistic and enveloping designs. The different items which have to be considered are given in Table 2. The categories of requirements for each area of analysis are indicated, assuming normal operation conditions or accident analysis. Each topic has to be assessed prior to the insertion of MOX fuel assemblies.

TABLE 2. SAFETY EVALUATIONS RELATED TO MOX FUEL ASSEMBLY LICENSES IN GERMANY

	Normal Operation		Accidents
	Categories of Requirements		
Area of Analysis	Reactor Core	Spent-Fuel Pool and New Fuel Store	Transients, LOCA,[a] External Events
Neutron physics	MOX-FA-Design Core characteristics	Subcriticality Decay heat Control rod worth	B worth Reactivity coefficients
Thermal hydraulics	Unchanged	—	□
System dynamics	Control rod worth	—	As above
Fuel rod design	Fission gas pressure Corrosion	—	Fuel rod failure limit
FA [b] structure design	Unchanged	—	Unchanged
LOCA analysis	□	□	Evaluated
Radiological aspects	Activity inventory	Activity inventory Release rates	Activity releases

[a] LOCA = loss-of-coolant accident.
[b] FA = fuel assembly.

TABLE 3. CURRENT STATUS OF MOX LICENSING FOR LWRS IN GERMANY

Reactor Plant type	Status of Licence	Pu_{fiss}-Content [w/o]	MOX-FAs per Reload	MOX-FAs in the Core [%]
PWR: KWO	in use	3.8	8	26
GKN I	in use	3.04		9
GKN II	in use	3.8 [a]		37
KKU	in use	3.5 [d]	16	33
KKG	in use [e]	3.07 [a]	16	33
KKI 2	in use	equiv. 4.0 U235	24	50
KWG	in use	3.2	16	33
KBR	in use	equiv. 4.0 U235	- [c]	[c]
KKP 2	in use	4.65	24	37 [b]
KKE	granted [e]	equiv. 4.0 U235	16	25
KWB A	in preparation [e]	equiv. 3.5 U235	24	42
KWB B	in preparation [e]	equiv. 3.5 U235	2 4	42
KMK	in preparation		24	39
BWR: GUN B/C	in use [e]	2.57/3.6	2x64	38
KKB	in preparation			
KKK	in preparation			

[a] changes in the carrier material and/or Puquality can be compensated
[b] temporary restriction
[c] according to amount of Pu generated in the plant
[d] max. nominal Pu_{fiss} content of fuel rod
[e] modification or extension in preparation

Mechanical design analyses taking into account the irradiation behavior are described in Ref. [12] and are content of another session at the present conference, as well as nuclear design.

The status of licensing for German PWRs and BWRs is indicated in Table 3.

One item currently being discussed in Germany in connection with the high burnup issue (as in other countries) is the RIA issue. Whereas German safety assessors say that the "experimental data base for high burnup MOX fuel rod behavior under LOCA and RIA conditions should be updated" [14], industry points out that RIA is not a safety issue because there are sufficient margins which can be demonstrated by 3D calculations [15].

Meanwhile, the German RSK (Reactor Safety Board) showed that they would accept a gradual increase of burnup under careful observation of all relevant phenomena for UO_2 and MOX fuel.

The following conclusion is taken from the cited paper of German safety assessors [14]: "As a conclusion it can be stated that MOX fuel influences some safety related parameters which has to be accounted for in the safety analyses. Up to an amount of about 50 % MOX assemblies in a normal LWR core, though, no effects were identified during the numerous licensing procedures concerning MOX insertion in German LWRs, which would indicate that an operation with MOX fuel were less safe or would demand an alteration in safety systems or even different rules and regulations than operation with UO_2 fuel only."

3.2.4. *Belgium*

The spent fuel definitively unloaded from the units Doel 1, Doel 2 and Tihange 1 between their start up and 1990 is being currently reprocessed. The resulting plutonium was originally devoted to be used in fast breeder reactors. Due to the delay of the fast breeder option, it was decided to use about 4.7 tons of Pu under the form of MOX fuel in existing PWR. Given the current uncertainties existing in the back-end policy (reprocessing and long term disposal of spent fuel assemblies are still open options in Belgium), very specific objectives were defined:

- MOX loading should not demand any modification on existing plant systems and equipment: for example, no additional control rod should be needed.
- No modification of the existing plant fuel management strategy should be required.
- The Pu recycling program must be coherent with the Pu recovery schedule.

Given these objectives, a feasibility study showed that:

- The recovered Pu had to be burnt at a relatively low (20 %) recycling rate in 2 units.
- The designed discharge burnup had to be significantly increased regarding the limits existing abroad (especially in France).
- The Pu content had to be as high as 7.7 % total Pu to be energetically equivalent to a 3.8 % U02 fuel.
- A specific "radial zoning" of the assembly had to be designed.

Given the low Pu recycling rate, the impact of the use of MOX fuel on the safety analysis was limited: only LOCA, steam line break, Rod ejection, Rod misalignment and Rod Drop accidents had to be reviewed.

Regarding the radiological consequences, the studies have shown that no existing release limit in normal operation and in accidental conditions was exceeded for MOX cores. It was also verified that the systems and equipment were not affected by the MOX presence in the core.

The Authorization to load MOX in the Doe1 3 and Tihange 2 reactors was granted in 1994, allowing a maximum of 37 MOX assemblies to be loaded in the core of each unit (on a total of 157).

Due to a limited international experimental background on MOX, the Belgian Regulatory Body required that larger uncertainties be applied on the calculated power distribution within the MOX assemblies.

The comparison between calculations and measurements performed during start up tests and operation did not show any larger discrepancies than for UOZ cores, except what regards the response of the incore fission chambers, which displayed a significant underestimation of the prediction compared to the measurement (4 to 10 %).

It was demonstrated that this underestimation did not affect the assembly power, but was mainly due to the detector response.

Nevertheless, due to this problem, an additional 3% penalty on the measured hot spots was required for MOX by the Belgian regulatory body.

A systematic underestimation of the measured critical boron concentration is also observed (between 30 and 60 ppm) leading to an underestimation of the actual cycle length.

3.2.5. Switzerland

The following assessment is based on a paper of the Swiss Federal Nuclear Safety Inspectorate, presented recently at a meeting on safety of operating reactors [16].

Experience with licensing and insertion of MOX fuel in Switzerland is reported to be very satisfactory.

Insertion started in 1978 in one PWR, where a loading of 34 % of the core with MOX fuel has been achieved. The presently licensed value is 40 %. For another PWR, the corresponding numbers are 16 % and 36 %. Operating experience in the a.m. plants has been satisfactory. No significant differences in performance of UO_2 and MOX fuel have been observed.

The basis of the licensing is a safety analysis, with similar considerations as already given in Table 2. Of special interest are all properties which are specific to MOX fuel. Therefore, a spectrum of operational transients and design basis accidents was evaluated for a MOX reference core, for one of the a.m. PWRs, and compared to a UO_2 reference core, e.g.:

- Control rod ejection accident
- Loss of coolant accident
- Main steam line break accident.

Additionally, detailed analysis of control rod worth, subcriticality and radiological risk related aspects have been worked out.

One interesting remark should be mentioned, related to mechanical fuel rod design for normal operation: "It was realized that the power histories considered for fuel rod design calculations would not cover the anticipated loading requirements for MOX cores. Therefore, a statistical (probabilistic) fuel rod design methodology, covering the more demanding fuel rod power histories without significantly affecting existing safety margins, has been evaluated and is being implemented" [16].

As demonstrated by the analyses, the insertion of MOX fuel in the considered range has only a small influence on the plant behavior. All safety criteria, valid for UO_2 fuel cores, could be fulfilled by the MOX cores, too, with the result, that no MOX specific restrictions exist.

3.2.6. Other countries

. USA

Between 1965 and 1979, 97 MOX fuel assemblies were irradiated at 3 PWRs and 3 BWRs. However, now, the activities with respect to MOX in the USA are not related to the recycling of Pu from commercial reactors, but only within the context of the disposition of weapons grade Pu.

- The Nether land

  Between 1971 and 1988, 12 MOX fuel assemblies were irradiated in BWR plant.

- Sweden

  Three MOX assemblies were irradiated at BWR in 1974.

- [illegible]

  70 MOX assemblies were irradiated at a BWR and a PWR between1965 and 1975.

SUMMARY

Current practice of MOX fuel utilization is partial loading as reload fuels to the existing plants. The fuel rods are basically of the same design as that for UO_2 rods, and MOX/UO_2 loading ratio, Pu content and burnup are limited so as not to demand any modification on existing plant systems and equipment.

In the licensing, similar procedures and safety analysis methods to those for UO_2 fuel are adopted in most of the countries, and it is requested to consider the effects of MOX fuel loading in the safety analysis. Experience indicates that no significant effects were identified to indicate less safety than the UO_2 cores up to a limited percentage of MOX loading in a normal LWR core.

The licensed burn up for MOX fuel is limited to the same or a little lower level of burnup compared with UO_2 fuel due to the lack of irradiation test data. The possible future task is to furnish test data of high burnup MOX fuels under normal operation and accidental conditions.

To extend the database under normal operation, some international projects such as DOMO, PRIMO, FIGARO, etc. have been established and have performed irradiation tests in test reactors. The

Halden project also has performed MOX irradiation tests at HBWR. As for transient and accidental conditions, CABRI and NSRR have performed MOX fuel irradiation tests under RIA conditions.

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Invited Paper

MOX FUEL FABRICATION AND UTILIZATION IN FAST REACTORS WORLDWIDE

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Abstract

The paper discusses the main results acquired from the activities on developing and updating the technology in the course of the MOX fuel production and its operation in Phenix, PFR, SuperPhenix, JOYO, MONJU, BN-600, BN-350 and BOR-60. The spent fuel reprocessing experience is also presented in the paper.

1. INTRODUCTION

Currently only in three countries, namely, Russia, France and Japan demonstration and commercial fast power reactors are in operation and MOX (UO_2PuO_2) fuel technology is under development for reactors of that type.

In some countries, e.g., UK, USA, Germany and oth., fast reactors and MOX fuel production facilities were either decommissioned and dismantled or suspended. Nonetheless, this situation was preceded by a rather long period of their operation and, as a result, a large scope of technological and experimental experience was gained on the MOX fuel operation, including its fabrication process and irradiation behaviour. This kind of experience was gained in Western Europe, USA, Japan, Russia and in the former Soviet Union in operating the reactors Rapsodie, Phenix, SuperPhenix (France); the MOX fuel experiments in DFR and PFR (UK); the KNK-2 investigations of MOX for SNR-300 (Germany); EBR-II and FFTF (USA); JOY0 and MONJU (Japan); MOX fuel irradiation in BOR-60 and BN-600 (Russia) as well as in BN-350 (Kazakhstan).

The experience gained currently is sufficient enough to understand what kind of problems might arise when very high burnup of MOX fuel is reached in fast reactors, i. e., the burnups that make fast power reactors competitive.

2. EXPERIENCE GAINED FROM THE MOX FUEL OPERATION IN FAST REACTORS

Below the data are presented that summarize the experience gained on the MOX fuel operation in experimental, prototype and commercial fast reactors.

For the facilities that operated and operate currently in Western Europe, USA and Japan the MOX fuel has always been the main one for all types of reactors except DFR.

In Russia, in the former Soviet Union, MOX fuel operated in the cores where uranium dioxide was used as driver fuel. An experience has been also gained in Russia in operating the experimental reactors with plutonium dioxide fuel. The main characteristics of the fast reactors fuel are presented in Table 1.

2.1. Experimental Fast Reactors

The onset of activities in Russia on the use of Pu as nuclear fuel dates back to the late 50-ies. In 1959 a 5 MW fast reactor (BR-5) started operation at Obninsk. Plutonium dioxide was used as nuclear fuel. The first loaded fuel rods operated in the reactor to a burnup of ~ 6,5% h.a. Then, following the testing of the core with monocarbide uranium fuel in the reactor, its power was raised to 10 MW (BR-10) and three more cores were consecutively tested in the reactor: two cores with plutonium dioxide fuel and one – with uranium mononitride. A maximum burnup of ~ 14% h.a. was reached in updated designs of plutonium dioxide fuel rods.

In 1977 the construction of the fast pulse sodium cooled reactor (IBR-2) was completed at Dubna. The reactor core with plutonium dioxide fuel rods has been successfully operated since 1978 until now (maximal burnup is ~ 6,6% h.a.).

In 1968 experimental reactor BOR-60 started operation in RIAR, Dimitrovgrad. Although the reactor was uranium dioxide fueled, its start up gave rise to systematic studies of oxide uranium-plutonium mixed fuel (MOX) used in power fast reactors. 16 pelletized fuel assemblies and 450 MOX vibro-fuel assemblies were irradiated in BOR-60.

Rapsodie operated at Cadarache, France, from 1967 to 1983. It was the first fast reactor designed with uranium-plutonium fuel that made it possible to acquire a large scope of experience for both fabrication and irradiation. The experimental program confirmed the feasibility of reaching high oxide fuel burnup and the high radiation resistance of cladding materials at high damage doses.

Experimental fast reactor DFR operated at Dounreay, UK, from 1959 to 1977. Its driver fuel was metallic one (uranium-molybdenum alloy, U-Mo). However, FAs with MOX fuel were tested in a lot of experiments.

In Japan experimental fast reactor JOY0 reached the first criticality with MK-1 core in 1977. Its rated power was 75 MW. The second phase of the reactor operation (MK-2) made it possible to reach a rated power of 100 MW beginning 1983. Various irradiation experiments were conducted in the reactor. The fabrication of fresh MOX fuel using plutonium, that was recovered from the reprocessed JOY0 spent fuel, and its testing allowed to close successfully the fuel cycle in 1984. The maximal burnup that was reached in the MK-2 core was 75 000 MW<day/ t.

KNK-2 reactor with a MOX fuel core operated at Karlsruhe, Germany, from 1977 to 1991. The fuel rods reached the burnup of – 100 000 MW<day/ t.

2.2. Prototype Fast Reactors

Four prototype fast reactors, namely, BN-350, Phenix, PFR and MONJU operated and are under operation throughout the world. Fast prototype reactor CNR-300 was also constructed in Germany, but it did not operate and its contribution to the MOX fuel development was connected with the MOX fuel core fabricated for CNR-300.

The BN-350 reactor has operated at Kazakhstan since 1972. Uranium dioxide is used as driver fuel for the reactor but FAs with plutonium oxide fuel were also successfully tested. In 1982 ten FAs with pelletized MOX fuel were loaded into the reactor core and in 1988 four more MOX fuel FAs were fabricated and loaded. In 1985-1987 two FAs with vibro-compacted MOX fuel were irradiated in the reactor. The main operational characteristics of the fuel are given in Table 1.

PFR operated at Dounreay from 1974 to 1994 with uranium-plutonium dioxide as standard fuel and it was intensively used as an irradiation tool. In the late 1982 Pu, recovered from the Dounreay reprocessing plant, was used to fabricate fresh FAs, thus closing the fuel cycle.

Phenix has operated at Marcoule, France, since 1973 and it is planned to be used till 2004. Its characteristics are comparable to those of PFR. Due to a successful start up and the effective operation up to 1990, Phenix provided an important experience on standard fuel and it allowed to acquire significant data for experimental fuel as well. The Phenix fuel was reprocessed at the appropriate plants and it was diluted by thermal reactors fuel. FAs, fabricated from reprocessed plutonium, were irradiated in Phenix, thereby closing the fuel cycle.

MONJU reached the fust criticality in 1994 and operated at 40% of its rated power until the sodium leak accident occurred in 1995. Plutonium enrichment of its inner core fuel reached **30%.**

2. 3. Commercial Fast Reactors

SuperPhenix operated at Creys-Malville, France, from 1985 to 1996 and it was stopped by French government' s decision. Its contribution is interesting from the commercial production viewpoint, but not because of the in-pile behaviour, due to the low burnup achieved following 320 EFPD of operation.

In Russia commercial fast reactor BN-600 started operation in 1980 and it has successfully operated until now. The driver fuel of the reactor is uranium dioxide, as in the case of BN-350. A definite number of FAs with MOX fuel is under permanent reactor irradiation. 20 FAs with pelletized MOX fuel and 6 vibro-compacted FAs have been successfully irradiated by now.

12 more pelletized FAs were fabricated in 1999-2000 to be irradiated

3. NUCLEAR FUEL FABRICATION

3.1. France's Experience [l-3]

3.1.1. In France, all standard and experimental fuel for fast reactors were fabricated in the Cadarache plutonium workshop, operated till 1991 by CEA and then by COGEMA. Its capacity was extended from 0.5 ton of MOX/year for Rapsodie line to 4 t/y for Phenix line and 20 t/y for SuperPhenix. Some PFR pins were also fabricated here since 1987. Now the production is mainly devoted to MOX for PWR, with an annual capacity of more than 40 t/y.

3.1.2. The process used consists in sintering pellets at high temperature in a reducing atmosphere. The successive steps are :

mixing and milling UOz and PuO_2 powders, in a mill with U balls,
granulation and incorporation of a lubricant (Zn stearate),
pressing of crude pellets,
sintering in a continuous circulating furnace, at 1650°C under Ar - 7 % Hz.

The diameter is reached after sintering without grinding (grinding is used for pellet obtained with a too large diameter).

Then the pins are fabricated under He atmosphere, by introduction of the fissile (and fertile) columns and other components inside the cladding. The end caps are welded by tungsten inert gas process. The spacing wire is wrapped by a specific machine and fixed to the end caps.

Clusters of pins are formed by fastening the bottom plugs to the rails of a bottom grid ; they are introduced in the hexagonal wrapper, which is finally welded on bottom and top structures.

3.1.3. Inspections are performed at each fabrication stage, to ensure a good quality of fabrication:

- for the pellets, the physical characteristics (diameter, linear mass, appearance), the chemical properties (Pu content, O/M, impurities and oths.) and the metallurgical characteristics are statistically checked,
- for fuel pins, individual checks concern : helium leak testing, leak tightness, ultrasonic examination of welds, ♋ count test, pellet stacks enrichment, radiographic examination,
- for the hexagonal wrapper, individual checks are made on the weldings, and on geometrical characteristics.

It must also be recalled that the cladding tubes and hexagonal tubes are individually checked (dimensions, absence of cracks, metallurgical characteristics) before being used in the plutonium workshop.

3.1.4. The tables 2 and 3 give some statistics illustrating the considerable experience gained in Pu fuel fabrication.

The technical options proved to be efficient to reach the objectives : the required fuel characteristics were held without noticeable difficulties, and no major evolution appears necessary.

Some points may be underlined :

- due to the use of contaminating and irradiating products, it was neccessary to increase mechanisation : from Rapsodie to SuperPhenix, all transfers between the various tight-containments became automatic, remote controlled ; also the equipments for fabrication and checking were automatised;
- a continuous effort was made to deal with the various fabrication wastes ⌋ they are selected according to their nature (activity, Pu content, etc.) and a part of them is reintroduced in the fabrication line;
- the radioprotection of workers was a constant concern, due to the increase of Pu quantities and the change from gas-graphite one to light water one. This challenge was successfully overcome, as illustrated by the following data : the averaged dose per man and per

year dropped from 7 to 3.5 mSv during the last 15 years ; since several years, practically all results are < 20 mSv/y, among which less than 1 % are in the range 15-20 mSv/y. This experience was benefit for the fabrication for PWR MOX fuel, the doses being kept very low by the extension of automatisation.

TABLE 1. OPERATIONAL CONDITIONS OF MOX FUE IN FAST REACTOR

Reactor	Max. LHGR, W/cm	Max. temperature of cladding "C	Burn-up, MW*day/t
Rapsodie fortissimo	450	650	90000 (in several FAs)
Phenix	450	650	90000 (inner reactor core) 11500 (outer reactor core)
Superphenix	420	620	70000
PFR	480	670	85000
KNK-2	430	600	66100/73400/1ooooo*
BOR-60 . *	490-520	700-710	100000-130000 (for pelletized fuel) 2800000 (for vibrofuel)
FFTF	430	680	100000-140000
BN-350 **	450480	650-690	70000-100000
6H-600 **	420450	670-690	95000-l 10000
Joyo (MK-I)	320	620	42000
Joyo (MK-II)	400	650	75000
Monju	380	675	94000

. min/average/max burn-up

. * the driver fuel for these reactors is uranium dioxide; the table gives some operational conditions for MOX FAs that were irradiated in the reactors

TABLE 2. CHARACTERISTICS OF FAST REACTOR CORES

Reactor	Number of S/A	Number of pins	Number of pellets	Fissile material (kg)	Pu (kg)
Lapsodie	64	2368	80000	563	200
Lapsodie fortissimo	66	4026	140 000	343	200
Phenix	103	22400	1565 000	5400	1000
Superphenix	364	98700	"8 000 000	55000	6000
PFR	72	23400	2100 000		
KNK2	14	2366	212 900	310	77
SNR 300	205	34030	4618400	17795	1536
Joyo (MK-I)	82*	7462	450000	310	166
Joyo (MK-II)	67	8509	520000	250	220
Monju	198*	33462	400000	-	1000 (fussile Pu)
BOR-60**	16 (pellets) 450 (vibro)	304 (pellets) 16650	20000	35 1915	7 383
BH-350**	14 (pellets) 2 (vibro)	1778 254	240000	355 50	90 12
BH-600**	20 (pellets) 12 (pellets)*** 6 (vibro)	4064 762	55000	815 150	205 30

* reactor core

** MOX FAs irradiated in reactor

*** manufactured for irradiation from 1999 to 2000.

TABLE 3. EXPERIENCE OF FUEL FABRICATION FOR FR AT CADARACHE (AT 31/12/98)

Type of fuel	From	Number of S/A	Number of pins
Rapsodie (*)	1964	611	28 600
Phenix (*)	1971	877	181 000
Superphknix	1979	769	208 400
P.F.R.	1987	-	9 555
Experiments in Superphknix	1995	3	799
Joyo (MK-I)	1972	119	-10000
Joyo (MK-II)	1980	352	-50000
Monju	1989	285	-50000

. *including experiments*

3.2. Japan's Experience [4,5]

3.2.1. In Japan all standard and experimental fuel for fast reactors was fabricated at the three MOX fuel fabrication facilities of JNC-Tokai:

- PFDF (Plutonium Fuel Development Facility), the purpose of which is the implementation of the basic research work and the fabrication of fuel for irradiation experiments. It started operation in 1966 and its maximum capacity is 300 kg Pu annually.

- PFFF (Plutonium Fuel Fabrication Facility), the purpose of which is the fabrication of driver fuel for JOY0 and FUGEN. It started operation in 1972 and its maximum capacity is 850 kg Pu annually.

- PFPF (Plutonium Fuel Production Facility), the purpose of which is the production of driver fuel for MON.JU and JOYO. It started operation in 1988 and its maximum capacity is 2.5 t Pu annually.

3.2.2. The fabrication process consists of the following steps:

- The starting materials are: UO_2, received from standard UF_6-UO_2 converters, and a co-converted $(U\text{-}Pu)O_2$ powder, obtained by microwave condenitration of plutonium and uranium nitrates mixed solution (Pu/U ratio is 1).

- The co-converted $(U\text{-}Pu)O_2$ or PuO_2 powders are mixed with UO_2 powder at the plutonium content specified for the final product.

- The powder is ball milled (a conventional ball mill is used), pre-compacted and granulated.

- Clean pellet scraps are crushed into powder that is recovered and reused.

- Clean powder scraps are sintered once and then treated in the same way as pellet ones.

- Dirty scraps are subject to storage and they are to be processed with purification.

3.2.3. The data, illustrating the MOX fuel fabrication experience, are presented in Tables 2 and 3.

The following aspects which are very important for the development of the fabrication technology should be noted:

- The use of pore formers for low density pellets (-85% TD) of MONJU.

- The development of the reduction technology for residual plutonium inside glove boxes.

3.3. Russia's Experience [6-8]

3.3.1. In Russia to fabricate the fuel containing plutonium, it was used or is under use either weapon's grade plutonium or that produced following radiochemical reprocessing of the spent fuel from the fast reactor breeding blankets (BN-350, BN-600).

At the first stage of R&D the mechanical blending of uranium and plutonium oxides was accepted as the main MOX fuel production technology. Uranium dioxide produced by means of ammonia precipitation and plutonium dioxide prepared through oxalate one (plutonium-ammonia pentaoxalate precipitation) were used. The technology using weapon's grade plutonium made it possible to fabricate rather a large quantity of MOX fuel that was successfully tested in SM-2, BOR-60, BN-350 and BN-600 reactors.

3.3.2. Until 1980 MOX fuel rods had been fabricated either at the experimental facility in the All-Russia Scientific Research Institute of Inorganic Materials, Moscow, (for SM-2, BOR-60) or at the pilot shop area of the PU "Mayak", Chelyabinsk (for BR-5, BR-10, IBR-2, BOR-60). In 1980 at the PU "Mayak" the pilot industrial facility "Paket" was specially constructed intended for a larger scale MOX fuel production. The facility's annual design output is -350-360 kg U-Pu oxide pelletized fuel (lo-12 FAs for the BN-600 and BN-350 reactors). Currently it is planned to reconstruct the "Paket" facility and increase its output to -50 FAs for the BN-600 reactor per year.

3.3.3. Along with the mechanical blending of uranium and plutonium oxides technology, a number of other methods has been developed in Russia.

The main task here is to minimize dusty operations to the extreme.

In 1978-1980 the All-Russia Scientific Research Institute of Inorganic Materials developed the technology of producing micro-sphere fuel through a sol-gel process. On the basis of those developments the facility "Zhemchug" was constructed. The gel formation was carried out by dispersing a cooled uranium-plutonium solution into a heated organic liquid. The further flow-sheet envisaged the washing of micro-spheres, their drying, calcination, sieving, reduction in a hydrogen containing medium, pressing and sintering of pellets. Using the above technology, pilot fuel rods were fabricated and successfully tested within 2 FAs in the BN-350 reactor.

However, the experience gamed showed that the sol-gel technology had serious shortcomings. The manufacture of pellets from granules, obtained through the process, entailed a

number of difficulties that inhibited the provision of the high and stable quality of pellets. Besides, the process resulted in a large amount of technological wastes that turned out rather dangerous. In 1987 the work using the above technology was stopped. In 1987-1988 the All-Russia Scientific Research Institute of Inorganic Materials developed a technology in accordance with which a pilot facility was constructed at the PU "Mayak" for producing low dust granulated fuel by uranium and plutonium ammonia co-precipitation method using surface-active substances (polyacrylamide).The facility was called "Granat" (Granulated Nuclear Fuel).

Both the method and the facility provided the production of low dust irregularly shaped granules with a stable and safe control of powder characteristics from which fuel pellets were readily manufactured that met the specifications. Using the technology, fuel rods for 13 FAs of the BN-600 and BN-350 reactors were manufactured.

A rather large scope of the work on carbonate co-precipitation of six valant uranium and plutonium has been carried out in the All-Russia Scientific Research Institute of Inorganic Materials. The work has been performing since 1985. The mastered technology is capable of preparing powders with a required bulk weight, rather good pressability and satisfactory flowability. Sintered fuel columns containing 2530% PuO_2 are a mono-phase solid solution that is 100% dissolvable in nitric acid.

Another method of fabricating MOX fuel – plasmo-chemical conversion (denitration of uranium and plutonium nitrate solutions mixture) – is under study. The potential feasibility of a rather large scale production of mixed oxides by means of this technology was demonstrated at a special facility. About 35 kg mixed powders prepared were used for producing pilot fuel rods for BOR-60.

All the above five methods of producing MOX fuel have a common result: namely, the manufacture of pellets for fuel rod loading.

3.3.4. A large scope of the work on vibro fuel rods is being carried out in Russia. This work is a constituent part of the task on creating the prospective fuel cycle of nuclear reactors based on the pyroelectro-chemical method of reprocessing irradiated fuel, resulting in high density granules prepared for producing vibro fuel rods. The particles density is 10,6-108 g/cm' and their size is from -10 ○m to 1 mm. In the course of the work physico-mechanical and technological characteristics of granulated fuel have been studied and the in-pile testing of experimental and pilot fuel rods has been carried out in BOR-60, BN-350 and BN-600 as well as their materials science studies.

In vibro fuel rods the problems of thermo-mechanical and physico-chemical pellet-cladding interaction are solved due to both the structural properties of the polydispersed granule based fuel column and the introduction of metallic uranium powder getter into a fuel composition.

Lately in the Research Institute of Atomic Reactors (RIAR), Dimitrovgrad, RF, studies have been performed in order to assess the feasibility of producing mixed oxide fuel from weapon's grade plutonium by a pyroelecro-chemical method resulting in its purification from some alloy additives, Ga in particular. The first batches of $UPuO_2$ granulated fuel have been produced and m-pile testing of 16 FAs (BOR-60) is being carried out. The technological features are as follows: high velocity of plutonium dissolution, a minimum of chemical and technological operations, compact equipment and the possibility of producing two types of granules: PuO_2 and $UPuO_2$.

These investigations reveal additional potentialities of the pyroelectro-chemical method combined with the vibrocompact technology aimed at solving the problems connected with weapon's grade plutonium utilization.

3.3.5. It is known that starting powder characteristics predetermine the choice of the pelletized fuel columns manufacture flowsheet.

Granulating operations are not needed for granulated (at the "wet stage") powders of "Granat type and of carbonate origin (AUPuK) having good flowability. These powders are immediately delivered for pressing. The powders prepared by the plasmochemical conversion method (PCC) and the mixtures of individual uranium and plutonium dioxides are subjected to the granulating operation to achieve the required flowability.

The technology of blending uranium and plutonium oxides and the preparation of PCC powders envisage special equipment that makes it possible to crush and mix (eddy bad blender) powders simultaneously for a few minutes. This method of powder mixing and crushing allows also to bring back and reuse in the technological process a rather large amount of pure wastes (scrap) obtained from the fuel pellets that were rejected for some reason or other. In the course of sintering the fuel pellets from such powders in standard modes (-1700-1750 ^{0}C for 2 hours in Ar-H_2 atmosphere) practically a fully homogeneous solid solution forms.

3.3.6. The subsequent technological operations envisage the loading of fuel pellets into a fuel rod cladding without grinding. The fuel rods are sealed by argon-arc welding using a non-consumable tungsten electrode (lower plug) and by magnetic field controlled arc (upper plug welding). The fuel rods are vacuum treated and filled with helium before the upper plug is welded.

The fuel rods spacing in FAs is carried out by means of a round wire of 1.15 mm diameter (central fuel rods) and an elliptic cross section strip (0.6 ⋄ 1.3) mm (circumferential fuel rods).

The wire is wound around fuel rods at a special machine and is contact welded at a cladding.

The control is performed in accordance with "The Program of Quality Assurance.. ." of the starting materials (tubes, wire, plugs, powders and 0th.) at each stage of manufacturing semi-finished products (fuel pellets, cladding tubes) and final products (fuel rods, FAs) as well as initial materials (tubes, powders, wire). The data on the MOX fuel production experience acquired in Russia are presented in Table 4.

4. OPERATION EXPERIENCE

The characteristics of the mixed oxide fuel will be reported in accordance with the following parameters:

- ability to reach high burnup;
- behaviour in off normal conditions;
- behaviour of failed fuel.

4.1. France's Experience [9-181

4.1.1. Reaching high burnup

The evolution of the mixed oxide is complex, under the influence of the radial and axial thermal gradients, the creation of fission products, and the interaction with the cladding. The numerous post irradiation examinations made on Phenix pins pointed out a transition region between 7 and 9 % bu that is characterized by the following processes inside fuel rods:

TABLE 4. EXPERIENCE OF MOX-FUEL FABRICATION IN RUSSIA (AT 31/12/98)

Type of fuel	From	Number of S/A	Number of pins	Place of fabrication
BOR-60 (pelletized)	1974	16	304	VNIINM (7 Fas) PU „MAYAK“ (9 FAs)
BOR-60 (vibro)	1980	450	16650	RIAR
BN-350 (pelletized)	1980	14	1778	PU „MAYAK“
BN-350 (vibro)	1984	2	254	RIAR
BN-600 (pelletized)	1985	32	4064	PU „MAYAK“
БН-600 (vibro)	1987	6	762	RIAR

- increase of fission gas release, shown in Fig. 1;
- fission gas bubble precipitation inside fuel pellet;
- decrease of intergranular caesium content;
- movement of fission product precipitates (MO, Pd, . ..) out of fuel pellet;
- opening of a so-called JOG-layer (Joint Oxide Gaine, as fuel to clad join) filled with fission products compounds (mainly caesium molybdate) without Uranium and Plutonium ; the evolution of the JOG width and its composition are illustrated in Fig.2, **3;**
- subsequent fuel shrinkage.

These observations have been confirmed by post-irradiation analysis of fuel pins (6.6 mm OD - annular pellets) highly irradiated (up to 21 at %) in PFR.

The role of the JOG-layer appears important for behaviour at high burnup, both with respect to stabilisation of fuel temperature and mitigation of fuel clad mechanical interaction (FCMI) to prevent its deleterious effects, the smear density should remain limited (-less than 0.85) ; fuel clad chemical interaction (FCCI) is another concern ; the affected zones on highly irradiated Phenix pins can reach 120-290 ○m ; and are located in the upper area of the active part stack and at the fissile-fertile interface ; the understanding made important progress, with evidence of the major role of Cs and Te produced by fission.

The evidence of these limiting phenomena implies to maintain an effort on R&D to improve understanding and increase the limits.

It must be recalled that another important limiting factor in the behaviour of clad and wrapper materials, for which substantial progress was also made-see the figure below.

Some outstanding experimental achievements in France

Burn-up : 27% on Rapsodie standard fuel
17,5% on Phenix standard fuel
7,5% on nitride in Phenix

Damage on core materials - claddings (in Phenix)
155 dpa on 15115 Ti SS
123 dpa on high Ni alloy
130 dpa on ferrito martensitic steel
91 dpa on ODS ferritic steel

Damage on core materials - wrappers (in Phenix)
155 dpa on martensitic EM10
127 dpa on 15115 Ti SS

4.1.2. Off normal conditions

Power and/or temperature transient may occur during reactor operation ; they can result in life limitations or failures, and so it is important to study them.

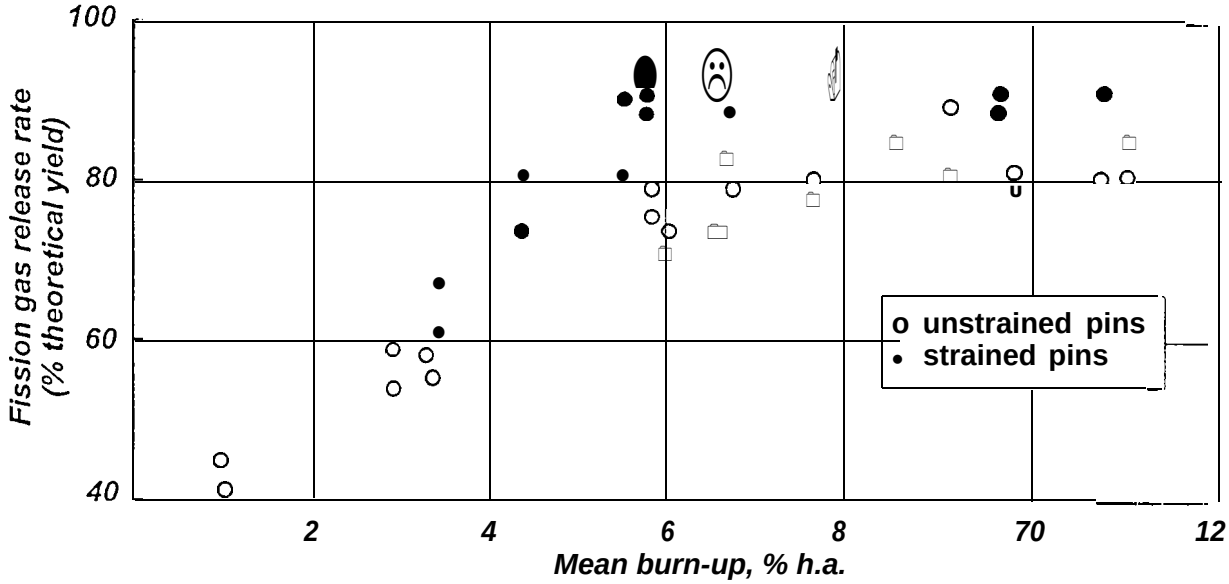

FIG.l. Fission gas release versus burn-up for PHENIX pins

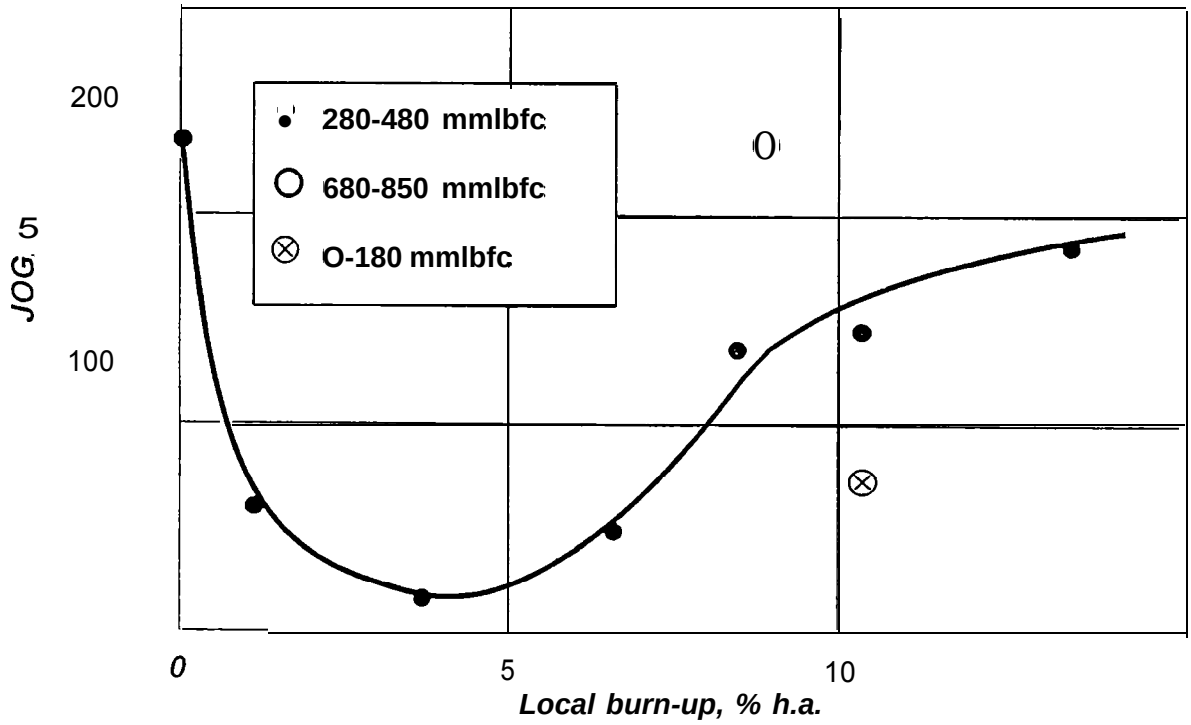

FIG.2. Axial evolution of the JOG* width in non unstrained PHENIX pins

* JOG - diametral extension of the fuel to clad joint

Since 1973, a comprehensive program was conducted in Cabri and Scarabee, consisting in about 60 different tests on fresh or preirradiated Phenix pins. As illustrated by the Fig.4, they provided information on failure condition. The demonstration was made than the mixed oxide fuel is able to survive a wide range of duty cycle power histories, and some off normal conditions including partial fuel melting.

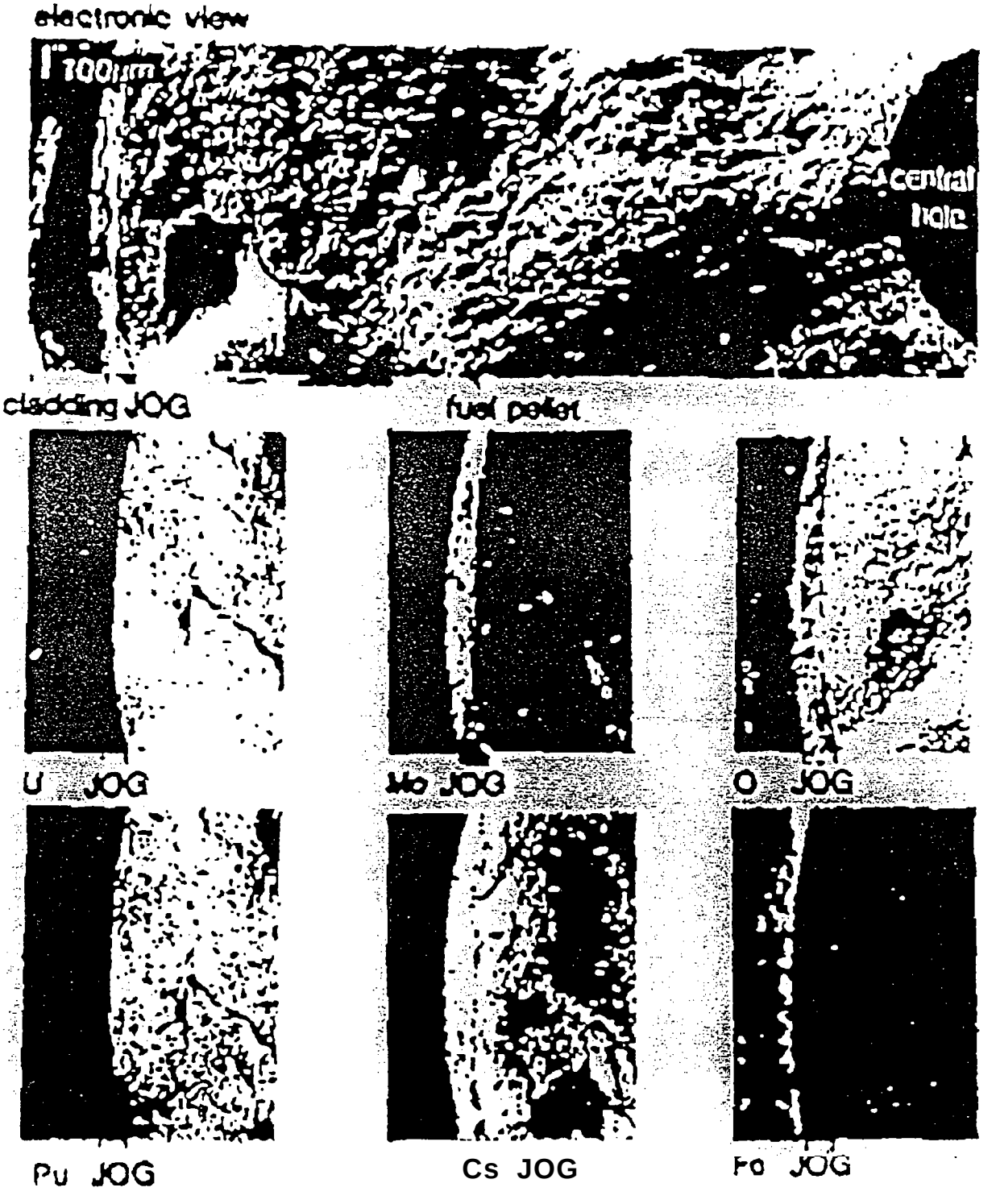

FIG.3. X-ray mapping of the JOG compounds at Peak Power Node at high burn-up without polishing

4.1.3. Failed pin behaviour

Failure of pin cladding are a concern, because the sodium and the mixed oxide can react. So an experimental programme was conducted on artificially defected pins in Siloé ; and a considerable experience came from Rapsodie (about 50 failures) and Phenix (15 failures), to which must be added about 45 failures in PFR and KNK-2.

The observations led to a good knowledge of the evolution of failed pins according to the cause and the age. The results are rather favourable : fuel loss is limited - for instance only one pin in Phenix led to loss estimated between 20 - 200 mg, there is no pin-to-pin propagation ; detection proved always possible. There is also no concern about the behaviour in storage condition.

These results should be confirmed for higher burnup and dose rates, taking especially into account the potential embrittlement of clad material at high doses.

No clad failure was observed in SuperPhenix ; which shows that a good control of fabrication may eliminate or set to a very low level the beginning of life failures.

4.1.4. Potential of mixed oxide fuel

All these results, and especially the statistical experience on standard fuel of PFR and Phenix, show that a burnup of 20 %, corresponding to a first target for a commercial reactor can be safely reached, in conditions as those proposed for PFR (annular pellets, smear density < 85 %, 520 W/cm at BOL, 650°C max for cladding). In fact no obvious physical limitation for higher values than 20 % appear ; for such ambitious targets, confirmation should be looked after, across experimental studies of the evolution of the phenomena described hereabove.

4.2. Japan's Experience

In the early period of development the irradiation experiments of fast reactor fuels, designed and fabricated in Japan, were carried out in DFR and in Rapsodie, though JOY0 MK-2 core, Phenix and FFTF (USA) were used for the irradiation experiments of MONJU fuels.

Fig.5 shows the irradiation achievements of Japanese fuels.

4.2.1. Reaching high burnup

A long life fuel development made clear the irradiation behaviour of fuel assembly with advanced austenitic satinless steel cladding up to 162 GW·day/t and with ferritic steel cladding up to 200 GW·day/t.

Development of the cladding material, which has both superior anti-swelling characteristics and high creep strength under the high temperature and high neutron dose circumstances, is important in order to accomplish the high burnup.

4.2.2. Off normal conditions [19]

A series of operational reliability tests of fuels under off normal conditions was carried out using EBR-2 (USA) from 1981 to 1995 as a collaboration research between Japan and USA.

The operational reliability test made it clear that the MONJU fuels had enough safety margin showing no fuel failure during transient operation up to the power of two times as much as the rated power, and that the in-reactor vessel on-line diagnosis of failed fuel was possible.

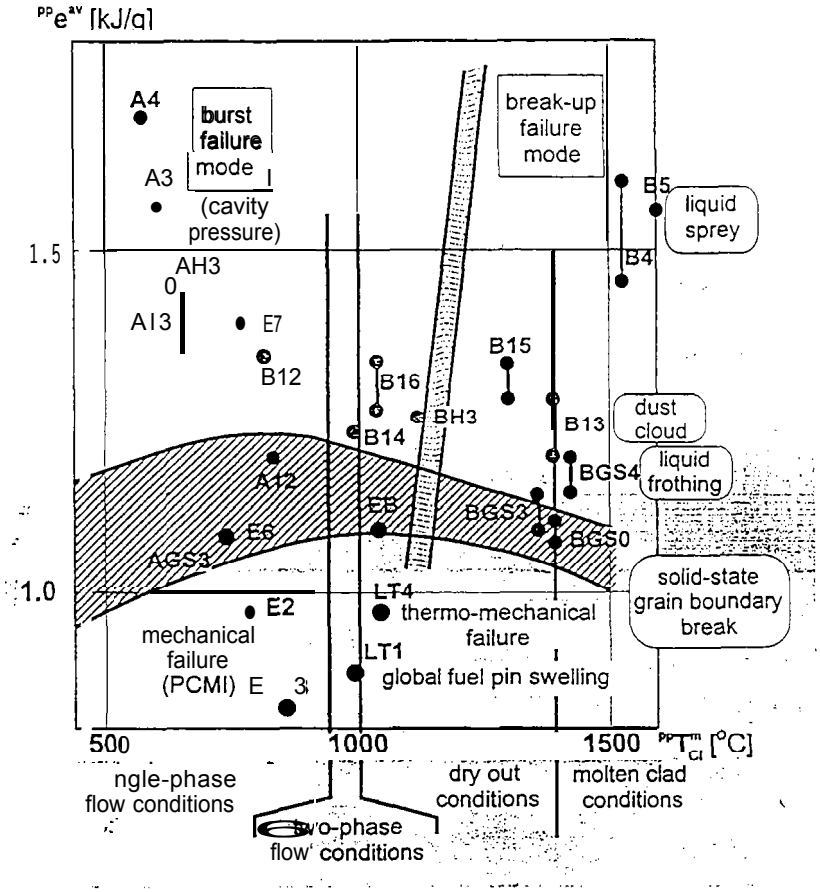

FIG.4. CABRI - related fuel pin destruction map fuel enthalpy and cladding midwall temperature at the PPN)

4.2.3. Failed pin behaviour

No fuel pin failure has been experienced in Japan.

4.3. Russia's Experience

4.3.1. MOX fuel assemblies testing in BOR-60, BN-350 and BN-600.

The onset of the pelletized MOX fuel irradiation dates back to the middle of the 1970s.The first stage of those activities was aimed at the experimental validation of the MOX fuel rods serviceability that was required for receiving permission to the MOX fuel assemblies irradiation in BN-350 and BN-600 reactors. The testing was carried out using prototype fuel rods under conditions similar to those of BN-350 and BN-600 reactors:

- burnup within 95- 135 MW·day/kgU;
- radiation dose within ~ 45-75 dpa;
- LHRG from 450 to 500 W/cm;
- maximum cladding temperature over the range of 690-7 10^{0}C (with overheating factors).

16 FAs were irradiated in BOR-60. All fuel rods were leak tight.

A maximum burnup and a dose for the first 10 FAs of the BN-350 reactor, loaded into the core in 1982, were restricted by a significant shape change of the wrappers due to swelling and radiation creep of structural materials (steel 16Cr – 11Ni –3Mo). A maximum burnup was over

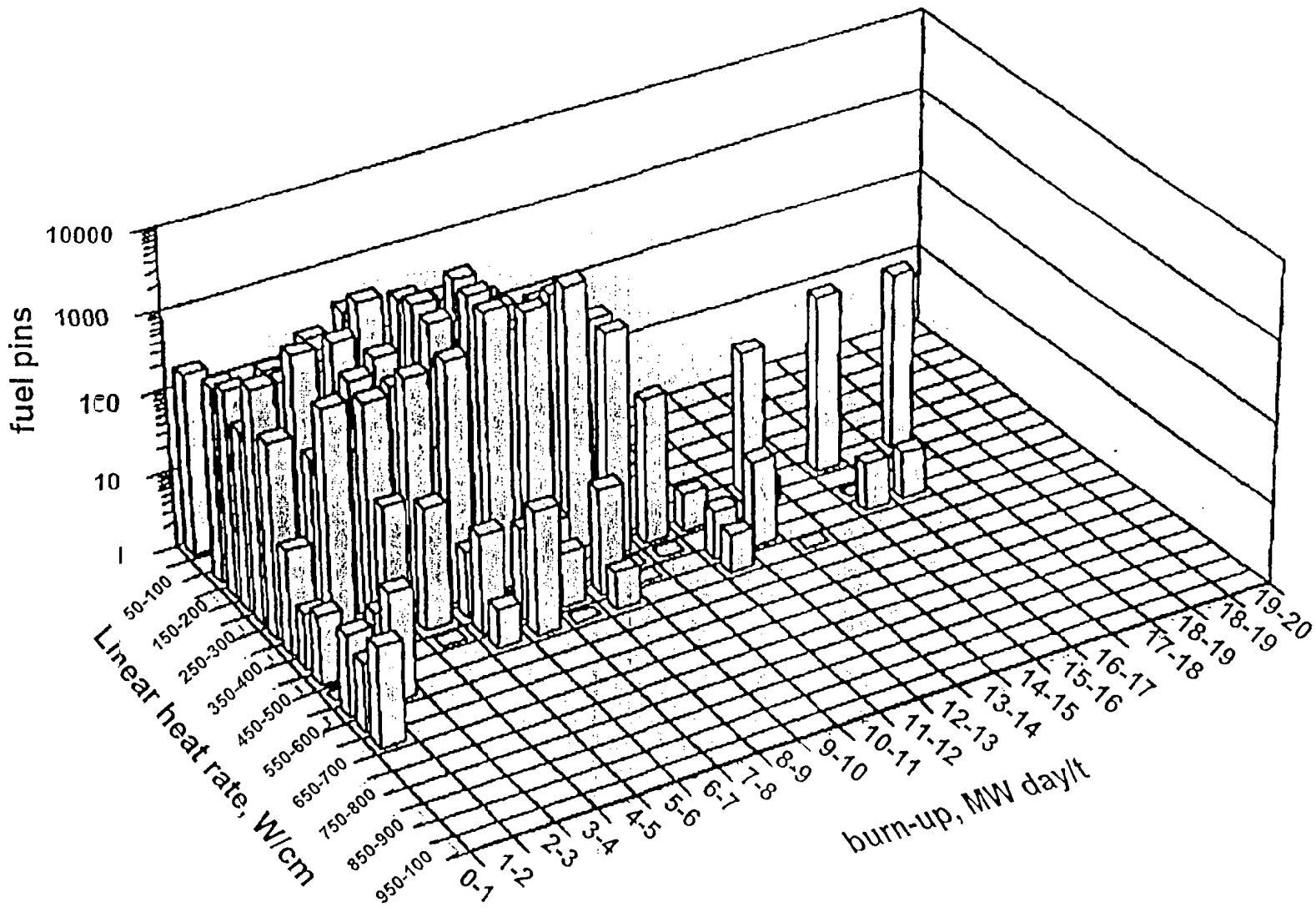

FIG.5. Irradiation achievements of Japanese fuels

the range of 4.5-5.0% h.a. and a maximum dose was within 40-45 dpa. Higher radiation resistance steels were used in the subsequent four FAs that allowed to achieve a burnup of ~ (10.5-l 1 .O)% h.a. (-100 MW·day/kgU). The fuel rods of all 14 experimental FAs in BN-350 were leak tight.

In fabricating MOX fuel assemblies for BOR-60 the wrapper structural material was changed. Steel of ferrito martensitic 13Cr – 2Mo–Nb-P-B type was used. The problem of wrapper shape changes was eliminated. A maximum burnup made 11.8% h.a. Maximum LHGR was 480 W/cm. Two of the fuel assemblies from that batch were subjected to examinations in the Beloyarskaya NPP's hot cell.

To manufacture fuel pellets (annular) for the fuel rods of the BOR-60, BN-350 and BN-600 reactors, a use was made of mixed oxide fuel that was prepared according to various technologies (mechanical blending of uranium and plutonium oxides, in accordance with „Granat" technology, plasmochemical conversion, „sol-gell" process) with a Pu content within 15-3 l%, a pellet density in the range of 10.2-10.7 g/cm^3 and oxygen factor from 1,95 to 2.00. A cladding diameter was 6.9x0.4 mm, a pellet outer diameter made 5.9 mm and the inner one was 1.7 mm. All rods with pelllettized fuel irradiated in reactor BN-600 are also leak tight (Table 5).

4.3.2. Materials science examinations of the BOR-60, BN-350 and BN-600 spent fuel rods has revealed the following:

- solid fission products are distributed in fuel for the most part proportionally to neutron fluence in the same way as in oxide uranium fuel , i.e., the migration of fuel and solid fission products through the fuel rod height is not observed;
- at a burnup higher than 3% h.a.: the fission products release from fuel under a cladding increases sharply. The fission products release from mixed and uranium oxide fuels is approximately identical and makes 95% relative to theoretical one at burnups higher than 10% h.a. (Fig.6);
- according to the metallographic examinations, the fuel column has a typical three-zone structure. A correlation among these zones is determined for the most part by the LHGR value. A gap between the fuel and cladding is filled with fission products. In the circumferential zone of the fuel bushings, manufactured from the mechanical oxide mixtures, UO_2 and PuO_2 appearances are observed. In the columnar zone such appearances are not available;
- in the mechanical mixture fuel at O/M ratio=2 it is observed the plutonium enrichment of the columnar crystals zone near the central hole (15-20% compared to the initial one). In the more homogenous initial fuel the plutonium enrichment achieves 2-7%. It depends upon the solid solution O/M value. The lower O/M value, the poorer plutonium enrichment of the bushings central part. The plutonium redistribution through the fuel length is not revealed;
- the oxygen chemical potential as an O/M function in fuel influences the depth of fuel-cladding chemical interaction. In the course of oxide fuel irradiation the radial oxygen redistribution through the fuel bushing takes place depending on the initial ON value (Fig.7). With increasing O/M in initial fuel and at a higher burnup, the oxygen potential grows (Fig.8) particularly at the bushings outer part that accelerates corrosion. The oxygen redistribution is not observed through the fuel rods active part. The radial oxygen redistribution takes place mainly in the core centre and above it at the most thermally intensive fuel areas;
- for the same steel grade the cladding corrosion depth increases at a higher burnup, temperature, LHGR and O/M in initial fuel. Matrix and inter-granular kinds of corrosion are observed. The compatibility of pelletized MOX fuel with the claddings, made of austenitic grade steels, is practically similar to that of uranium dioxide in the same irradiation modes. The influence of the fuel fabrication process and a plutonium content on the corrosion is not revealed.

4.3.3. In conclusion it is to be noted.

The results of the complex studies into pelletized mixed oxide fuels for BOR-60, BN-350 and BN-600 reactors allow to make a conclusion that the fuel rods serviceability resource

TABLE 5. PARAMETERS AND RESULTS OF EXPERIMENTAL FAs WITH PELLETIZED MOX FUEL IRRADIATION IN REACTOR BN-600

	Irradiation parameters			
	Max. LHGR, W/cm	Max. cladding temperature, °C	Max. burn-up, % h. a.	
15	48,2	695	IO,2	Leak tight
16	47,5	690	IO,2	Leak tight
17	48,5	695	IO,6	-"_
18	46,5	695	IO,2	-'I-
19	45,2	675	9.1	-"_
20	47,5	695	9.4	-"_
21	46	670	9,5	-"_
22	48,5	700	11,8	m"_
23	46,4	690	II,O	m"_
24	43,2	660	II,5	-"_
25	45,I	680	II,5	m"_
26	45,4	680	II,5	-"_
27-34	46-48	690-695	11.5	w"_

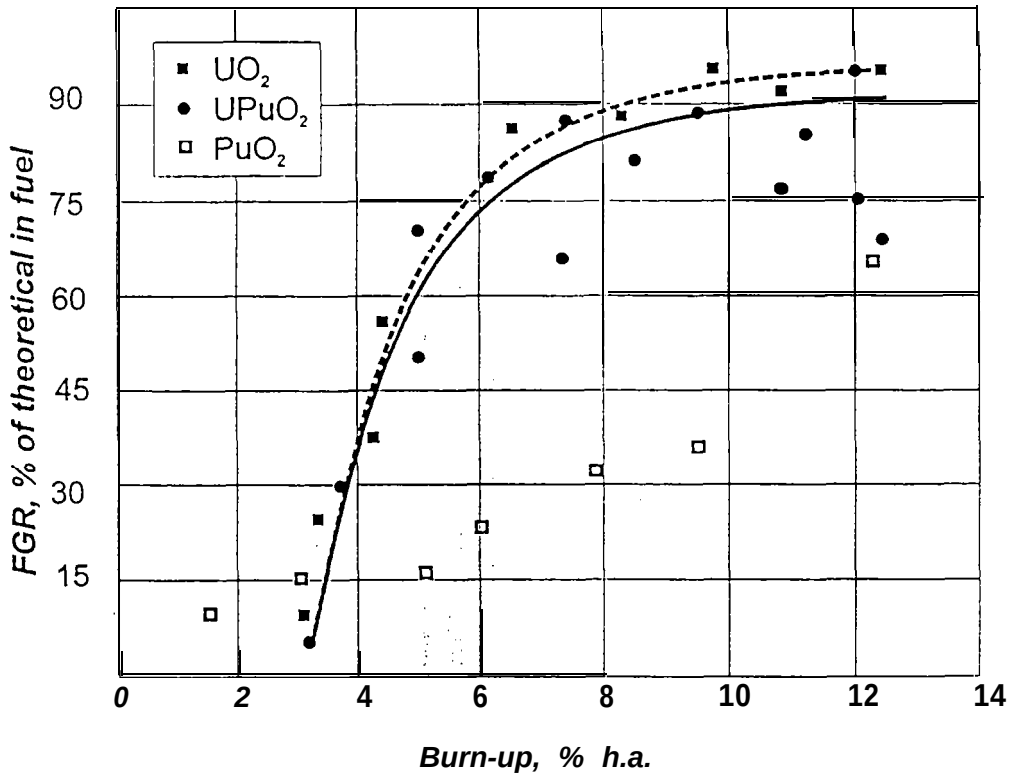

FIG.6. Fission gas release from U, U-Pu and Pu oxide fuel as a function of burn-up (BR-5, BR-10, BOR-60, BN-350)

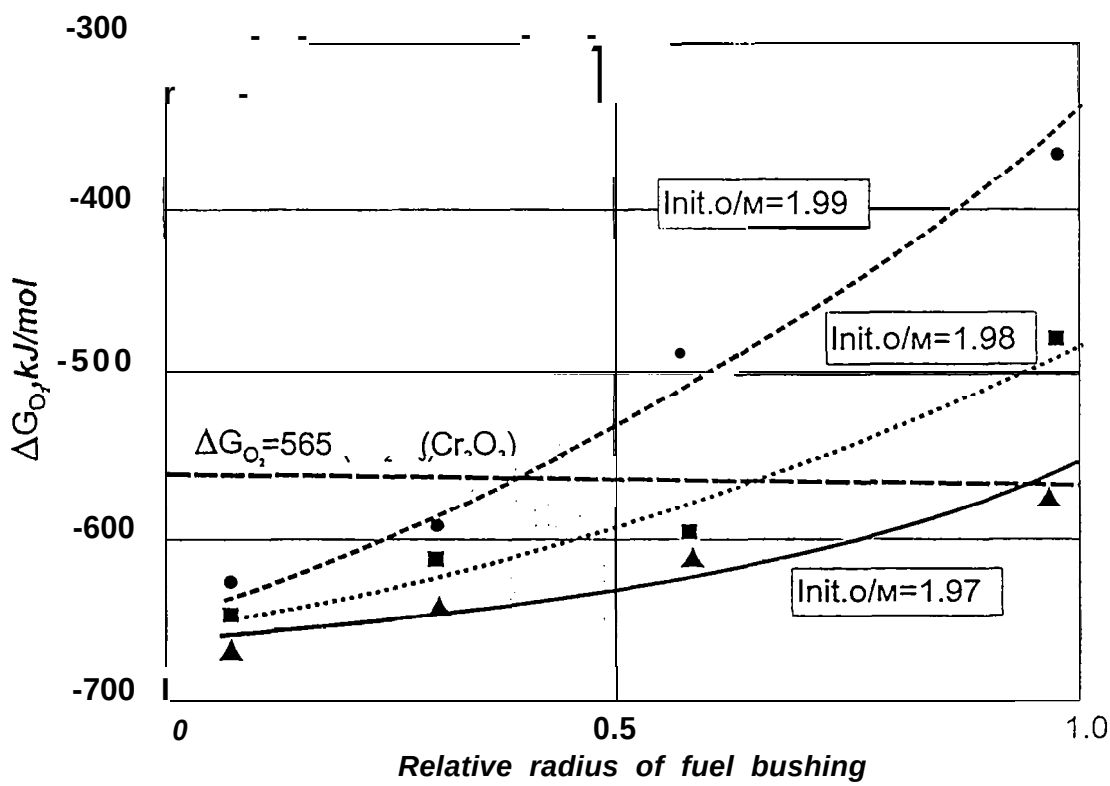

FIG.7. **Change in oxygen potential of U-Pu oxide fuel along** fuel bushing radius after irradiation in **BN-350**

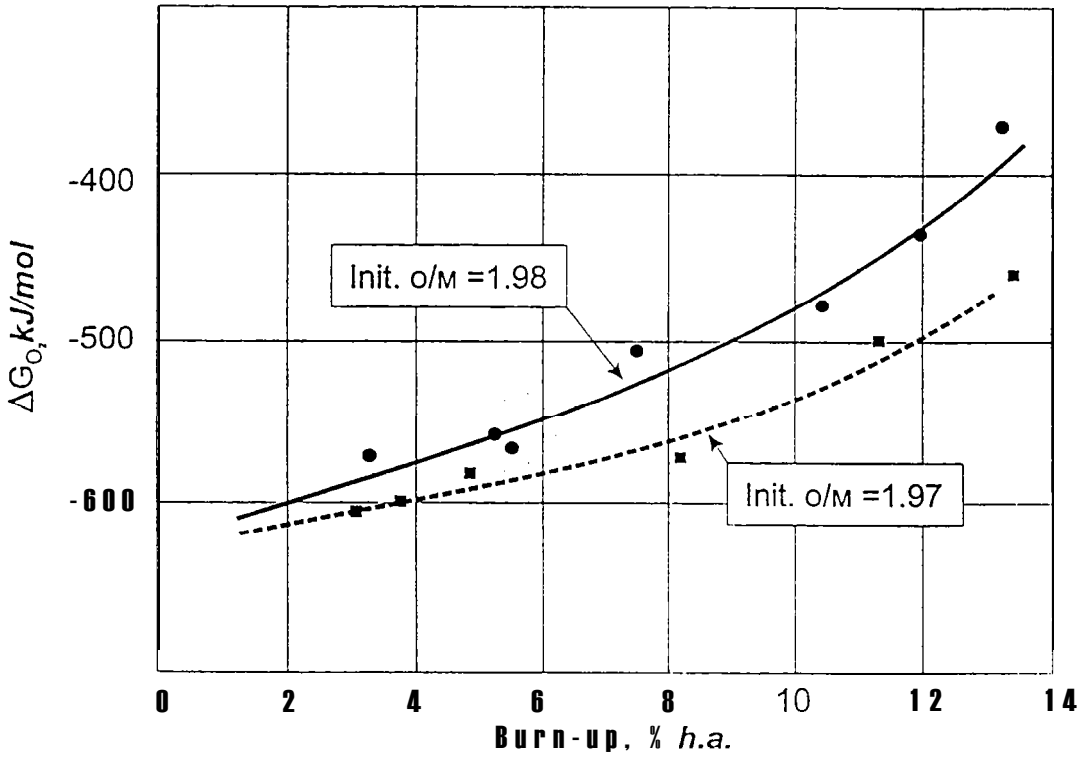

FIG.8. **Change of oxygen potential of U-Pu pelletized oxide fuel vs. initial stoichiometry (BOR-60)**

up to burnups of -14% h.a. has not been exhausted and the fuel rods retain their serviceability. With regard to many properties, mixed oxide fuel is similar to that of uranium oxide under irradiation. It is neccessary to enhance the requirements for the technology of preparing a mixed fuel solid solution by achieving its higher homogeneity and decreasing the O/M value in fuel. Leaky pelletized MOX fuel rods have not been revealed. Parameters and results of experimental FAs with pelletized MOX fuel irradiation in reactor BN-600 are presented in Table 5 as exsample.

4.3.4. The vibro MOX fuel was irradiated in BOR-60 (-450 FAs), BN-350 (2 FAs) and BN-600 (6 FAs) (Table 6).

In the irradiation of that fuel the loss of fuel rods tightness was observed in some cases (Table 6). The loss of tightness took place at the stage, when the vibro fuel parameters were not enough validated and optimized (efficient fuel density, getter content and oths.).

TABLE 6. IRRADIATION RESULTS OF FAs WITH VIBROFUEL RODS

	Fuel composition	Reactor	Irradiation parameters			Irradiation results
			Max. LHGR, W/cm	Max. cladding temperature, °C	Max. burn-up, % h. a.	
1	$(20\%Pu+80\%U)O_2$	BN-350	50,7	~700	4,7	Non-leak tight
2	-«-	BN-350	48,0	690	6,9	Non-leak tight
3	-«-	BN-600	40,3	691	6,8	Leak tight
4	-«-	BN-600	45,0	694	9,6	-"-
5	-«-	BN-600	45,0	683	9,3	-"-
6	-«-	BN-600	42,0	682	9,2	-"-
7	-«-	BN-600	44,0	665	9,0	-"-
8	-«-	BN-600	44,0	677	9,1	-"-

5. REPROCESSING

5.1. France's Experience

The interest of fast reactors is to allow the best utilisation of natural uranium by closing the cycle and so using the Pu formed from ^{238}U. So the ability to process the irradiated fuel is a key issue ; it was another positive feedback of the experience on European Fast Reactors. As indicated in table 4, all type of fuels except PFR were reprocessed in French plants ; and the Dounreay reprocessing plants successfully dealt with PFR fuel.

It should be noted from Table 7 that a coherent development was made in France for reactors, plant fabrication (see § 3.1) and reprocessing plants : AT1 for Rapsodie, APM for Phenix (a part of Phenix fuel was also reprocessed in the UP2 plant at La Hague by dilution with gas/graphite fuel).

The core of the process (extraction cycles) is the same for FR and PWR fuels ; The specificity for FR fuel comes from their geometry, the composition of clads, the high Pu content, the high burn-up. The dissolution is more difficult when the Pu concentration increases, and it appears desirable to remain under a Pu concentration of 30 % ; the behaviour of cladding material (passage of elements in solution) has repercussions on the down-stream process steps. Nevertheless the experience gained show that no insuperable problem was met.

In conclusion, the demonstration is made that reprocessing is possible, either in specific plants, either by dilution -dilution remains a possible solution for Superphenix fuel. Closure of the fast reactor fuel cycle has also been demonstrated, both in Phenix since 1980, and in PFR since 1982.

TABLE 7. PRODUCTION OF REPROCESSING FACILITIES FOR FR FUEL IN FRANCE

[redacted]	Reactor	$PuO_2/(U\ Pu)O_2$ %	Years	Tonnage (ML)
	Rapsodie	25 to 30	69 to 79	0.91
(La Hague)	Phenix	18	78 to 79	0.18
APM	Rapsodie	30	75 and 94-95	5.63
(Marcoule)	KNK1	enriched uranium	75 to 76	1.65
	KNK2	enriched uranium	92 to 94	1.45
	Phenix	enriched uranium	77 to 78	2.30
	Phenix core 1	18	79 and 83	6.74
	Phenix core 2	25	79 to 83	
	Phenix cores 1 and 2	18 and 25	88 to 93	8.85
HAO, (UP2 La Hague)	Phenix core 1	18	79 to 84	10.07
TOTAL				37.78

5.2. Japan's Experience

A basic research for the reprocessing of fast reactor spent fuel has been carried out since 1982 at CPF (Chemical Processing Facility) of JNC Tokai. The plutonium was recovered from the JOYO's spent fuel in 1984 at CPF.

The RETF (Recycle Equipment Test Facility) is under construction adjacent to CPF and intended for hot tests.

5.3. In Russia the MOX fuel, irradiated in BN-350 and BN-600, has not reprocessed yet. The enriched uranium fuel of fast reactors is being reprocessed.

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Invited Paper

OVERVIEW OF SAFEGUARDS ASPECTS RELATED TO MOX FUEL

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Abstract

Recent developments in the light of the IAEA verification requirements for MOX fuel at reactors and bulk handling facilities are discussed. Impact of the Additional Protocol and Integrated Safeguards System is briefly addressed. Agency's work undertaken with regard to the nuclear arms control and reduction is presented.

1. INTRODUCTION

International nuclear material safeguards consist of a complex control system based on nuclear material accountancy with the technical objective of providing for " . . . the timely detection of diversion of significant quantities of nuclear material from peaceful activities to the manufacture of nuclear weapons or of other nuclear explosive devices or for purposes unknown, and deterrence of such diversion by the risk of early detection" (para. 28, INFCIRC 153).

The uniform implementation of safeguards is maintained in all States with comprehensive safeguards agreements through application of technical implementation criteria which provide detailed requirements and procedures for how safeguards are to be implemented in any given circumstance. The intensity (i.e. the frequency and extent) of the IAEA's verification of nuclear material inventory is determined by the values assigned to technical implementation parameters such as a significant quantity and timeliness (ref: "timely detection"). The Agency has defined a "significant quantity" as the amount of a particular material (e.g. 8 kilograms of plutonium) as the amount of material that a State would need to make a nuclear explosive device. The detection time used in safeguards planning and evaluations is the maximum time that may elapse between diversion and its detection by the Agency safeguards. For direct-use nuclear materials -- such as unirradiated mixed oxide -- the detection time is specified as one month.

At the end of 1998 a total of 62.4 tonnes, equivalent to 7797 SQs, of unirradiated plutonium outside reactor cores was subject to IAEA safeguards. During 1998 fresh MOX was loaded into the cores of 14 reactors, and there were half a dozen plutonium conversion and MOX fabrication plants safeguarded by the Agency. In 1998 the Agency used ca 10000 Person-days of inspection effort of which one quarter was devoted for facilities using unirradiated plutonium. In order to reduce inspection costs the Agency has during recent years enhanced co-operation with the State or Regional Accounting Systems by introducing joint/common use of equipment and inspection procedures, coordinated R&D and training. However, both in Europe and in Japan the use of MOX LWRs is going to grow in coming years.

2. NEW METHODS AND TECHNOLOGIES

Over the years, the Agency has had to develop increasingly sophisticated verification methods to provide credible assurance that plutonium is not diverted from MOX conversion and fabrication facilities, and from reactors using MOX fuels. Some important accomplishments in this area are:

- the development of improved destructive analysis methods for laboratory analyses of all bulk forms of plutonium;
- the development of measurement procedures permitting independent on-site measurements of amounts of plutonium and plutonium isotopic ratios in feed materials, intermediate product materials, fuel rods and fuel assemblies, and in scrap and waste materials, through non-

destructive assay methods, including spontaneous fission assay methods by neutron coincidence and multiplicity measurements, isotopic ratios through high resolution gamma ray analysis, and calorimetric assay methods;

- the development of non-destructive assay methods for determining process hold-up within glove boxes and transfer equipment;
- the development of computer software for use during inspections to process operator inventory and flow information to expedite inspections, including the streamlining of the preparation of sample plans and the extrapolation of inspection findings from the use of random sampling.

During last decade there has been a steady increase in the number of unattended verification and monitoring systems which have resulted an increase in the verification coverage and at the same time reduce on-site safeguards inspections. The systems operate in unattended mode and combine surveillance, non-destructive assay techniques and or process monitoring devices. The advantages of such systems are: reduced inspection effort, 100% verification with reduced levels of intrusiveness to the operation of the nuclear facility and reduced radiation exposures to inspectors and plant operating staff. Such assay and monitoring techniques are applied at complex , and especially automated and remotely operated facilities.

More recently, a variety of advanced unattended systems for remote monitoring and transmission through telephone lines or other secured data transmission channels have been developed. The instruments used in such applications include digital surveillance cameras, electronic seals, and radiation monitors and sensors. Remote monitoring offers the possibility of replacing on-site activities which involve unattended assay and monitoring systems with data collection, review and evaluation at a remote location, in essentially real time. The use of remote monitoring is anticipated to be in connection with a reduced number of inspections, either announced or unannounced. Unannounced inspections have the potential to reduce inspection frequencies significantly when applied randomly on large group of similar facilities.

2.1. Safeguards of MOX Fuel at Reactors

The approach the Agency has taken is to concentrate the NDA verification activities to the fuel fabrication facilities by maintaining the continuity of knowledge on fresh MOX during shipment and storage at LWR reactors until the core loading using containment and surveillance techniques/l/. The MOX fuel is normally stored at LWRs in a wet storage; only at a few installations the elements are being placed to dry storages. The shipping containers are sealed by metal cap seals and/or VACOSS seals which are removed at the reactor by the IAEA inspector prior to placing fuel into the storage. Until core loading fresh MOX fuel is covered by surveillance where the Agency is currently introducing digital surveillance devices such as ALIS and DMOS to replace MIVS or Digiquad equipment.

Of particular importance is the requirement to verify the category change of fuel during core loading i.e. the downgrading from unirradiated to irradiated direct-use material. The current practice is to apply surveillance to confirm the transfer of each fuel assembly to the core and confirm that it has not been removed from there until the core is closed. There are two options to cover this : human surveillance with a presence of inspectors during the relevant operations and/or unattended video surveillance including underwater cameras with a recording frequency of the order of one minute per image. Whichever method is applied for verification the inspectorate needs to ascertain that all assemblies went into the core and none of them have been removed, concealed as a dummy or discharged as a spent fuel. Depending on the type of reactor the verification effort in the human surveillance option is 20-30 PDIs when the surveillance option typically requires 6-10 PDIs.

Now when authenticated digital surveillance systems such as DMOS come available the Agency can in future rely increasingly to the activities done by the SSAC such as the New Partnership Approach and/or to take the advantage of remote monitoring. The Agency has also acquired a number of CdTeZn

detector systems which can be used for the verification (gross defect) of fresh MOX under water without or with minimum movement of fuel assemblies /2/. For the partial defect measurement of fresh MOX fuel the Underwater Coincidence Counter is used.

Unattended monitoring system in other plutonium fueled reactors such as MONJU, JOY0 and FUGEN are based on GRAND electronics which are using neutron detectors on fuel transfer paths, connected to an electronic cabinet containing redundant data collection and computer software enabling data collection, retrieval and evaluation. The systems have uninterruptible power supplies to ensure reliability /3/.

2.2. Safeguards of MOX at Bulk Handling Facilities

The basic requirement is to verify PuO2 and MOX powder in the annual physical inventory taking with a high detection probability for gross, partial, and bias defects. For fuel rods, fuel assemblies, and other fuel items verification for gross and partial defects is required. Other bulk material such as waste needs to be verified fro gross defects. These requirements are also valid for the verification of receipts and shipments but to minimize the need of reverification the Agency uses intensively C/S measures such as seals to ensure the continuity of knowledge of nuclear materials. In addition, it is worth of noting that direct use material not verified should not exceed 0.3 SQ in physical inventory taking or 0.6 SQ in inspections for timely purposes. In particular with regard to the in-process inventories special arrangements need to be done.

Since early 1980's the High Level Neutron Coincidence Counting (HLNC) has been the main method used by the Agency for verification of plutonium bearing materials. The application of unattended assay and monitoring systems based on the HLNC techniques was first introduced in 1988 at PFPF in Japan and was considered a major advancement for reduction of inspection effort and simultaneously improved safeguards coverage. More recently advanced systems have been introduced in co-operation with Euratom at WBNP and WBFP in Belgium./4/

During last decade increasing amount of automatization has become a reality in the process lines and material storages. These developments restrict access to the process area, and consequently to nuclear material, and make also frequent clean out of glove boxes difficult and costly. To overcome these problems a number of systems for the measurement of plutonium contents in glove boxes such as the Glove Box Assay System (GBAS), Material Accountancy Glove Box System (MAGB) to verify plutonium in clean scrap, Plutonium Scrap Multiplicity Counter (PSMC) for dirty scrap have been developed. Waste Drum Assay System (WDAS) and Big Box Assay System (BBAS) for large containers have been introduced for the verification of plutonium in waste. In addition, procedures for other accountancy issues such as characterization of materials and nuclear decay (loss) corrections have been improved /5/. Near-Real-Time-Accountancy (NRTA) is a key part of the verification scheme at PFPF.

Recently solution monitoring system for plutonium nitrate has been introduced at PCDF in Japan /6/. The system is based on sensors introduced inside the tanks containing the nitrate solution which are then connected to electronic devices to process the signals and computers for collection and storage of data. The data are collected every few seconds and filtered to enable analysis by inspectors. The authentication of data is ensured by application of containment measures on the electronics cabinet containing electronic devices and data collection computers operated with authenticated software.

3. TOWARDS STRENGTHENED AND INTEGRATED SAFEGUARDS SYSTEM

In the past few years , the Agency has intensified its efforts to strengthen the effectiveness and improve the efficiency of the safeguards system including its efforts to enhance its capability for detecting undeclared nuclear material and activities. A number of strengthening measures endorsed by the Board were implemented by the Secretariat on the basis of the legal authority contained in existing safeguards agreements. In May 1997, the Board of Governors approved the text of a Model Protocol

additional to the safeguards agreements which, when implemented, will further increase the assurance of the compliance by States with their safeguards agreements provided by the safeguards system as a contribution to global nuclear non-proliferation objectives. To date, Additional Protocols covering 40 States have been signed.

The Secretariat is working on the integration of the traditional nuclear material accountancy-based safeguards measures implemented pursuant to safeguards agreements and on the information-based safeguards measures implemented pursuant to additional protocols. The aim of this Integrated Safeguards System is to optimize the combination of all the safeguards measures available to the Agency in order to achieve maximum effectiveness and efficiency in fulfilling the Agency's obligations under safeguards agreements. To do this, the system has needed to move beyond its focus on nuclear material accountancy - essentially a quantitative audit system designed to keep track of material declared to the Agency - to a system based on more qualitative assessments. This has entailed development in three major directions: more information, wider access to locations and greater use of advanced technology in areas such as remote monitoring and environmental sampling. In implementing this system, the Agency's objective is to achieve optimum effectiveness and efficiency by balancing fully the traditional nuclear material accountancy system with the strengthened measures. When confidence level grows with the new measures, alternative trade-off measures will be sought to reduce costs without sacrificing the over-all detection capability.

4. FUTURE PROSPECTS OF VERIFICATION

Thus far we have dealt with the development of the new integrated safeguards system and with the present verification responsibilities of the Agency. We would like to conclude the role of the IAEA safeguards by mentioning work undertaken with regard to the verification of nuclear arms control and reduction agreements. In addition to a complete ban on nuclear testing, two actions have always been as indispensable to nuclear arms reduction and nuclear disarmament: freezing the production of fissile materials for nuclear weapons purposes and the gradual reduction of stockpiles of such materials.

Last August, the United Nations Conference on Disarmament formed a Committee to negotiate a treaty prohibiting the production of fissile material for use in nuclear weapons or other nuclear explosive devices. According to the CD's rules and procedures, that Committee was disbanded at the end of their 1998 Session. When the Committee is re-established, the Agency is prepared to offer assistance, if requested, in developing the technical verification arrangements for the treaty.

In September 1996, the Agency, the United States of America and the Russian Federation agreed to investigate the technical, legal and financial issues associated with Agency verification of fissile materials in those States that had been removed from defense programs in conjunction with bilateral nuclear arms reduction treaties. Currently , the United States and the Russian Federation have each designated 50 tonnes of plutonium as no longer required for their respective defense purposes. Under the trilateral discussions, arrangements are being developed that would allow classified forms of fissile material to be submitted to Agency verification, pending steps later to convert the fissile material to unclassified forms and render it unsuitable for further use in nuclear weapons. As progress in the technical arrangements is made, a model verification agreement is being developed. It is anticipated that this initiative will lead to parallel bilateral fissile material verification agreements with the United States and the Russian Federation, not earlier than 2000.

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MOX FUEL FABRICATION

(Session II)

Invited Paper

OVERVIEW OF MOX FUEL FABRICATION ACHIEVEMENTS

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Abstract

Such overview having been adequately covered in an OECD/NEA publication providing the situation as of end 1994, this paper is mainly devoted to an update as of end 1998.

The Belgian plant, Belgonucleaire/Dessel, is now dedicated exclusively to the fabrication of MOX fuel and has operated consistently around its nameplate capacity (35tHM/a) through the 1990s involving a large variety of PWR and BWR fuels.

The two French plants have also achieved routine operation during the 1990s. CFCa, historically the largest FBR MOX fuel manufacturer, is utilizing the genuine COCA process for that type of fuel and the MIMAS process for LWR fuel: a nominal capacity (40 tHM/a) has been gradually approached. MELOX has operated at 100 tHM/a, as defined in the operating licence granted originally.

The British plant, MDF/Sellafield with 8tHM/a nameplate capacity is devoted to fuel and has manufactured several small fabrication campaigns.

In Japan, JNC operates three facilities located at Tokai: PFDF, devoted to basic research and fabrication of test fuels, PFFF/ATR line, for the fabrication of Fugen fuel and of corresponding fuel for the critical facility DCA, and PFPF for the fabrication of FBR fuel.

In Russia, fabrication techniques have been developed to fuel four BN-800 FBRs contemplated to be constructed and be fuelled with the civilian Pu stockpile. Two demonstration facilities Paket (Mayak) and R.IAR (Dimitrovgrad) fabricated respectively pellet and vipac type FBR MOX fuel for BR-5, BOR-60, BN-350 and BN-600.

The paper includes a brief description of each of the fabrication routes mentioned, as well as the production of respectively LWR and FBR MOX fuel in each fabrication facility, since the start-up of the plant, since 1 January 1993 and since 1 January 1998 up to 31 December 1998.

1. INTRODUCTION

This invited paper provides an overview on the historical evolution of MOX fuel fabrication and on the achievements resulting therefrom. It does not elaborate on lessons learned, on current developments and on today's perspectives, as these are the topics of the contributed papers. Such overview having been adequately covered in an OECD/NEA publication [1] and in a presentation at TOPFUEL' [2] and being complemented by several more limited scope presentations at various conferences [e.g. 3,4, 5 & 6], relevant parts of these publications are summarized herein. Since [1] presents the situation as of end 1994, since [2] deals only briefly with fabrication and since the other references quoted hereabove present the views of selected fuel fabricators, this invited paper is mainly devoted to a consensual update as of the end of 1998.

The paper deals with the fabrication of MOX fuels as well for LWRs (including therein the closely related ATR fuel) as FBRs. It is recognized that the two types of fuels have quite different characteristics impacting both on the fabrication process and on the quality requirements :

- the Pu content of FBR fuel is up to seven times higher than the Pu content of LWR fuel;
- having to operate at higher temperature and to higher burnups than LWR fuel, the smear density of FBR fuel has to be lower;
- the higher plasticity of FBR fuel, resulting from the higher irradiation temperature, justifies less restrictive specification tolerances and quality requirements than for LWR fuel;
- uniformity of Pu isotopic composition within a batch of FAs is a key performance related quality for LWR fuel (Pu 239 and Pu 241 being fissile, both with quite different reactivity values, and Pu 240 and Am 241 being neutron absorbers), while it is rather unimportant for FBR fuel (all the Pu and Am isotopes being fissile to some degree);
- the FR cladding is a steel for FBR fuels and a Zr alloy for LWR fuel.

Most industrial fabrication plants are therefore devoted exclusively to either LWR or FBR fuel. Dual purpose facilities have nevertheless operated successfully in the past (BN/Dessel, Siemens/Hanau) and dual purpose fabrication lines continue to operate (COGEMA/Cadarache) or are even contemplated for future plants (Russia).

The paper is restricted to operating fabrication plants and to facilities utilized as prototype for future industrial plants. The characteristics of those plants and facilities are briefly summarized, as well as the fabrication processes adopted. An update is presented on the actual fabrication records, on the fuel quality, on the safeguards implementation, on the personnel exposure and on the management of fabrication scraps and wastes. The challenges facing the fuel fabricators are briefly outlines, in order to provide a perspective for the contributed papers in this session. This overview paper is indeed complemented by the contributed papers dealing more particularly with the actions undertaken to progress further and with the advances and trends being implemented or contemplated.

2. OPERATING FABRICATION FACILITIES

Significant MOX fuel fabrication activities, as a preparation for industrial **deployments, were** conducted since the 1950s in Belgium and the USA, since the 1960s in Germany, France, the UK, Japan and Russia, and since the 1970s in India.

In the **USA,** five pilot facilities, with a cumulated fabrication capacity of 50 to 70 tHM per year, were in operation in 1976, when President Carter took the political decision to indefinitely defer reprocessing. As a result, the facilities were shutdown and decommissioned. Most of this valuable experience is by now obsolete.

In **Germany,** the **Siemens/Hanau** plant, which started operation in 1972, as a dual purpose (FBR and LWR) facility, reached an effective capacity of 20 to 25 tHM per year of LWR fuel in the 1987-1991 period. It was shutdown, as a result of a contamination incident on 19 June 1991, by order of the Hesse Ministry of Environment. This plant is now being decommissioned. Since the operation experience is recent enough to still have some relevance, further mention of this plant will be included in this paper, whenever appropriate. A larger plant (120 tHM per year), constructed on the same site and almost ready to start operation, never received operation licence and is now abandoned.

The **BN/Dessel** plant started operation in 1973, on the basis of R&D conducted over the previous fourteen years in successive facilities from laboratory to pilot scale and pursued during the first few years of operation of the plant. During an initial period of operation (10 years), the plant was equipped to fabricate as well FBR as LWR fuel and did indeed fabricate both types of fuel. Based on lessons learned, the plant was temporarily shutdown and refurbished in 1984 and the capacity was upgraded to 35 tHM per year for LWR fuel, the only fuel to be manufactured thereafter. Since the mid-1990s, the plant is being backfitted, without interrupting fabrication, to incorporate improvements resulting from accumulated experience and deemed necessary to meet more challenging future requirements. During the first ten years of operation, the end-product of the plant was LWR and FBR FAs. Since the mid- 198Os, the end-product is FRs and assembling is performed in the adjacent FBFC Int'l/Dessel U fuel manufacturing plant, to benefit from their large scale production (800 to 900 FAs per year). The FBFC Int'l plant is equipped with a dedicated MOX fuel assembling, control and storage facility devoted to the LWR FRs produced by BN/Dessel and CFCa (COGEMA/Cadarache). Their experience dates back to 1963, with the manufacture of the first PWR MOX FA incorporating FRs fabricated by BN.

The **CFCa** plant is the ultimate achievement of a facility, which started operation on a pilot scale in 1962 and was devoted to FBR fuels. In the mid-1980s with EDF's decision to utilize MOX fuel in their PWRs, one of the FBR lines was converted to LWR fuel fabrication and started operation on PWR fuel in 1990. More recently, the second fabrication line, up to then still devoted to FBR fuel, has been modified into a dual purpose facility, capable of fabricating either FBR or LWR fuel. The resulting capacity is 30 tHM per year for LWR fuel if no FBR fuel is being fabricated. The mounting of the FAs is performed within CFCa for FBR fuel and at FBFC Int'l for LWR fuel.

Construction of the **MELOX/Marcoule** plant was decided in 1985, as a consequence of EDF's decision to load MOX fuel in 16 of their 900 MWe PWRs. The MOX operation started in 1995. The plant was originally to be devoted to this purpose and take over the fabrications for EDF conducted at BN/Dessel and COGEMA/Cadarache during the interim period of construction of MELOX. In this perspective, the plant was licensed for a production of 100 tHM per year. The mono-product destination of this plant enabled to introduce more automation than in the BN/Dessel and FBFC Int'l plants, which served as technology basis, and to achieve an extremely low fabrication cost. As originally equipped, MELOX had a nameplate capacity of 120 tHM per year. It has been boosted to 160 tHM per year, on a three-shift basis, by incorporation of additional equipment (a.o. a third sintering furnace). With the addition of a West Fitting Building, the capabilities of MELOX are

being extended to BWR fuels and the production rate could reach 250 tHM per year in the 2000s. Details about these current developments are provided in a contributed paper to this session [7].

MDF/Sellafield, with a current nameplate capacity of 8 tHM per year, is a refitting, for LWR fuel, of a facility which produced FBR fuel between 1970 and 1988. The purpose is to get manufacturing experience of MOX fuel in preparation for SMP/Sellafield, a plant with a nameplate capacity of 120 tHM per year, which is ready to operate, but has not yet received the licence to start U commissioning, while the plant was initially scheduled to start operation in 1997 and has been ready to begin U commissioning since April 1998.

In **India,** a pilot scale fabrication plant is in operation at Tarapur, producing now only BWR fuel, but foreseen to be adapted later to produce CANDU and FBR fuel.

After conducting development for some years at laboratory scale in PFDF, still in operation, JNC started **PFFF** with two completely separated lines, one for ATR fuel (1972), still in operation, and one for FBR fuel (1973), shutdown in 1987. The latter was indeed replaced, in 1988, by **PFPF, a** fully automated plant, with a fabrication capacity adequate to fuel the Joyo and Monju FBRs. After a few years of operation, PFPF production has been temporarily suspended in 1998 the adequate stockpile of reload fuel for Joyo and Monju (which has not resumed operation since the 1995 sodium leak in the secondary heat transport system) provided the opportunity to performed planned maintenance and refitting of the facility on the basis of lessons learned.

In Russia, three small scale facilities started operation in the 1970s for FBR fuel : the Grana/Chelyabinsk lab-scale facility and the Paket/Chelyabinsk small scale facility, both fabricating pellet fuels and the NIIAR/Dimitrovgrad integrated reprocessing-refabrication facility, producing Vipac fuel. The last two facilities are being upgraded from lab-scale to pilot scale. Three contributed papers in this session provide more details on the evolution of those facilities.

Table 1 summarizes the main characteristics of the commercial MOX manufacturing facilities functioning today. The seven operators of those facilities have an aggregate experience of some 150 years MOX fuel fabrication, to which must be added the valuable technology background of the now defunct Siemens MOX operations and of the lab-scale and pilot scale facilities predecessors to the facilities listed in Table 1. The available capacity for LWR (+ ATR) fuel is some 200 tHM per year, which is inadequate to serve the customers' demand for moxification of their separated Pu : it is the main cause of the increasing Pu stockpile over the past decade. The available fabrication capacity for FBR fuel is over 20 tHM per year. It is redundant in the European Union and in Japan, since the three operating FBRs would require only some 5 tHM per year if they were operating steadily at full power and furthermore their lifetime load factor has been below 40%. Under their present operation licence, the two Russian FBRs, BOR-60 and BN-600, require 0.4 - 0.5 tHM per year for their refuelling, quite steadily, since BN-600 has exhibited a high lifetime load factor (74%) : it can be accommodated by the existing fabrication facilities. If BN-600 becomes licensed to operate with an hybrid core, the requirements would become 1.0 – 1.5 tHM per year, still achievable in the current Russian fabrication facilities.

3. FABRICATION PROCESSES

The various fabrication processes developed historically and the ones applied currently are well described in some detail in [I].

3.1. Conventional process

The conventional fabrication route is a direct application of the most common industrial fabrication process for U fuel, the enrichment of U being replaced by a mechanical blending of the feed powders : UO_2 and PuO_2 (or $U\text{-}PuO_2$ for the Pu delivered by the JNC reprocessing plant). As the blended powder is not free-flowing and therefore inadequate to feed the pelletizing press, the

powder is preconditioned by precompaction, in a slugging press, followed by granulation, the granules being obtained by crushing the slugs. The challenge in this process is to obtain a uniform distribution of the Pu in the product. It is not so much a problem for FBR fuel in which the ratio of UOZ to PuO_2 powder is 2 to 3. But it is a difficulty for LWR (and ATR) fuel, the UO_2 to PuO_2 powder ratio being 10 to 60. When the feed powder is (U-50% Pu)O_2, as is most common for the JNC plants, achievement of a uniform distribution of the Pu in the product is eased, since the UO_2 to (U-SO% Pu)O_2 powder ratio is 0.5 to 1 for FBR fuels and 5 to 30 for LWR (and ATR) fuels.

Fig. 1 illustrates a schematic flowsheet of such conventional fabrication process. It represents the process applied by JNC in PFFF and PFPF and, approximately, by Mayak in PAKET. While JNC most commonly starts from co-denitrated (U-50% Pu)O_2, PAKET is fed by PuO_2, as all other current MOX fuel manufacturing plants. After preparation of the MOX powder, all the processes are similar, with minor specificities. Pelletizing is carried out in hydraulic presses : only Siemens/Hanau had implemented rotary presses in their new large plant. Sintering is most commonly conducted in continuous furnaces, except in PFFF equipped with batch furnaces and in PFPF, where contingency batch furnaces have been installed, after experiencing problems with the automatic transfer mechanism of the continuous furnaces installed originally. Center-less grinding is now performed dry, which, in principle, does not require subsequently drying of the ground pellets. However, JNC is still including a drying-degassing step before quality control (QC) of the pellets. TIG welding is commonly adopted, both for the seam and seal welds of the end plug. Only Siemens/Hanau was using resistance welding, which can be performed under pressure and therefore did not necessitate a venthole machining in the end plug and a separate seal weld. For FBR FAs, the assembling operation starts, in most cases, with fitting an helical spacing wire to the rods and involves the introduction of a bundle of FRs into the FA shroud. For LWR FAs, the FRs are positioned in magazines and drawn from the magazines through the FA skeleton. For ATR FAs, individual FRs are directly loaded into the FA skeleton.

3.2. Powder processing

A particular example of conventional processes is provided by the elaborate process currently applied by JNC in PFPF (Fig. 2). The additives utilized include a binder, to improve granulation, a poreformer, to achieve the low density specified for FBR fuel, and the lubricant, as universally used to optimize pelletizing. Excellent quality fuel can be fabricated by the current process, which has however the inconvenience to require de-gassing the green pellets prior to sintering and is expensive, due to the several process steps involved. In this respect, JNC has begun the development of a short process which omits the homogeneous blending and granulation steps, in a cost-reduction perspective [8].

The **BARK** plant is also utilizing the conventional process, with the particularity that the blending and ball milling operations are conducted in a single attritor mill, as initiated by BNFL-UKAEA.

A simplification of the conventional feed powder processing route (which is to MOX fuel what the enrichment operation is to U fuel) was developed in the 1970s by CEA and applied in the Cadarache fabrication plant under the name **COCA.** It is based on the use of an optimized ball mill acting as blender and of a forced extrusion of the lubricated micronized powder through a sieve, resulting in free-flowing granules adequate for feeding the pelletizing press (Fig. 2). This process, originally developed for FBR fuel, has been adapted for LWR fuel and utilized from 1989 to 1994 for manufacturing MOX fuel for EDF's PWRs, but later abandoned in favour of the MIMAS process now currently applied for LWR fuels in this plant. Amongst the reasons are some inconveniences of the COCA process for manufacturing LWR fuel, the evidence that COCA fuel did not exhibit an improved irradiation behaviour compared to MIMAS fuel and the effort and expenses required to accumulate a database on COCA LWR fuel to the same level as the already existing and still expanding database on MIMAS fuel. As a result, the COCA process is now considered only for FBR

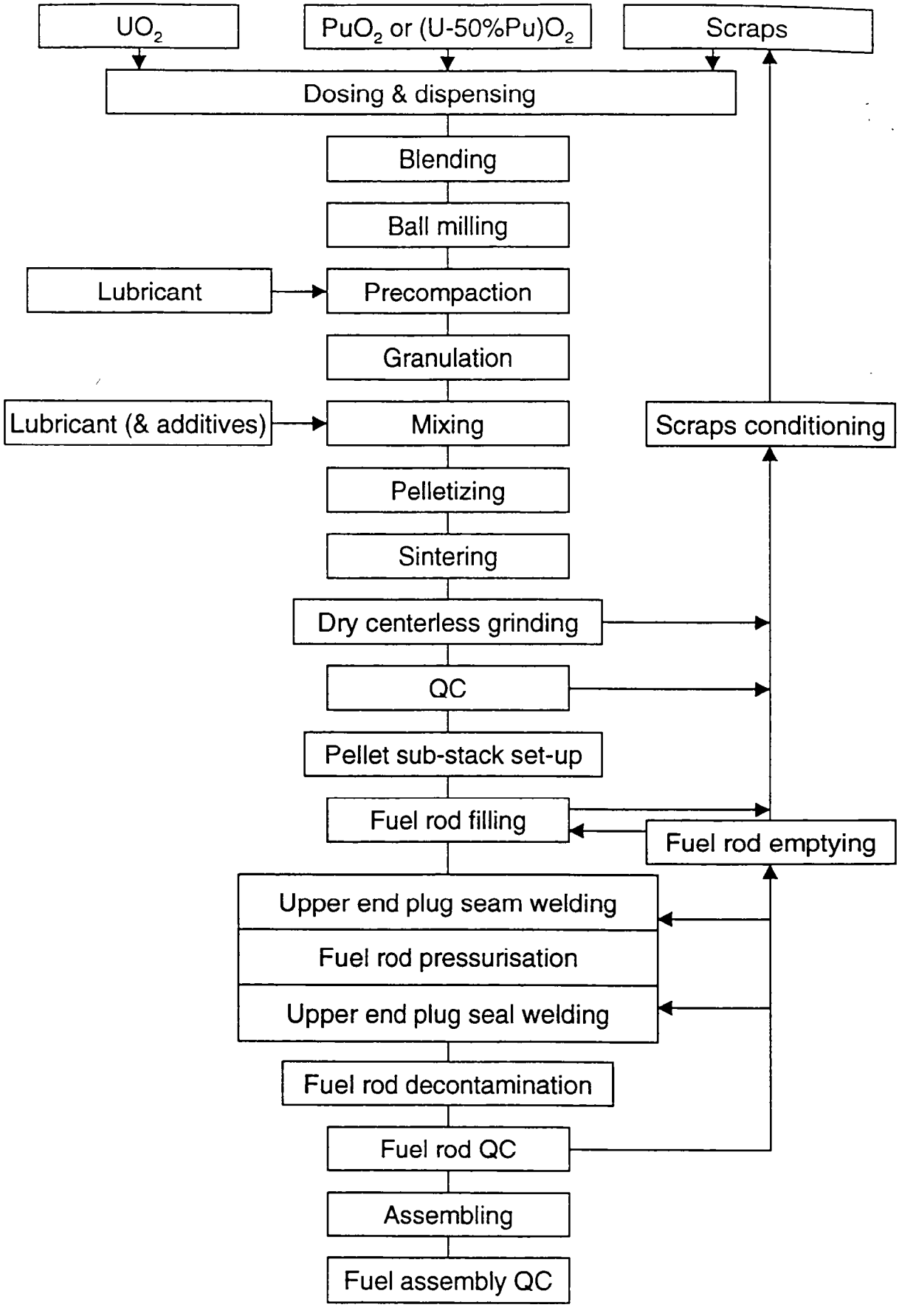

Fig. 1. Flowsheet of a conventional fabrication process

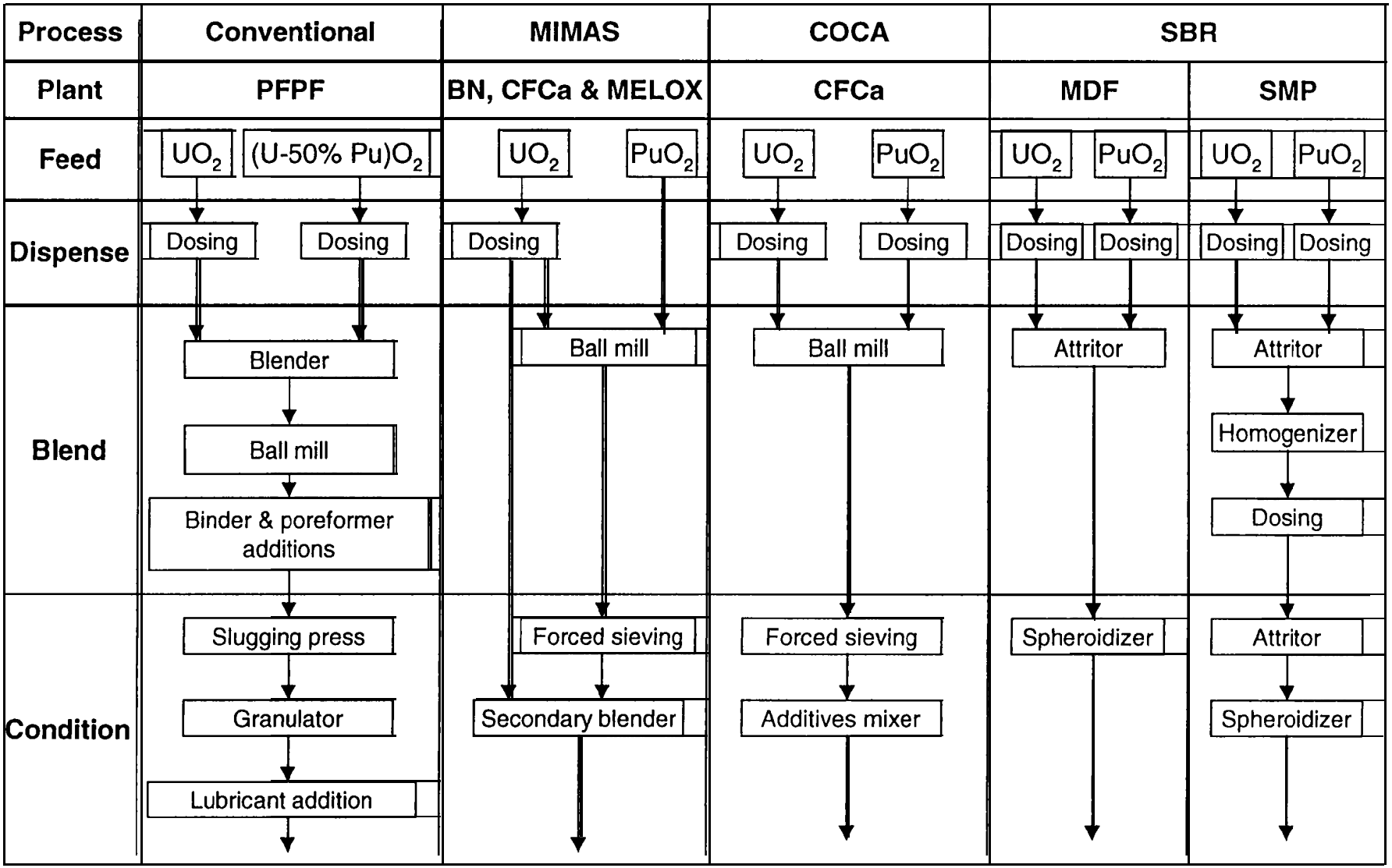

Fig. 2. Schematic flow sheets of the powder processing routes

(in these simplified flow sheets, the incorporation of additives and the scrap recycle are not represented)

fuels and selected in this respect for the contemplated DEMOX fabrication plant, designed by a joint French-German-Russian team for dispositionning of Russian W-Pu.

An original development was conducted in the 1980s by BNFL-UKAEA and resulted in the Short Binderless Route, **SBR,** based on the application of alternative process equipment for the blending and granulation functions (Fig. 2). The traditional ball mill is replaced by an attritor, an off-the-shelf mill widely used n the pharmaceutical industry and renowned for providing intimated blends of constituents in a short processing time. The precompaction-granulation equipment is replaced by a spheroidizer, working on the powder agglomeration process invented by CEN.SCK in the 1970s for the fabrication of fuel kernels, the pit of coated particles fuelling HTRs. The name SBR reflects the two genuine characteristics of the process :

- the binder additive, incorporated in ancient nuclear fuel manufacturing routes, is not used : as a result, a de-waxing step of the green pellets prior to sintering is not necessary and the process is in this respect similar to modem U fuel fabrication processes ;
- the processing time and the flowsheet are short and the equipment can be stacked, so that the powder be discharged by gravity from the feed dispensing and dosing glove-box through the processing equipment into the hopper of the pelletizing press.

The simple sequence of one attritor mill and one spheroidizer, utilized in MDF, has been sophisticated for SMP [8], by the addition of one homogenizer and one attritor mill (Fig. 2) main purpose is to increase the size of a powder lot from 50 kg MOX, the load of an attritor and of a spheroidizer, to 150 kg MOX, the load of the homogenizer, with the benefit of reducing the number of QCs and obtaining a larger quantity of fuel with uniform Pu isotopic composition.

The **MIMAS** process invented by BN in the early 1980s is an adaptation of the BN reference fabrication process developed earlier and applied commercially through the 1970s in the Dessel plant. The reference process consisted in a single blending of PuO_2 powder with free-flowing UO_2 powder, resulting in a blend of adequate flowability to feed the pelletizing press. This extremely short powder processing route provided fuel of adequate quality. The interdiffusion of Pu and U during sintering was not preventing the presence of high Pu content agglomerates, which size was smaller than the specified maximum “Pu particle” size defined on the basis of RIA behaviour experience. When the reprocessor decided that MOX fuel had to be soluble in a pure nitric acid solution, the reference fuel was not acceptable anymore : like pure PuO_2, MOX material with a Pu content higher than 40 – 50 % Pu can only be dissolved in nitric acid with an oxidizing additive, e.g. hydrofluoric acid. To meet this new specification, the pure PuO_2 feed to the blender was replaced by a co-micronized mix of UO_2-(typically) 30% $Pu0_2$, which is the principle of the MIMAS process (Fig. 2). Not only does the intimate contact between the co-micronized UO_2 and PuO_2 provide for adequate interdiffusion during sintering and therefore solubility, but larger contact area between the more abundant fine powder and the free-flowing UO_2 powder results in a less heterogeneous MOX structure than in the earlier reference process. In parallel, Alkem (subsequently part of Siemens) had developed the sibling OCOM process, with the same success in fabricability and improved fuel behaviour.

The process being developed at **ERC/NIIAR** by the radiochemistry department starts with spent fuel and produced Vipac fuel. It is based on molten-salt dissolution in a “chlorator-electroiyser” made of pyrolitic graphite, the electrorefined UO_2 and/or MOX is deposited on the cathode as a loose crust that is crushed and sized to produce the adequate size fractions fed into the fuel rod by vibro-compaction. More details on the process can be found elsewhere [e.g. IO]. Although further development is being pursued, the facility has already produced fuel [11].

3.3. Fabrication technology

Beside the general outline provided in section 3.1. and the specificities of powder preparation routes described in section 3.2., each fabrication plant is characterized by genuine processing

approaches, influenced by their design bases and licensing limits. In this respect one can mention, amongst others, the minimum Pu-240 content of the Pu to be processed, its maximum Am content, the maximum percentage of Pu in the MOX fuel and the maximum allowed personnel exposure, which are provided, as illustration, in Table 2 and infuence the equipment adopted and the plant lay-out.

The **BN/Dessel** plant has developed the original MIMAS process and has fitted it to the requirements of its 40 tHM/yr industrial operation, nl. :

- achieving an almost uniform isotopic composition within a fabrication campaign, by computerized selection of the feeding sequence of PuO_2 supply cans to the process, within the available PuO_2 feed stock, guaranteeing thereby the energetic equivalence of all the FAs throughout their utilization ;
- automatizing the sophisticated ball mill to provide for an homogeneous and uniform distribution of the PuO_2 in the 60 kg MIMAS master blends ;
- maximizing the proportion of scrap that will be dry recycled in the process, up to 18% (i.e. an equivalent of half of the master blend) ;
- optimizing the secondary blender to obtain a uniform distribution of the master blend in the free-flowing UO_2 and selecting the capacity of the blender (80 kg) as a compromise between simplification of product traceability and minimization of the scraps and waste arisings, taking due consideration of the required plant flexibility;
- in general, conceiving all the equipments and their size be easily serviceable in line with the objectives of the plant : producing fuel of a large variety of specifications in rather small fabrication campaigns (typically 4 to 23 tHM, each comprising 3 to 6 different fuel compositions and/or FR types). This optimization of the equipment results in reduced inter-campaign loss of capacity and minimized scraps and waste arisings;
- adopting a FR filling and welding unit with minimum intrusion of the cladding into the glove-box, to minimize FR surface contamination, decontamination and contamination monitoring.

CFCa, which had culminated as the largest FBR fuel plant with two dedicated fabrication lines, has acquired and implemented the BN/Dessel LWR fuel technology to launch its industrial LWR operations, while, at the same time, testing automated transfer and QC devices for MELOX. The adjacent CEA Pu facilities provide for scientific research into the process parameters, resulting in an opportunity to continuously improve fabrication, complementing the large trial-and-error database acquired by the past industrial operation of the MIMAS process.

The **MELOX** plant is the first large scale LWR fuel facility. The MIMAS process, as implemented at BN/Dessel and CFCa, was adapted to the specific objectives set for the plant to fabricate a mono-product, nl. single specification MOMX fuel for the set of identical EDF PWRs, in large fabrication campaigns (up to 65 tHM, consisting each of only three discreet Pu contents. In this frame :

- complete automation has been implemented from the selection and opening of the PuO_2 canisters to the emptying of programmatically chosen individual PuO_2 cans ;
- the ball mill has been developed to a 150(?) kg capacity category, providing the possibility to use up to 50 % pellet scraps as ingredient and resulting in the same excellent homogeneity and uniformity of PuO_2 distribution in the MIMAS master blend as the original 60 kg capacity ball mills ;
- a high capacity (670 kg) secondary blender, consisting in a conical screw mixer with a double envelope air cooling system has been adopted ;
- the filling, welding and decontamination of the FRs has been enclosed in one single glove-box, containing essentially the same industrial equipment as used for U fuel in the FBFC plants ;
- the FA manufacture, QC and handling have been fully automated ;

. the waste is minimized through sorting and treating the waste and irrecoverable scrap in a dedicated building of the MELOX site, in the liquid effluent treatment unit of COGEMA Marcoule and in the centralized UCD and URP facilities at COGEMA La Hague.

As its name indicates, the MDF was conceived as a test bench for the SBR concept and not as an industrial fabrication facility : the emphasis was to put it rapidly into operation, rather than to optimize the layout. Attention was mainly exercized on the demonstration of six genuine features :

. the vertically integrated attritor-spheroidizer system ;
. the transport system of pellets : pick-and-place of the green pellets in the sintering boats and cushion transfer ;
. the laser QC of pellets ;
. the loading system of pellets into the FRs ;
. the B-impregnated bakelized wood panels for gamma and neutron shielding ;
. the elaboration of all the QC procedures and techniques required for commercial fuel.

Being housed in a building previously devoted to the development of FBR fuel and using some equipment of this previous activity impacts on the operability and capacity (8 tHM/yr) of this facility as well as on the dose rate to the personnel, while however actually producing commercial fuel. Its purpose of providing information for the design and construction of SMP has been fulfilled. Although elaborating on SMP is beyond the scope of this paper devoted to fabrication achievements, it is worth mentioning how the technology demonstrated in MDF has been adapted to fit in a large scale industrial plant, on the basis of lessons learned :

- in the feed powders receipt and dispense unit installed on top of each of the two identical powder processing columns a dual PuO_2 feed has been provided : it allows metered aliquots of Pu of two different isotopic compositions to be incorporated in a MOX powder lot to uniformize the isotopic composition within a fabrication campaign;

- an homogenizer and a second stage attritor mill has been added (Fig. 2) to bulk together three 50 kg sub-lots from the first stage mill, with the advantage of constituting 150 kg powder lots. The equipment and process parameters of this new processing column have been developed in a full scale mockup, operating on UO_2 and located on the BNFL Springfields premises ;
- the four continuous furnaces are capable of operating at up to 1750 C ;
- each of the two identical FR fabrication and inspection lines is fit for both PWR and BWR fuel;
- FA facilities consist of one automatic PWR assembling and inspection line and one automatic BWR inspection line;
- the plant is operated from a control panel using virtual reality to ensure that it is ergonomically correct;
- an Export facility has been built adjacent to SMP, recognizing the storage and packaging needs specific to BNFL's customers.

The **BARC** facility, devoted to the development and demonstration of an industrial fabrication technology, will not be described in detail here, as it is the topic of a contributed paper in this conference [11].

Similarly, the **PFFF** and **PFPF** plants are described each in a contributed paper [12 & 13]. PFPF has raised the fabrication technology to a very high degree of automation : even maintenance and repairs of equipment within the fabrication line is foreseen to be performed by dedicated robots enclosed in each glove-box. The problems encountered (a.o. the historical high level of Pu hold-up, the jamming of the sintering furnace, etc...) and the experience resulting therefrom are invaluable for progressing in a proper selection of MOX fabrication plant design philosophy and appropriate technology.

The **Russian** facilities, to which four papers are devoted in this conference, are developing technology for future industrial fabrication plants and, in this context, producing demonstration quantities of MOX fuel. It will define the technologies and equipment appropriate for future deployment.

4. FABRICATION RECORDS

Tables 3 and 4 provide, respectively for LWR and FBR fuels, the productions of the facilities since start of operation until 3 l December 1998. The definitively shut down Siemens/Hanau facility and the FBR line of PFFF have been incorporated as a recognition to their significant contribution to MOX fuel industrialization. Small additional quantities of fuel have been fabricated in the 1960s by operators of these fabrication plants in earlier laboratory (and sometimes demonstration) facilities : BN (at Mol), Siemens (at Karlsruhe) and BNFL (at Risley). In the tables and throughout this section are only included the deliveries accepted by the customers : additional quantities still to be accepted by the customer have, of course, been fabricated.

Although the fabrication experience of both types of fuel are equivalent FR-and FA-wise, the quantity of fuel produced and of Pu processed is larger for LWR fuel than for FBR fuel and the LWR experience is more contemporary.

The fuels mentioned in Tables 3 and 4 are for a large variety of NPPs in twelve countries (Table 5).

The experience over the past decade encompasses a broad range of characteristics (Table 6), covering the full range of industrial MOX fuel of current design. Particularly important is to notice that the personnel exposure is decreasing or stabilizing over the years, notwithstanding the generally increasing radioactivity of the Pu being processed and of the Pu contents of the fuel being fabricated. Indeed, while Pu issued from reprocessing GCR fuel was a common feed in the 1980s almost all the feed is now Pu issued from reprocessing of LWR fuel, with quite higher specific gamma and neutron activities (Table 7). The benefits of automation can be illustrated by the past experience of JNC : 324 mSv/tHM in the FBR line of PFFF in 1987 (fabrication of Joyo fuel) and 161 mSv/tHM in PFPF in 1993 (fabrication of Monju fuel).

5. FUEL QUALITY

As is evidenced in a quite large number of publications by as well fabricators as customers, the fuel produced today meets the specification requirements and is of quality equivalent to top grade U fuels. A good illustration is the burnup achieved by commercial MOX fuels in NPPs (Table 8). It is beyond the scope of this paper to overview all the quality attributes of MOX fuels, FRs and FAs. Only two characteristics, differentially approached and achieved by each genuine fabrication route, will be reviewed : the homogeneity of Pu distribution in the MOX fuel and the uniformity of Pu isotopic composition within a fabrication campaign.

5.1. Homogeneity of Pu distribution

An homogeneous distribution of the Pu within the fuel serves four purposes :

- impart to MOX fuel the same resistance to RIA failures as U fuel. This historically first requirement resulted from the SPERT power burst experiment conducted in the 1960s and concluding that a fissile particle of adequate size could pierce the cladding below the fuel failure limit of homogeneous fuel (170 cal/g radially averaged fuel enthalpy). On these bases, a maximum Pu-rich agglomerate size is specified, generally corresponding to a pure PuO_2

TABLE I – CURRENT CHARACTERISTICS OF OPERATING MOX FUEL FABRICATION FACILITIES

COUNTRY	FACILITY	OPERATOR	START OF OPERATION	CAPACITY tHM / yr	FEED	PRODUCT	PROCESS
BELGIUM	BN/Dessel	BELGONUCLEAIRE	1973	35	PuO_2	LWR FRs	MIMAS
	FBFC Int'l	FBFC	1987 [a]	120-200 [b]	FRs	LWR FAs	Assembling
FRANCE	CFCa	COGEMA	1962	10	PuO_2	FBR FAs	COCA
			1989	35 [c]	PuO_2	PWR FRs	MIMAS
	MELOX	COGEMA	1995	100 [d]	PuO_2	PWR FAs	MIMAS
UK	MDF	BNFL	1994	8	PuO_2	PWR FRs	SBR
INDIA	Tarapur	BARC	1994	18	PuO_2	BWR FAs	Conventional [e]
JAPAN	PFFF	JNC	1972	10'	$(U\text{-}50\%Pu)O_2$	A T R FAs	Conventional
	PFPF	JNC	1988	5 [g]	$(U\text{-}50\%Pu)O_2$	FBR FAs	Conventional
RUSSIA	Paket	Mayak	1986	0.3	PuO_2	FBR FAs	Conventional
	ERC	RIAR	1981	1	spent fuel	FBR FAs	Vipac

a new part of an U fuel fabrication plant in operation since 1963
b 120 tHM/yr (700 FAs/yr) if only BWR ; 200 tHM/yr (440 FAs/yr) if only PWR.
c capacity if no FBR fuel is being fabricated
d capacity restricted by licensing
e under « conventional » are meant pellet fabrication processes based on compaction-granulation of the feed powders, as is used in most U fuel fabrication plants
f the licence is based on Pu processed annually, nl 850 kg Pu I yr
9 the licence is based on Pu processed annually, n12.5 t Pu / yr

particle of 100 μm diameter. Although more recent NSRR tests conducted by JAERI have failed to reveal any influence of Pu-rich particles on the power excursion failure threshold, the MOX specifications continue to include a maximum Pu-rich particle size limit : modern fuel fabrication technologies can easily meet this specification item.

- minimize power peaks resulting from Pu mal-distribution. While the pellet-to-pellet enrichment is practically constant in U fuel, the Pu content of pellets varies in MOX fuel, as a result of mechanical blending of the constituents. The power peaks resulting therefrom penalize the admissible power ratings of MOX fuel. As a result a pellet level homogeneity of Pu distribution is included in the specifications. We will call it “macro-homogeneity”, to distinguish it from the “micro-homogeneity”.

- insure solubility in pure nitric acid solutions. As indicated previously, this demand was raised in the early 1980s by the industrial reprocessors. As MOX crystallographic lattices containing less than 40-50 % are soluble, the solubility criterion requires that the Pu content be below the solubility threshold in the individual grains and that the pellet structure affords adequate access of the acid to the grains. This Pu distribution attribute will be called “micro-homogeneity”.

- minimize fission gas release (FGR) and therefrom resulting rod internal pressure (RIP). MOX fuel unavoidably releases more fission gas than equivalent U fuel, due mainly to the difference in the radial distribution of power within the pellet as burnup proceeds and to the usually higher power ratings of MOX fuel late in life. If Pu is mal-distributed in the pellet, resulting in Pu-rich agglomerates of a size larger than fission spike lengths, a saturation effect at the locally very high burnup spots results in an additional increase of FGR. This effect, adequately predicted and quantified by FR modelling codes, has been observed in early heterogeneous MOX fuel. It is part of the micro-heterogeneity features.

The **macro-homogeneity** depends on the sophistication of the blending technology. Each fuel fabricator has devoted a large effort to optimizing its blending equipment and procedure. All other things being equal, the fabrication routes involving a progressive dilution of PuO_2 into UO_2 can more easily achieve macro-homogeneity than the processes mixing directly PuO_2 and UO_2 to the final required composition. Examples of such progressive blending processes are the MIMAS process, with the intermediate master blend, and the JNC process, with the co-denitrated $(U\text{-}50\%Pu)O_2$ feed.

TABLE 2 -DESIGN BASIS AND LICENSING LIMITS

PLANT	MIN. %Pu 240/Pu	MAX. %Am/Pu	MAX. %Pu/HM	MAX.EXPOSURE [a] mSv/yr
BELGONUCLEAIRE	0	1.0 [d]	Unlimited [c]	5
CFCa	17		Unlimited [c]	5
MELOX	17	3.0 [e]	12.5	5
MDF	18	3.6 [e]	6.5 [f]	5
SMP	17	3.0 [b]	15 [f]	5
PFFF nd	[g]	1.0	5.7	15
PFPF	nd	3.0	32	15

a Design and internal limitations. The licensing limit is set by ICRP at 50 mSv/yr
b Average per fabrication campaign. Higher values are admissible, but Impact on the production capacity
c FBFC licence for FAs restricted to 10% Pu / HM
d for individual PuO_2 delivery cans upon anival at the facility
e for individual PuO_2 delivery cans at the time of feeding the fabrication line
f % Pu fiss. / HM
g minimum not defined by licensing

TABLE 3 – LWR FUEL FABRICATION RECORDS AS OF 31 DECEMBER 1998 (ROUNDED FIGURES)

FACILITY	SINCE FACILITY START-UP				1993 – 1998	1998
	t HM	t Pu [a]	FRs	FAs	t HM	t HM
BELGONUCLEAIRE	400	20	210 000	1 200 [b]	210	38
Siemens/Hanau	158	6.4	77 000	380		
CFCa	180	11	90 000	340 [b]	150	36
MELOX	238	13	144 000	619 [c]	238	101
MDF	14 [d]	1	7 300	36	14	na [e]
BARC	1	0.0	290	8	1	na
PFFF [f]	110	1.7	20 000	695	24	0.5
TOTAL	1 100	53	550 000	3 300	640	180

a contained in the delivered fuel

b mainly manufactured at FBFC

c Includes 92 FAs Incorooratina pellets and FRs fabricated at CFCa

d the philosophy is to make diffierent fuel designs (Table 5) to support business in SMP

e data not available

f ATR fuel

TABLE 4 – FBR FUEL FABRICATION RECORDS AS OF 31 DECEMBER 1998 (ROUNDED FIGURES)

FACILITY	SINCE FACILITY START-UP				1993 – 1998	1998
	t HM	t Pu	FRs	FAs	t HM	t HM
BELGONUCLEAIRE	4.2	1.3	14 000	70		
Siemens/Hanau [a]	5.9	1.9	26 000	100		
CFCa	11Ø	22	430 000	2 300	0.2	0
Sellafield [a]	13	2.6	98 ØØØ	3ØØ		
PFFF	4	1.1	43ØØØ	375		
PFPF	IO	2.5	63 000	401	5	0
Paket	2	0.4	4 800	38	na [b]	na
RIAR	Ø.7	Ø.3	na	31Ø	na	na
TOTAL	15Ø	32	68ØØØ	39ØØ	6	negl. [c]

a now being decommissioned

b data not available

c less than 1 tHM

The **micro-homogeneity** depends on the blending and sintering technologies. In this respect, the single step blending fabrication routes are better suited to approach perfection. The picturesque achievements of the SBR process are a good example. Through proper optimization of the powder processing and sintering steps, the MIMAS process, which produces genetically micro-heterogeneous fuel, has been improved, so that now a negligible amount of the Pu is in agglomerates of sufficient size to enhance FGR and that the interpretation of irradiation results shows the Pu particle size effect

TABLE 5 – NPPs FOR WHICH THE FUEL WAS FABRICATED
(STATUS 31 DECEMBER 1998)

	PLANT	[a]	BN	SIEMENS	CFCA	MELOX	PFFF	PFPF	OTHERS [b]
BE	BR 3	P	x [c]						
	Doel 3	P	+ [d]						
	Tihange 2	P	+						
CA	NPD	H	x						
CH	Beznau 1	P	+						M
	Beznau 2	P	+	+					M
	Gösgen	P	+						
DE	Grafenrheinfeld	P	+	+	+				
	Isar 2	P			+				
	Philippsburg 2	P	+	+	+				
	Obrigheim	P		+	+				
	Neckarwestheim 1	P		+					
	Neckarwestheim 2	P			+				
	MZFR	H		x					
	KNK	F	x	x					
	Lingen	B		x					
	Brokdorf	P	+	+					
	Unterweser	P	+	+	+				M
	Grohnde	P		+	+				
	Gundremmingen A	B		+					
	Gundremmingen B	B	+	+					
	Gundremmingen C	B	+						
	SNR	F	x	x					
	VAK	B		x					
FR	CNA	P	x	x					
	Blayais 1	P				+			
	Blayais 2	P	+		+	+			
	Chinon B 4	P				+			
	Dampierre 1	P	+		+	+			
	Dampierre 2	P	+		+	+			
	Dampierre 3	P				+			
	Dampierre 4	P				+			
	Gravelines 1	P				+			
	Gravelines 2	P				+			
	Gravelines 3	P	+		+	+			
	Gravelines 4	P	+		+	+			
	Phenix	F	+		+				
	St. Laurent B1	P	+		+	+			
	St. Laurent B2	P	+		+	+			
	Superphenix	F			x				
	Tricastin 1	P			+	+			
	Tricastin 2	P			+	+			
	Tricastin 3	P			+	+			
	Tricastin 4	P				+			

TABLE 6 – NPPS FOR WHICH THE FUEL WAS FABRICATED (CONT'D)

	PLANT	[a]	BN	SIEMENS	CFCA	MELOX	PFFF	PFPF	OTHERS [b]
GB	DFR [e]	F	x						
	PFR [e]	F	x		x				
IN	Tarapur 1	B							T
	Tarapur 2	B							T
IT	Garigliano	B	x						
JP	Fugen	A					+		
	Joyo	F					+	+	
	Monju	F						+	
	Takahama 4 [f]	P							M
	Fukushima1-3 [g]	B	+						
KZ	BN-350	F							P+R
NL	Dodewaard	B	x						
RU	BOR-60	F							R
	BN-600	F							P+R
SE	Oskarshamn 1	B	+						
TOTAL	PWR + PHWR	36	18	9	17	16	-	-	4 (M)
	BWR + ATR	12	6	4	-	-	1	-	2 (T)
	FBR	11	4	2	3	-	1	2	2 (P) + 3 (R)

a A = ATR ; B = BWR ; F = FBR ; H = PHWR ; P = PWR
b M = MDF ; P = Paket ; R = ERC/RIAR ; T = BARC
c x reactor now shutdown
d + reactor still operating (not necessarily with MOX fuel)
e the majority of the fuel was fabricated in a now shutdown BNFL facility
f and / or Takahama 3
g and / or Kashiwazaki Kariwa 3

TABLE 7 – EXPERIENCED RANGE OF LWR FUEL FABRICATION CHARACTERISTICS OVER THE PAST DECADE

CHARACTERISTICS	BN	CFCA	MELOX	MDF	PFFF
Fuel types	6	5	1	4	1
tHM / campaign	4 – 29	4.6 – 15	33 – 90	1.3 - 4	1.5 – 8.7
Max. % Am / Pu [a]	2.0	1.0	1.5	0.6	0.7
Nber of Pu contents / campaign	3 – 6	3	3	3	2
Max. % Pu / HM	8.4	6.6	6.7	8.6	2.4
Cladding types	6	4	1	4	1
Collective doses [b]	130 (1986)	72 (1994)	20 (1996)	nr [c] (1994)	22 (1981)
mSv/tHM (year)	40 (1994)	34 (1997)	12 (1997)	47 (1997)	23 (1994)
	29 (1998)	30 (1998)	11 (1998)	nr (1998)	nr (1998)

a mean value of a fabrication campaign
b includes the plant personnel and the external contractors
c not representative due to the limited quantity of fuel fabricated.

TABLE 8 – RELATIVE RADIOACTIVITY OF Pu FROM DIFFERENT ORIGINS . (REPRESENTATIVE VALUES)

PU ORIGIN	GCR	PWR	BWR
Gamma	1.0	2.2	2.5
Neutron	1.0	2.9	3.8

to be indiscernible. As for the fuel solubility objective of micro-homogeneity, all the fuels are now confronted by a proper optimization of the fuel micro-structure to meet the specification criterion ; the Pu distribution achieved in the current industrial fabrication processes is not a factor anymore.

5.2. Uniformity of Pu isotopic composition

PuO_2 is produced by the reprocessing plants in batches of typically 90 to 100 kg Pu and conditioned in cans of 2.7 to 3.2 (COGEMA) or 5 to 7 (BNFL) kg Pu for delivery to the fabrication plants. While PuO_2 precipitation and finishing is a batch operation at La Hague, resulting in strictly identical isotopic composition of the Pu throughout the batch, it is a continuous process at Sellafield, resulting in a "rainbow" transition affecting the first and the last cans of each PuO_2 batch. Depending on the size of the MOX fuel fabrication campaign and on the average Pu content, anywhere from 3 to 4 PuO_2 batches are involved in one fabrication campaign.

The variability of Pu isotopic composition amongst the PuO_2 batches to be incorporated in a fabrication campaign depends on the panoply of types (PWR, BWR, AGR, Magnox) and burnups (first cores to extend burnup reloads) of fuel having been reprocessed. With the progressive exhaustion of first cores being reprocessed, the variability of Pu isotopic composition is progressively diminishing, but still much too large to be neglected, since it critically affects the MOX fuel design and performance. To compensate for the variability of Pu isotopic composition, BN initiated a Pu equivalence formulation, enabling to correct the Pu contents of each fabrication campaign and of each MOX fuel batch within a fabrication campaign, relative to the design basis Pu contents. This approach has been adopted by all fuel designers and is now universally applied, except when NPP licence limits require to consider (also) the fissile Pu content. For FBR fuel, variability of Pu isotopic composition corrected by application of an adequate equivalence formula does not result in deterioration of the fuel quality. For LWR fuel, whatever the sophistication of the equivalence formula, the fuel performances are affected by non-uniformity of the Pu isotopic composition within a MOX fuel fabrication campaign. For instance, in a PWR, a power peak up to 6 % can result from a variability of only 2 % Pu-239 in the isotopic composition, even when the Pu contents are adjusted through a sophisticated equivalence formulation [14]. It illustrates the importance of approaching uniformity of Pu isotopic composition within a fabrication campaign of LWR fuel.

In the BN/Dessel plant, it is achieved by computerized selection of the 5 to 6 La Hague cans being incorporated in the ball mill to produce one master blend. It results in a good uniformization of the Pu isotopic composition amongst all the master blends within a fabrication campaign (for which 3 to 15 PuO_2 batches are provided by the customer). If this single uniformization step were insufficient, the MIMAS process provides intrinsically the possibility of feeding the secondary blender with different master-blends, providing an additional uniformization step. The isotopic composition uniformization is equally achieved in the LWR fuel fabricated at CFCa; since the same MIMAS process and the same ball mill are utilized.

The ball mill capacity being larger in the MELOX plant (150 kg), 13 to 15 La Hague cans are incorporated in each master blend, resulting in an even more perfect uniformity of Pu isotopic composition amongst all the primary blends prepared for one fabrication campaign.

In SMP, uniformization of the Pu isotopic composition is being approached by an elaborated receipt and dispense unit, enabling to load in each attritor mill charge optimized aliquots of two different PuO_2 batches. The subsequent homogenization of three primary attritor charges into a 150 kg MOX powder lot, further improves the uniformity of Pu isotopic composition. Since each MOX powder lot contains the equivalent of less than two BNFL PuO_2 cans, the dosing of appropriate aliquots of two of the several (typically 9 to 30) PuO_2 feed lots incorporated into a fabrication campaign constitute a significant step towards homogeneity of the Pu isotopic composition.

The other MOX fabrication facilities do not provide for a specific opportunity to cross-blend the PuO_2, or co-denitrated (U-50 %Pu)O_2 feed batches within the MOX manufacturing process.

6. SAFEGUARDS

The BN/Dessel plant has the longest past history of safeguards inspections, having been granted on IAEA facility attachment in 1978 and having been through the dual IAEA and Euratom inspection regime, before implementation of the combined Euratom-IAEA inspection regime, now ruled by the so-called “New Partnership Approach”.

The current safeguards situation of the MOX fabrication plants is given in Table 9. Additionally, all plants are, of course, also under the safeguards jurisdiction of the responsible agency of their national government.

An overview of the safeguards aspects [15] and two examples of safeguards applications [16 & 17] are presented in this symposium.

7. SCRAPS AND WASTES

Scraps are generated by the process itself (e.g. centerless grinding fines or sludges), by the rejects (e.g. non conforming pellets) and by the surpluses fabricated within a fabrication campaign before switching to the next fabrication campaign (in-line contingency inventories of MOX powder, pellets and FRs). When the campaign size is small compared to the fabrication capacity of the plant (or dedicated fabrication line in the plant), and the fabrication process is well under control, the surpluses constitute a majority of the scrap arisings. As the fabrication campaigns are frequently relatively small, adequate management of the scraps is an important consideration having economic (fabrication cost) and environmental (personnel exposure and waste generation) impacts. In this perspective, all the MOX fuel manufacturers have developed and/or are elaborating technologies (therein including scrap conditioning) to recycle into the process lo-15% scraps without prejudicing MOX fuel quality. With their long experience of the MIMAS process, BN/Dessel can now operate with up to 18 % of the feed constituted of scrap. Similarly, the MIMAS process in MELOX can accommodate 50 % rejected pellets in the master blend : this corresponds to approximately 10 % of the final blend. Trials made by BNFL in the full scale Springfields mock-up of the SMP powder preparation tower have provided the SBR process with a technology to incorporate conditioned scrap and a trial in MDF has been successful.

Waste arisings originating from plant operation and maintenance, as well as waste due to originate from plant backfitting and ultimately decommissioning have also received proper attention. Indeed, waste management influences fabrication costs, personnel exposure, licensability and public acceptance. At the BN/Dessel plant, by identifying and optimizing the waste generating operations and by educating the personnel, Pu contained in the waste has been reduced to less than 0.1 percent

TABLE 9 – PEAK FUEL ASSEMBLY AVERAGE BURNUP (GWd/tHM) REACHED BY COMMERCIAL MOX FUEL, AS OF 31 DECEMBER 1998

PLANT	PWR	BWR	ATR	FBR
BELGONUCLEAIRE	51	30	-	-
CFCa	38	-	-	100
MELOX	32 [a]	-	-	-
MDF	39	-	-	220 [b]
Tarapur	-	17 [a]	-	-
PFFF	-	-	38	102
PFPF	-	-	-	61 [a]
Paket	-	-	-	114
ERC / RIAR	-	-	-	150

a not any fuel discharged yet
b from a BNFL facility now being decommissioned

TABLE 10 – SAFEGUARDS INSPECTION OF THE FABRICATION FACILITIES

	INSPECTED BY	EURATOM	IAEA [a]	
Belgium	BELGONUCLEAIRE	yes	A	(1978)
	FBFC Int'l	yes	C	(1990) [b]
France	CFCa	yes	no	
	MELOX	yes	C	(1999)
UK	MDF	yes	C	(1998)
India	Tarapur	-	no	
Japan	PFFF	-	A	(1979)
	PFPF	-	A	(1988)
Russia	Paket	-	no	
	ERC/RIAR	-	no	

a A facility having been granted a full scope facility attachment
C facility designated by IAEA to Euratom for safeguards with IAEA control limited to the shipping area
( ) date at which the facility attachment was granted or the designation notified

b the original facility attachment, granted in 1980, is applicable only to U fuel

of the Pu contained in the delivered MOX fuel [1], notwithstanding the policy of not stripping Pu from the waste : it illustrates accomplishments achievable by feedback from lessons learned. At MELOX, licensing authorization imposes to reduce the radioactivity releases and the Pu wastage to almost zero : as mentioned in Section 3.3., dedicated facilities have been commissioned at the MELOX/Marcoule site and at La Hague, to achieve this target. Beside this effort of all the manufacturing plants to minimize Pu in the waste streams, improvement programmes are also pursued to reduce the volumes of each radioactive waste category.

8. CHALLENGES

In line with the evolution observed over the past few years, MOX fuel fabrication will be confronted with increasingly demanding and difficult targets and conditions.

The evolution to more radioactive Pu feeds will continue, with the processing of aged Pu stockpile and the separation of Pu from higher burnup U fuels. It can be contemplated that the incentive to reduce personnel exposure will also persist.

With the degraded Pu isotopic compositions and the higher design burnups of MOX fuels, the Pu contents will continue to increase, potentially impacting on personnel exposure and on quality of MOX fuel, which might have to be fabricated to tighter specifications accounting for the more severe irradiation duties.

The safeguards will be strengthened with the enforcement of IAEA's 93+2 Programme, resulting in additional constraints.

The waste minimization objective will continue to be pursued and, in spite of this context, the fabrication costs will have to be mastered, as the continuing decline of uranium prices and the fierce competition amongst U fuel fabricators affect the MOX fuel competitivity.

9. CONCLUSION

The MOX fuel manufacturing industry has reached maturity as a result of the long operating experience of some fabrication plants. The large scale MELOX and PFPF plants are examples of extension of existing technologies into more advanced and larger facilities.

The lessons learned from experience have been instrumental not only in designing and starting up the new facilities, but also in backfitting the plants which had operated since a couple of decades.

In the next millennium, the industry will be confronted to additional challenges, which will require the same progressive improvements as experience will progress.

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MELOX FUEL FABRICATION PLANT: OPERATIONAL FEEDBACK AND FUTURE PROSPECTS

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Abstract

As of December 1, 1998, 32 Europeans LWRS are loaded with MOX fuel. It clearly means that plutonium recycling in MOX fuels is a mature industry, with successful operational experience in fabrication plants in some European countries, especially in France.

Indeed, the recycling of plutonium generated in LWRs is one of the objectives of the full Reprocessing - Conditioning - Recycling (RCR) strategy chosen by France in the 70's. The most impressive results of this strategy, is the fact that 31 of the 32 reactors are loaded with MOX fuels supplied by the COGEMA Group from the same efficient fabrication process, the MIMAS process, improved for the MELOX plant to become the A-MIMAS process.

In France, 17 reactors are already loaded and 11 additional reactors are technically suited to do so. Indeed, the EDF MOX program plans to use MOX in 28 of its 57 reactors. An EDF 900 Mwe reactor core contains 157 assemblies of 264 rods each. 52 fuel assemblies per year are necessary for a "UO_2 3-batches -MOX 3-batches" core management. In that case, a third of the UO_2 and a third of the MOX assemblies are yearly replaced, that means 36 UO_2 fuel assemblies and 16 MOX fuel assemblies.

Some MOX fuelled reactors have now switched from the previously described core management to a so-called "hybrid core management." In that case, a quarter of UO_2 assemblies is yearly replaced. The first EDF reactor loaded with MOX fuel was Saint-Laurent Bl, in 1987. The in-core experience, based on several hundred assemblies loaded, with reloading on a l/3 cycle basis, shows that there is no operational difference between UO_2 and MOX fuels, both in terms of performance and safety. MOX fueling of 900 Mwe EDF's PWRs, with a limited in-core MOX ratio of 30 %, has only needed minor adaptations, such as addition of control rods, modification of the boron concentration in the cooling system and precaution against radiation exposure, easy to set up (optimisation of the fresh MOX fuel handling process, remote inspection equipment). In such conditions, plant safety is not affected and operation remains the same.

To cope with the growing MOX fuel demand, some countries have equipped themselves (or should equip themselves in the near future) with the state-of-art MOX industrial capabilities. This growing demand is obviously linked with a higher diversity in fuel designs requirements. The empowerment of the MELOX plant, the first high-throughput MOX fuel fabrication facility in operation in the world, is in keeping with this situation: the MELOX West Fitting Building (MWFB) resulting from an optimized design know-how, is the demonstration of the COGEMA Group high capability of adaptation. With the MWFB, the completion of a versatile fabrication plant adapted to the international fuel market is reached.

INTRODUCTION

Recovering and recycling of a large proportion of reusable material (Uranium and Plutonium) after combustion in reactors, usually named as Reprocessing-Conditioning-Recycling strategy, is a natural way for a sustainable and responsible development of the nuclear fuel cycle. This nuclear energy policy based on a closed fuel cycle meets the twofold requirement of an environmental-driven approach and of an optimal use of natural resources.

The RCR strategy achievement lies in the implementation of multiple recycling, itself relying on a well running Mixed Oxide (MOX) fuel fabrication.

For almost thirty years, European utilities have been involved in reprocessing and recycling programs. As of February 1, 1999, 32 Light Water Reactors (LWRs) had been loaded with MOX fuel in Europe. It clearly means that plutonium recycling in MOX fuels is a mature industry, with successful operational experience in fabrication plants in some European countries, especially in France. French experience in MOX fuel industrial utilisation has been growing. considerably since its start in 1987. Short term EDF's strategy implies an increased number of MOX - IoadedLWRs : up to 28 reactors are expected to run with MOX fuel in France. The growing needs in MOX fuel are also linked with a higher diversity in fuel design requirements for both BWRs and PWRs.
Foreseeing this growing and diversified needs in MOX fuel elements, not only in France but also in other European countries and in Japan, COGEMA Group pooled in 1997 the production resources of three independent plants by setting-up an homogeneous and versatile MOX fuel fabrication tool, known as the "COGEMA Group MOX Platform" (CGMP).
The CGMP is the result of the wish of harmonisation in terms of process and quality control, while ensuring a high degree of safety and a permanent respect of environmental quality.

This paper firstly presents the cumulated in-core experience and the evolution in the MOX fuel demand, showing a real expansion of the MOX fuel market. The COGEMA Group MOX Platform, whose main objective is to satisfy such an evolving demand, will then be described. The MELOX plant, which holds an important place within the CGMP, illustrates COGEMA MOX production high flexibility through the implementation of the proven MIMAS process.

1. The MOX fuel market: an expanding market

Facing the upcoming deregulated market for electricity supply, Utilities have to further enhance their technical performance and economic competitiveness.

An opfimised management and an outstanding in-core performance

One of the main objectives of Utilities that have opted for plutonium recycling is to bring the overall performance of MOX fuel up to that of UOX fuel, in particular in terms of discharge burn-up.

Several types of core management exist in Europe. Germany has the highest authorised in-core MOX ratio in Europe, up to 50 % for the three KONVOI PWRs (Isar-2, Neckar-2 and Emsland). The maximum in-core MOX ratio reaches 40 % in Switzerland (Beznau-1) and 20 % in Belgium.

In France, EDF has been undertaking "hybrid" fuel management since 1993. In this case, a core contains up to 48 MOX Fuel Assemblies (FAs) representing an in-core MOX ratio of about 30 %. This hybrid cycle is performed with 4 batch fuel management for U02 assemblies and 3 batch fuel management for MOX. EDF's short term strategy involves switching from the so-called "hybrid" core management to the same 4 batch core management for both types of fuel, which leads to the same cycle cost as U02 4-batch management. Feasibility studies have shown no major difficulties and safety analysis are expected to be launched during the year. The targeted discharge burn-up for both types of fuel is about 50 GWd/t. The increase in discharge burn-ups has already been initiated in some

European countries, such as Germany (typical average discharge burn-up of 45 GWd/t with some FAs irradiated to 60 GWd/t).

In terms of safety and performance, the in-core experience, based on more than 1500 MOX Fuel Assemblies loaded in European reactors, shows that there is no operational difference between U02 and MOX fuel. MOX fuelling of 900 MW EDFs reactors has only needed minor adaptations, such as the addition of control rods and the modification of boron concentration in the cooling system.

Because of the high in-core performance and competitiveness of MOX FAs, Utilities are prone to use more and more MOX fuel in reactors, as shown hereafter.

The increasing number of moxified reactors

Taking advantage of excellent operational feedback in reactors, the trend is towards expanding utilisation of MOX fuels in reactors.

The choice of recycling uranium and plutonium made by several countries, which is a result of a long term nuclear policy, goes back a long way. The first MOX loadings date back to 1972 in Germany (Obrigheim) and 1978 in Switzerland (Beznau-1). In France, further to EDF's decision in 1985 to load reactors with MOX fuels, the decision to build and commission a new industrial-scale plant dedicated to MOX fabrication, MELOX, was taken by EDF together with COGEMA and Framatome. The first French reactor loaded with MOX fuel was Saint-Laurent B 1 in 1987. The operational experience of MOX fuel use in France has been characterised since the mid 90's by a fast increase. As of February I, 1999, 17 French reactors have been loaded with MOX fuel, three others have authorisation to use it and 8 others are technically adapted to be loaded with MOX fuel. On the whole, 32 reactors today are loaded with MOX fuel in Europe.

In the near future, Japanese reactors will also use MOX fuel. The COGEMA Group will produce MOX fuel for Japanese utilities. Indeed, Japanese regulators recently approved the first loading of a BWR (TEPCO). Preliminary approval for a PWR (KANSAI) by Fukui Prefectural Governor paves the way for an industrial MOX utilisation in Japan. By 2010, eleven utilities shall be using MOX in 16 to 18 units.

Diversified needs

Besides the increase in terms of volume, MOX fuel demand is characterised by a wide variety of fuel designs, responding to an heterogeneous worldwide reactor pool.
Fuel Vendors have to meet those diversified needs by producing MOX FAs for both BWRs and PWRs:

- 14x14, 15x15, 16x16, 17x17 and 18x18 MOX FA's for PWRs
- 9x9, 10x10 and 11x11 MOX FA's for BWRs

2. The COGEMA Group MOX Platform

A versatile too/

To meet the growing and diversified MOX fuel demand, the COGEMA Group runs an outstanding industrial tool : the "COGEMA Group Mox Platform" (CGMP), including the MELOX plant, the Cadarache plant and a percentage of the BELGONUCLEAIRE PO plant production capacity. For the period 1995-2000, this percentage amounts to 70 % and for the period 200 I-2006 it will reach 50 %.

The CGMP also includes FBFC-International, dedicated to fuel rods assembling. The CGMP is already able to produce 180 tHM/y (tons of Heavy Metal per year) of any MOX fuel design and the targeted production capability is expected to reach 350 tHM/y.

As of December 31st, 1998, about 750 tHM have been manufactured by the plants of the CGMP and almost 1600 FAs have been delivered to Belgium, French, German and Swiss Utilities (of various types, from 14 x 14 to 18 x 18 PWR FAs, and from 8 x 8 to 10 x 10 BWR FAs). Benefiting from the CGMP's high flexibility, this product range can be extended to other designs such as 11 x 11 BWR FAs.

The CGMP is highly versatile in terms of:

- **fabrication.** A given product would strictly be the same in terms of quality wherever it would be produced in the CGMP. Moreover, some of the production functions are interchangeable : as an example, fuel rods manufactured at the COGEMA Cadarache plant can be either assembled at MELOX or FBFC-International,
- storage. The fuel rods and assemblies storage capacity of any plant can be, if necessary, put to another plant's disposal,
- **FAs transport accommodation.** The wide variety of casks (FS 65, FS 69, TNB 176, MX, SIEMENS packaging...) possibly used in the CGMP illustrates the high flexibility in terms of FAs consignment.

High qualify products

The main objective of the CGMP is to satisfy the Fuel Vendors evolving demands, using any of its available production tools. This implies all the products manufactured by the CGMP present the same level of quality. The switch from the former - single blending step - COCA process to the - two blending steps - MIMAS process, now the common process implemented in the « Platform », illustrates this wish of harmonisation. Moreover, all three plants have been granted the IS0 9002 certification.

Qualification program with Fuel Vendors

Through CGMP, COGEMA is committed to .the Fuel Vendors' qualification program, taking advantage of the partnership with the main actors of the worldwide nuclear fuel industry

- In Europe with FRAGEMA and SIEMENS,
- In Japan with NFI, MHI, TOSHIBA and HITACHI

Certifications are expected from MHI in 2000, and from TOSHIBA, HITACHI in 1999-2000.
On February 1st, 1999 MELOX was certified by NFI for the following activities :

- Off line vendor qualification for production of MOX 17x17 B type FAs for nuclear power plants.

According to the NFI "certificate of approval", MELOX general organisation Quality Assurance system and technical capability meet NFI's requirements and are suitable for the production of mixed oxide FAs.

Regarding product quality, more than 80 characteristics are assessed during the pellet fabrication in the CGMP. With more than 30 parameters checked, the rods are also stringently inspected. About 15 visual and dimensional inspections are performed on fuel assemblies.

The impressive feedback experience, in terms of MOX plant design and plant operation, gained through the progressive implementation of CGMP has led to the quick and reliable start-up of the MELOX plant. MELOX reached the scheduled nominal production of 100 tHM/y within only two years, as explained hereafter.

3. MELOX contribution to the COGEMA Group MOX Platform

Towards a new generafion of MOXplanf

The construction of MELOX was initiated in 1990. The equipment was tested first in inactive conditions and then with U02 powders. After progressive introduction of plutonium in the production building, MELOX was brought on line in 1995. With the achievement of the scheduled capacity of 100 t HM in 1997, MELOX reached industrial maturity; the 1997 production rate rose up to one 900 MW PWR Fuel Assembly per day, corresponding to 264 rods or 100,000 fuel pellets. In March 1998, the MELOX production capacity reached the level of 20 tons per month.

The production capability of MELOX has been continuously developed, leading to the delivery of mixed oxide fuels not only for the French reactors (as originally planned in 1985) but also for foreign customers as soon as 1999.

To satisfy the specific needs of each utility, new equipment now completed will enhance the plant flexibility. A **multi-design assembling line** adapted to all MOX fuel designs, for both PWRs and BWRs, is under start-up at the MELOX plant.

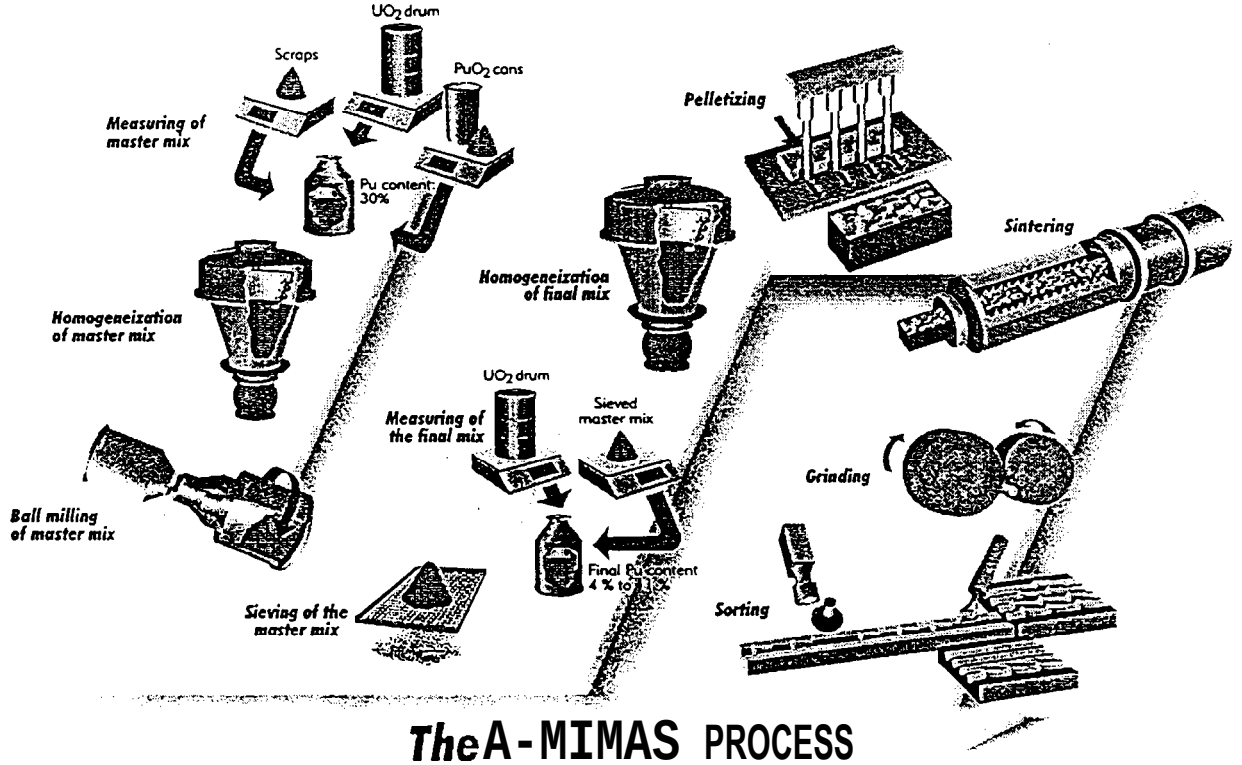
The Advanced-MIMAS Process, a fool for the *MELOX challenge*

The reference process implemented at MELOX is the A – MIMAS process, based on the sound and efficient MIMAS process. The latter had been implemented in the DESSEL and CADARACHE MOX plants, each currently producing 40 tons HM of MOX fuel per year.
With the implementation of the multi-design assembling line and the well-mastered A-MIMAS process, MELOX meets the customers specific requirements :

- the plant can receive any type of raw product
- the plant is able to produce any type of FAs for both PWRs and BWRs.

. An industrial process

The A-MIMAS process, perfectly adapted to high-scale production, and the MELOX plant layout are described hereafter :
MELOX receives raw products, PuO2 and U02, separately stored. The constant isotopy all along one



The A-MIMAS PROCESS

MOX production campaign (one of the main A-MIMAS features) is ensured through complete automation, from the selection and opening of canisters to the opening of chosen PuO2 cans. Selected PuO2 cans are then placed in a buffer store before the two blending operations.
The PuO2 powder is micronised in a ball mill with a portion of U02 powder to form a primary blend containing 30 % of Pu. This primary blend is then diluted with a free flowing U02 powder allowing the production of a secondary blend at a specific Pu enrichment, which is then homogenised. Scraps can be recycled at both stages in the process line after a specific treatment. The powders are fed into a large size blender (able to reach 650 kg), where the secondary batches are blended with additive components -lubricant and a small quantity of pore forming agent.

The powder obtained is pressed to form cylindrical pellets which are then passed through a high temperature furnace. The grinding operation is then performed to obtain the required diameter. The pellets are finally controlled and inserted into rods. Rods are then cleaned and checked with automatic control machines. The rods are placed in a metallic structure to form a Fuel Assembly. The completed assemblies are cleaned and submitted to final fabrication controls, then placed into storage before being sent to the customers.

Two of the main advantages of the A-MIMAS process are firstly a complete recycling of scraps and secondly a blending operation in two steps which allows to obtain final pellets, characteristics very close to those of UO_2 pellets.

. A large variety of inputs

The MELOX plant offers Utilities flexible characteristics of MOX fuel fabrication thanks to its complete automation. The plant enables to meet several constraints, in particular those related to the inputs qualities. The use of aged plutonium coming from high burn-up fuels, as well as high plutonium content is allowed through the techniques used at MELOX.

Moreover, the trend of an increased discharge burn-up of MOX fuel leading to a high plutonium content in the fabricated fuel is taken into account in the design of the MELOX plant.

. Multi-design innovative pellet fabrication equipment

To cope with various specifications and multi-client requirements, that is to say to produce all types of MOX fuel, multi-design equipment has been implemented:

a 4^{th} multi-design press with accommodations allowing a quick reconfiguration to all types of fuel designs,
a 3^{rd} grinding line accommodating an innovative « dust » removal system developed at the CDA, the COGEMA Advanced Development Center: this system allows maximum recovery of the grinding powders that can be then recycled after a specific treatment. Extensive tests with UO_2 pellets have also been performed at the CDA demonstrating the highest level of quality formulti-design pellets, handling and processing,
a 2" rod control/inspection line (multi-design),
a 2^{nd} assembling line (multi-design) allowing the fabrication of all the FA types previously mentioned.

Production capacities adapfed to a markef in expansion

As of December 31^{st}, 1998, the total number of FA's delivered to EDF (the French Utility) had reached 1032. Thanks to the plant adaptation, MELOX capabilities will continue to be developed in the coming years. By the 2000's, up to 250 tHM per year could be produced at MELOX, for both PWR and BWR.

Moreover, the COGEMA Group benefits from the global CGMP prospective capability to produce up to 330 t HM/y of MOX fuel.

This production capacity level is consistent with current and future Utilities MOX fuel needs, in Europe and also in Japan.

CONCLUSION

The CGMP, the first high-throughput MOX fuel manufacturing tool in operation in the world, is able to satisfy the growing MOX fuel demand highly linked with a wide diversity in fuel designs.
The progressive implementation of the CGMP, which constitutes the best answer in terms of product quality, delivery reliability and cost effectivness, illustrates the COGEMA Group high capability of adaptation.

The MELOX plant can be considered an outstanding achievement, taking full benefit from previous experience, while introducing several innovative features in terms of the fabrication process, radiation protection as well as environmental concerns. With the MELOX PWR and BWR multi-design assembling line, a next generation plant is under start up.

With the CGMP and in particular the MELOX plant, the completion of a versatile fabrication tool adapted to the evolving international fuel market has been reached.

OPERATIONAL EXPERIENCES IN MOX FUEL FABRICATION FOR THE FUGEN ADVANCED THERMAL REACTOR

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Abstract

The Japan Nuclear Cycle Development Institutes, JNC, has fabrication the MOX fuel for the Advanced Thermal Reactor, ATR, "FUGEN" in the Plutonium Fuel Fabrication Facility, PFFF, since 1974. For these 25 years, the MOX fuel fabrication has been progressed in stable manner after overcoming several problems at the start up of FUGEN fuel fabrication. Through the experience, improvements on process equipment and conditions have been taken place to achieve efficient MOX fuel fabrication on an engineering scale as 10 tons MOX per year. Main features of current fabrication process are digested as one step blending with ball milling, pelletizing without granulation and sintering with batch type furnaces. This fabrication process has been demonstrated and confirmed to be applicable techniques for the MOX fuel fabrication on this scale. This paper discusses the FUGEN fuel fabrication focused on the MOX pellet fabrication with operational experiences and improvements on the process.

1. INTRODUCTION

In 1966, the JNC has started development of Advanced Thermal Reactor, "FUGEN", adopting heavy water moderating and light water cooling system. The FUGEN was expected as the bridge between Light Water Reactor, LWR and Fast Breeder Reactor, FBR. The construction of FUGEN started in December 1970 and its first criticality was achieved in March 1978. The FUGEN fuel assembly has a unique cylindrical shape and consists of three layers of fuel pins. The numbers of fuel pins in each layer are 4, 8 and 16 from inner to outer layer respectively. The pellets with same plutonium content are loaded in the inner and middle layers pins and the other pellets with lower plutonium content are loaded in the outer layer pins. The structure of fuel assembly for FUGEN is shown in figure 1.

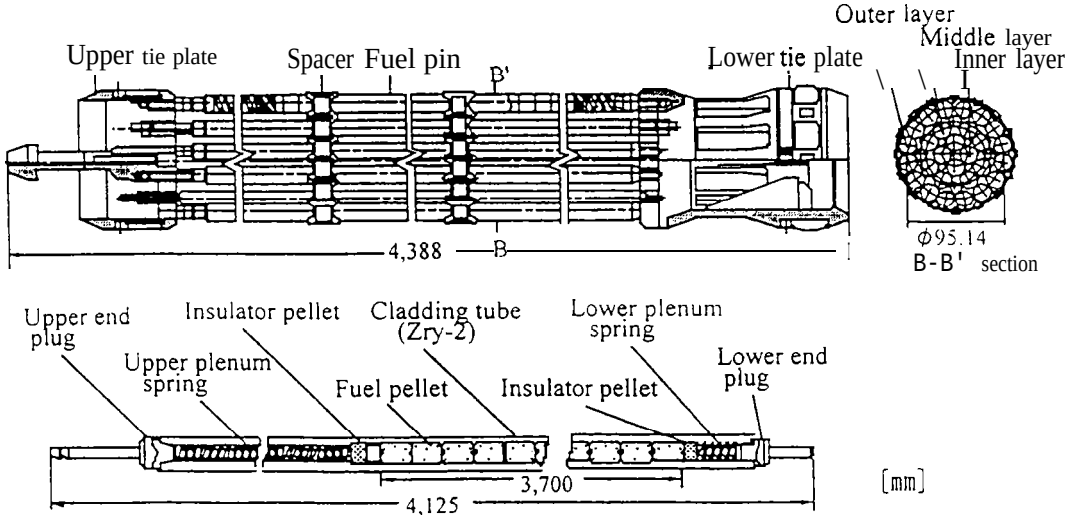


FIG. 1. FUGEN Fuel Assembly

In parallel with development of the reactor, the JNC started development of **MOX** fuel fabrication technology at the Plutonium Fuel Development Facility, PFDF, constructed by the design of NUMEC, U.S.A. Company, in 1966. Based on the experiences concerning plutonium handling and test fuel fabrications in the PFDF, the JNC designed and constructed PFFF in 197 1. In the PFFF, two MOX fuel fabrication lines, one is for FUGEN (hereinafter referred to as ATR line) and another is for the experimental fast breeder reactor "JOYO" (hereinafter referred to as FBR line) are installed. The PFFF ATR line was designed to fabricate MOX fuel through semi-automated operation with using reactor grade plutonium on the proto-typical scale of commercial MOX fuel fabrication. The annual fabrication capacity of ATR line in PFFF is about 10 tons of MOX. The fuel fabrication can be carried out through the control panel located beside the glove box and process equipment by operator from weighing to fuel assembling. The PFFF ATR line has started its operation with MOX fuel fabrication for the Deuterium Critical Assembly, DCA, in 1971 with 30 kg of MOX per day of fabrication capability. Total amount of MOX fuel fabricated for DCA was 9.1 tons of MOX. After DCA fuel fabrication, the ATR line was improved its production capability up to 60 kg of MOX per day for FUGEN fuel fabrication in 1974. The MOX fuel fabrication for FUGEN initial core fuel was completed in 1978.

2. PROCESS FLOW AND LAYOUT

The flow chart of current pellet fabrication process for FUGEN is shown in figure 2. Three kinds of feed materials such as plutonium dioxide or co-converted MOX powder, uranium dioxide and recovered powder are weighed to meet the fuel specification. The weighed powders are blended homogeneously with ball mill machine. After lubricant addition, the blended powder is pelletized into green pellets directly without granulation. Prior to sintering, the green pellets are processed in the de-waxing furnace to remove the lubricant from the matrix of the pellets. Followed by sintering, the pellets are ground by wet grinding method to satisfy the specification of diameter and surface flow. Finally, the pellets are inspected to be satisfied with fuel specifications such as dimensions, surface appearance and density as products. The scraps generated in the pellet fabrication are recycled as a feed material after calcination and reduction.

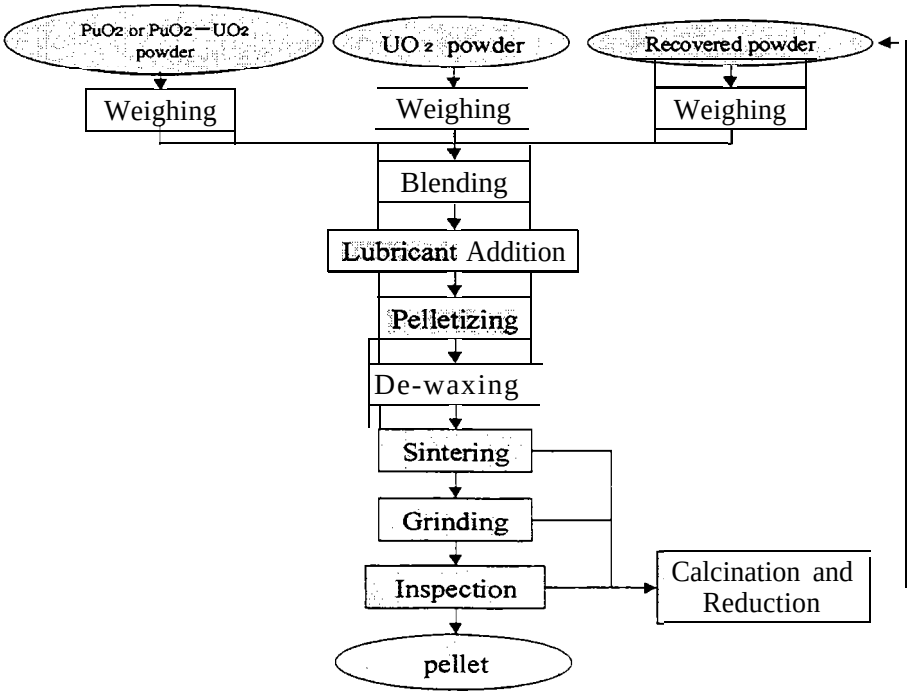


FIG. 2. The flow chart of current pellet fabrication process for FUGEN

The JNC began to utilize the standardized giant size glove boxes, 2.5m x 1m x 6m, in the PFFF to restrain the initial construction cost. Each glove box is connected to the transfer tunnel at its end so as to transfer nuclear materials through this tunnel between glove boxes along fuel fabrication process. Layout of glove boxes in PFFF is shown in figure 3. As moving from left to right in figure 3, the fuel fabrication is proceeded along the flow chart.

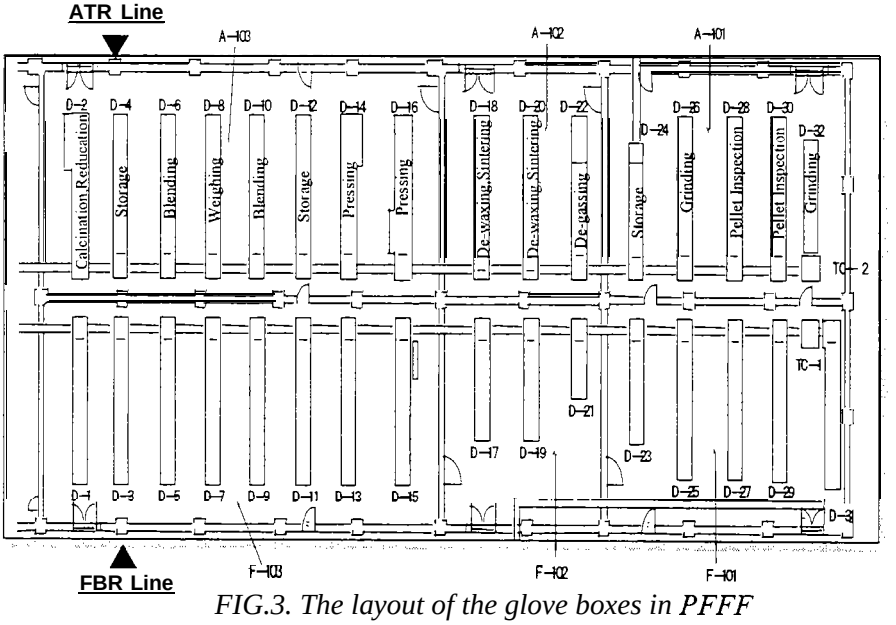


FIG.3. The layout of the glove boxes in PFFF

3. EXPERIENCES IN MOX FUEL FABRICATION

The MOX fuel fabrication process mainly consists of three processes such as pellet fabrication, fuel pin fabrication and assembling. Because the fuel pin fabrication process starting with pellet loading into a cladding tube has a similarity with uranium fuel fabrication, the MOX pellet fabrication process consisting of powder, pelletizing, sintering and finishing steps is main topic to be discussed here.

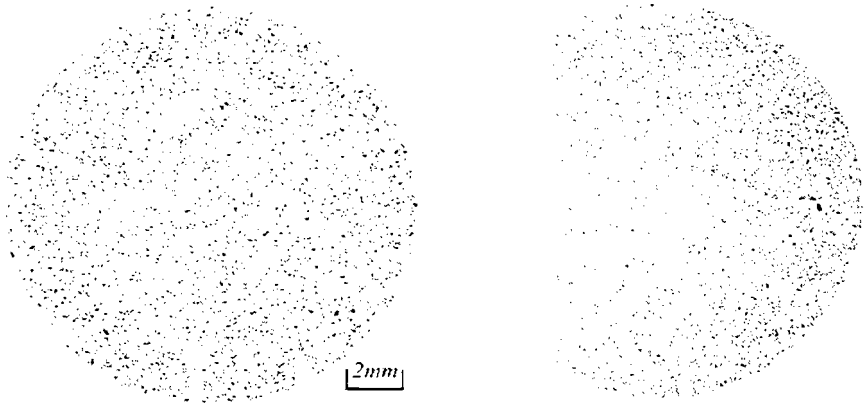
3.1 .Powder Preparation

The main features of the powder preparation employed in ATR line are one step blending with ball mill and powder preparation without granulation. In the one step blending method, the weighed feed powders to meet the specification are blended homogeneously only with ball mill machine. At the start-up of fuel fabrication for FUGEN in 1975, a two steps blending method was utilized with the purpose of the homogeneity of blended MOX powder. For the first step of two steps blending method, a VI type blending machine which had an excellent blending capability was utilized. For the second step, a ball mill machine was utilized in order to improve the homogeneity of the plutonium in the fuel pellets. The distribution of plutonium spots in the sintered pellet is the indicator of the homogeneity of blended MOX powder, which is brought by the performance of ball mill. In order to confirm the homogeneity of plutonium and uranium in a pellet, the observation of plutonium spots distribution for these two methods were carried out. Photo. 1 shows the typical example of the plutonium spots distribution in the sintered pellets taken by alpha auto-radiography method for one step blending and two steps blending respectively. Significant deference is not observed between them. Figure 4 shows the histogram of the diameters of plutonium spots measured in the Photo. 1. In case of two steps blending, the averaged diameter of plutonium spots is 39 μm and the maximum diameter is about 80 pm, while more than 98% of plutonium spots are under 60 μm. Regarding the one step blending, the averaged diameter is 44 μm and the maximum diameter is about 100 μm, while more than 96% of plutonium spots are under 60 μm. Both of averaged and maximum diameters of plutonium spots in one step blending are slightly larger than

those in two steps blending. However, both of these values obtained by two blending methods were low enough to be satisfied with specification limit, which is 200 μm with 100% plutonium concentration.

At the beginning of MOX fuel fabrication in the ATR line, Stainless steel was used as a material for the ball mill pot and ball, that caused a problem of MOX powder adhering on the surface of ball and inner surface of pot. To resolve this problem, a silicon rubber lining was applied for the inner surface of pot, and an alumina ball was introduced instead of stainless steel ball. In addition, the replacement of ball mill was taken place for the improvement of blending performance and for increase of capacity. After this replacement and confirmation of blending performance of new ball mill machine described in previous paragraph, one step blending method has been introduced in ATR line.

For the purpose of process control, the maximum size of plutonium spot has been measured for every production lot up to the present. The measurement of plutonium concentration in the spot is applied only for the production lot of which the maximum diameter of plutonium spots exceeds 200 μm. According to the measurement results of plutonium concentration in the spot, the maximum value in them is 18 %. Concerning the measurement of plutonium spots distribution, it had been carried out for every lot of feed plutonium material, and was abbreviated in 1992 because the accumulated data were low enough to assure the specification limit for the plutonium spot.



(1) By two steps blending (2) By one step blending

Photo. I. The picture of plutonium spots taken by alpha auto-radiography method.

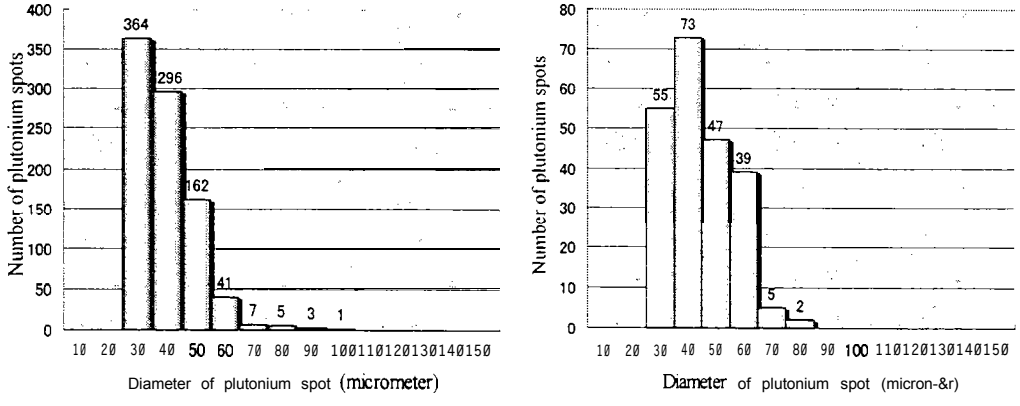


FIG.4. The distribution of the diameters ofplutonium spots

The flowability of blended powder to be supplied to pelletizing step is important element to realize pelletizing without granulation. From the beginning of fuel fabrication for FUGEN, the MOX powder has been prepared for pelletizing without granulation. The ball mill crushes MOX powder into the micrometer order size particles, which forms secondary particles in the following lubricant addition step as if granulation were taken place. That is the key technique to realize pelletizing without granulation. Various kinds of feed plutonium materials including plutonium dioxide, co-converted MOX powder and so on have been able to be utilized for the fuel fabrication. Because ball mill machine is enough capable of eliminating the differences in characteristics of feed materials. Therefore, the formation of the secondary particles at lubricant addition process creates enough flowability which brings easy powder filling into the die of pressing machine for FUGEN pellet.

3.2.Pelletizing

For process control purpose, the density and dimensions of green pellet are important parameters to control the quality of sintered pellets. At the early stage of pressing for a production lot, frequent sample taking is carried out to measure the density and dimensions of green pellets to determine adequate pressing conditions such as hydraulic pressure and so on, that is normal procedure for pelletizing operation. The standard deviation of green pellet density obtained through this procedure is less than 0.2 g/cm3 in the latest fuel fabrication for FUGEN, while the averaged green density is about 6 g/cm3 with 10% of allowance. This quality of green pellets is enough to satisfy specification of sintered pellet density after sintering. Figure 5 shows a cut view of FUGEN fuel pellet with its specification.

Fissile material enrichment of pellet	
Outer layer	1.84 ± 0.08 wt.%
Inner and middle layers	2.21 ± 0.09 wt.%
Dimensions	
Height	18 ± 1 mm
Diameter	14.4 ± 0.03 mm
Density	95 ± 2.0 %T.D
Oxygen to metal ratio	1.97 ~ 2.02
Plutonium spot	
A maximum diameter of plutonium spot < 800 μm	
However, concentration of plutonium spot which maximum diameter exceeds 200μm should be less than the value given by following equation.	
$C = -4.1824*10^{-7}D^3 + 9.3648*10^{-4}D^2 - 0.7314D + 212.18$	
C : Concentration of plutonium in a maximum Pu spot	
D : Maximum diameter of Pu spot	

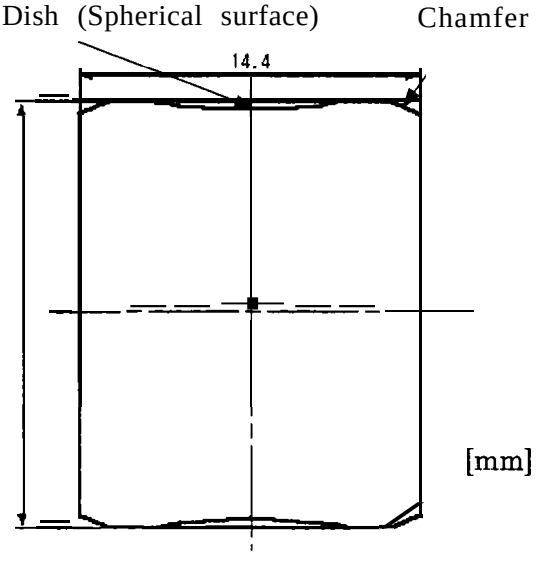


FIG.5 A cut view of FUGEN fuel pellet with its specification.

3.3 .Sintering

The sintering is the most important process to determine the qualities of MOX fuel pellet such as density, solubility of plutonium and uranium in the pellet, and oxygen to metal ratio and so on. In the ATR line, conventional batch type furnaces with cylindrical configuration have been utilized for de-waxing and sintering. This type of furnace has excellent thermal uniformity and easiness of maintenance although the production capability is small. 4 sets of batch type de-waxing and sintering furnaces were installed at the beginning of fuel fabrication for FUGEN. Each furnace can sinter one production lot of pellets equal to 30 kg of MOX, and the daily sintering capability of ATR line is 4 lots or 120 kg of MOX.

Each furnace is connected to the bottom of glove box as shown in figure 6. The green pellets are loaded into the furnace by manual operation with the crane installed at the ceiling of glove box. A production lot composed of normally 800-1000 pellets is stacked up on 11 molybdenum sintering dishes, and placed vertically into the furnace for sintering operation. 5% hydrogen and 95% nitrogen mixture gas is used for sintering atmosphere. The operating conditions in temperature for de-waxing to eliminate lubricant are 200 “C/h of rising rate, 2 hours keeping at 800 °C and cooling down by natural. On the other hand, the sintering furnace is operated with the conditions of 400 “C/h of rising rate, 2 hours keeping at about 1700 °C and 600 °C/h of cooling rate in temperature. The structures of de-waxing and sintering furnaces are shown in figure 7.

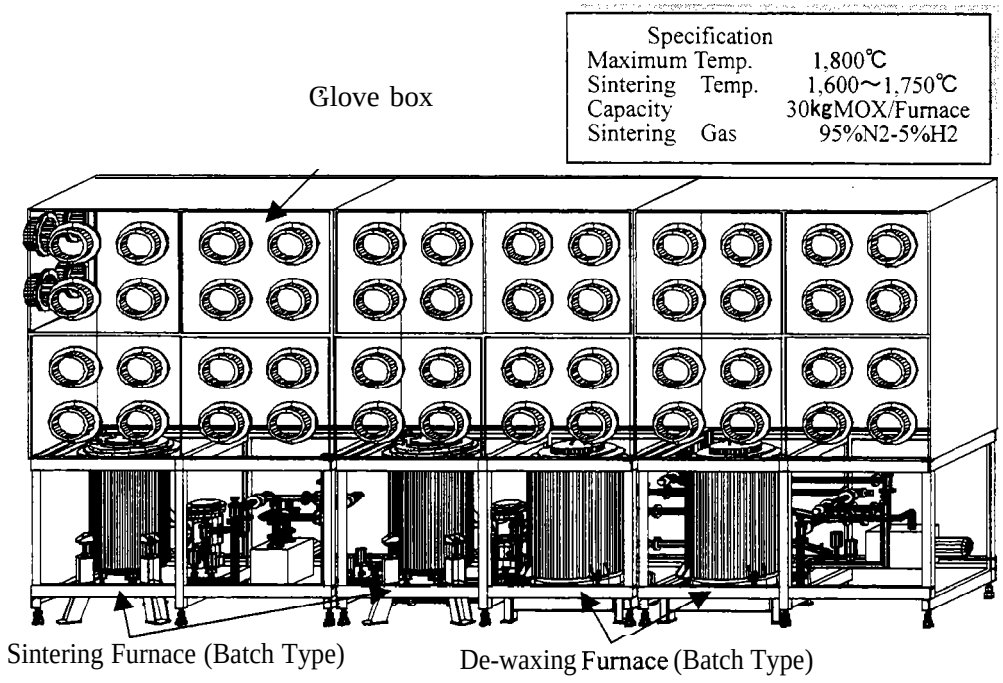


FIG. 6. The de-waxing and sintering furnaces for ATR pellet fabrication (Batch type)

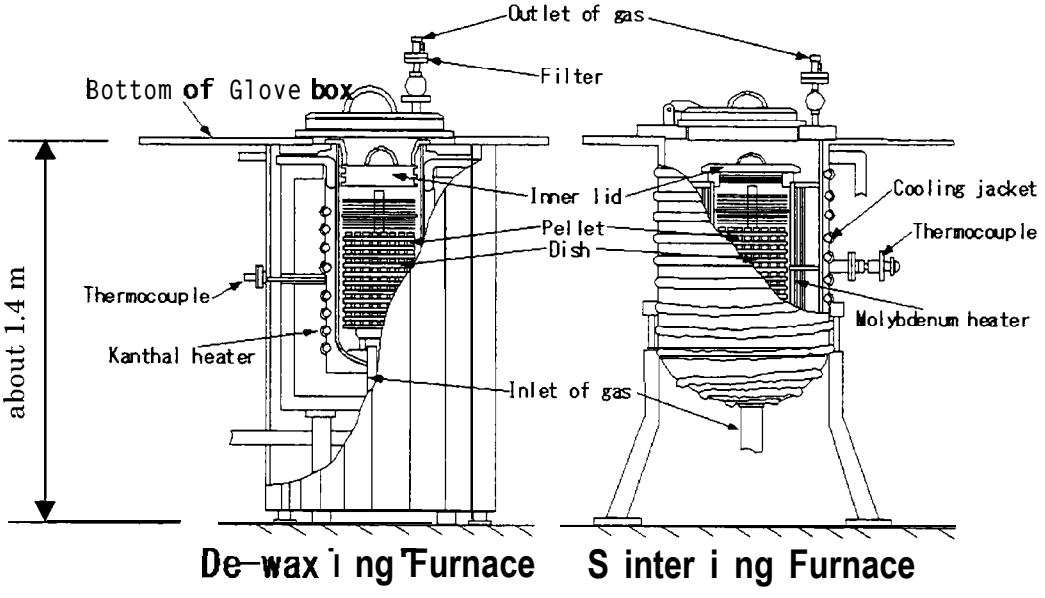


FIG. 7. The structures of de-waxing and sinteringfurnaces

The first replacement for all of 4 sintering furnaces had been carried out one by one during the period from 1982 to 1985 in parallel with fuel fabrication. The second replacement for all of them had been also carried out from 1995 to 1996 in parallel with fuel fabrication. Each sintering furnace itself still had enough durability after about 600 times of thermal cycles caused by sintering operations for about 10 years. The replacements of the sintering furnaces had been taken place mainly because of deformation of outer cylindrical jacket of the sintering furnace by thermal stress.

Because of the excellent thermal uniformity brought by cylindrical configuration, the standard deviation of sintered pellet density is small enough to assure the specification. Figure 8 shows the variation of averaged standard deviation of sintered pellet density in each campaign from the 11th to 28th reloaded fuel fabrications. In each campaign, more than 30 lots of pellets were fabricated. The standard deviation within these campaigns is estimated as 0.16%T.D. (Theoretical Density) in average. Concerning averaged sintered pellet density of each production lot, it is depend upon the sintering condition which is brought by the performances of sintering furnaces. Figure 9 shows the variation of averaged sintered pellet density in each production lot in the current campaign. The standard deviation of averaged sintered pellet density within this campaign is 0.32%T.D. They are small enough to be satisfied with the specification of 95±2.0%T.D. for sintered pellet density.

The production capability of ATR line with using 4 sets of furnaces is about 10 tons of MOX per year. The performance of this type furnace may not be sufficient for a commercial scale plant for MOX fuel fabrication. At the Plutonium Fuel Production Facility, PFPF, continuous type furnaces have been introduced and demonstrated for the commercial scale of MOX fuel fabrication for FBR, and batch type furnaces are also installed as the back up for them, which are designed based on the experiences gained at ATR line.

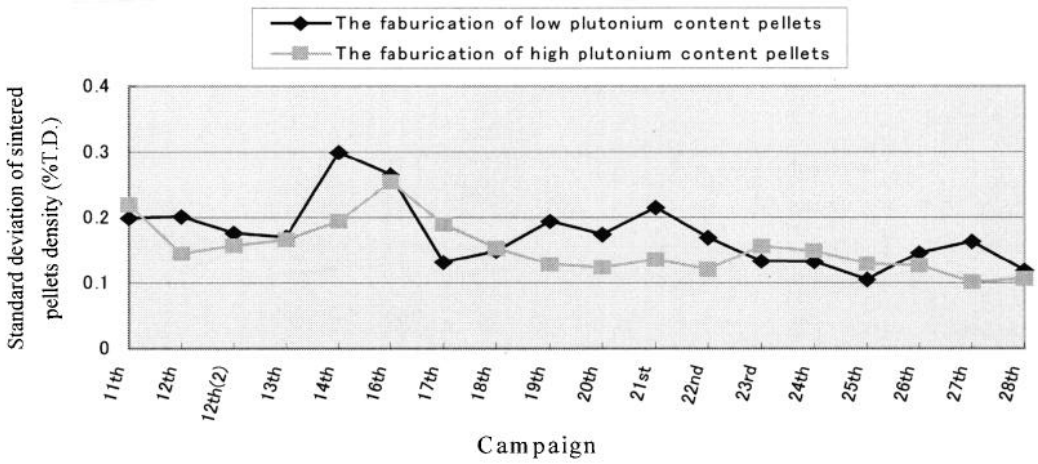


FIG.8. Averaged standard deviation within the campaign (From 11th to 28th)

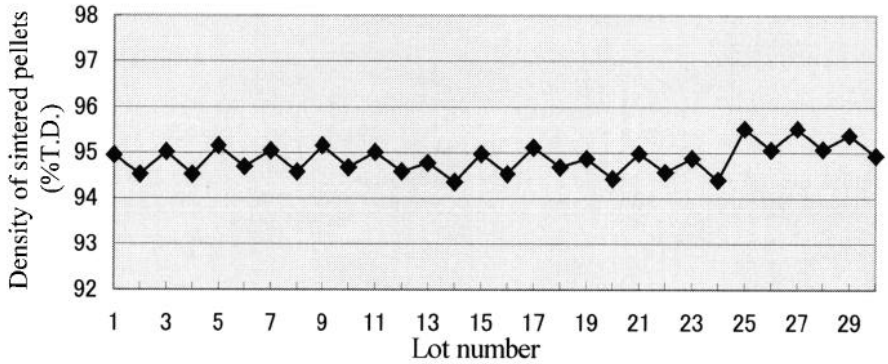


FIG.9. Averaged density of sintering pellets in the latest campaign

3.4.Finishing

The centerless grinding is essential step in the finishing process. From safety point of view, the management of the ground dusts in the glove box which is different from uranium dioxide pellet grinding is necessary. In order to meet the specification, a wet grinding method is applied for the all pellets grinding in FUGEN fuel fabrication. The standard deviation of outer diameter of ground pellets is less than 6 μm, which gives enough allowance for the specification of 14.4±0.03 mm. Severe critical control should be adopted because of the small critical mass resulted from the wet system. The ground dusts are collected together with cooling water and recovered by the centrifugal separation system to be recycled as one of feed materials after calcination and reduction.

The measurement of dimensions and density is the last step in pellet fabrication. This step is necessary to assure the quality of pellet. The early stage of fuel fabrication, the measurement was carried out by manual operation. Since 1980, it is carried out with automatic operation by laser in order to decrease radiation exposure during measurement and increase accuracy in measurement. Based on this experience, the measurement system by laser is also employed in the PFPF.

4. RESULTS OF 25 YEARS FABRICATION

The accumulated MOX fuel fabricated for FUGEN reached to 128 tons of MOX or 689 fuel assemblies as of March 1999. The amount of MOX fuel fabricated in the ATR line is shown in figure 10. Through these 25 years experiences, the ATR line has demonstrated MOX fuel fabrication on the engineering scale with a yield of more than 90%. With regard to the MOX handling technology, the ATR line has adopted dry process except grinding and this dry process is the indispensable technology to the current MOX fuel facility on a large scale.

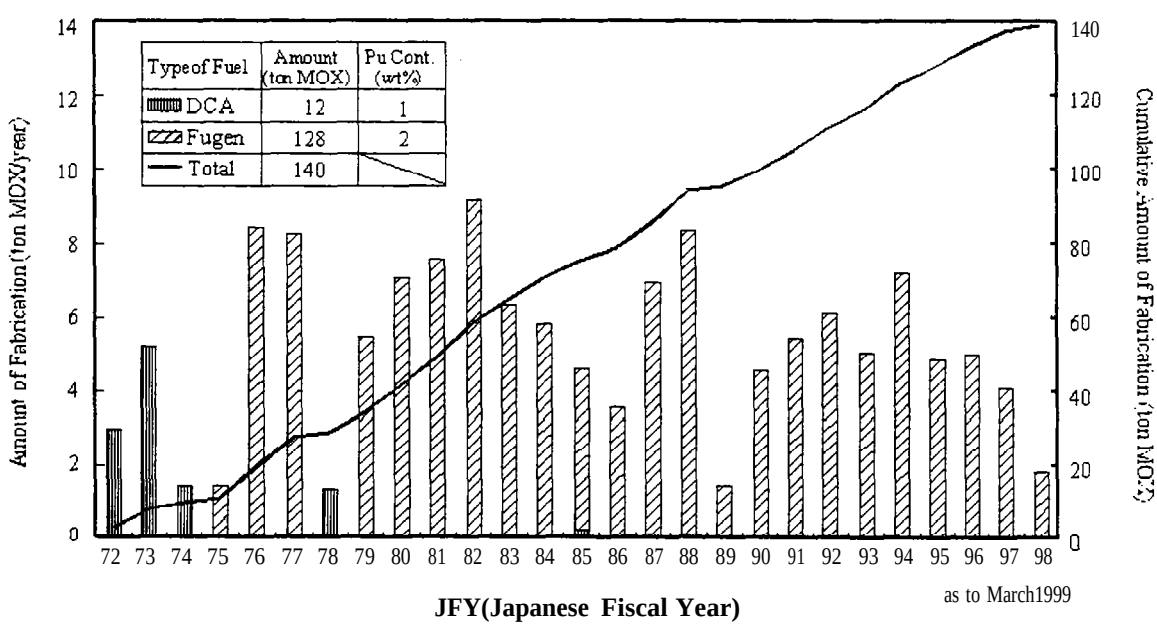


FIG. IO. The amount of MOX fueI fabricated in the ATR line

Up to the present, any failure has never been observed in all products of ATR line including 683 fuel assemblies loaded into FUGEN. In the ATR line, various kinds of plutonium recovered by JNC as well as foreign countries have been utilized as a feed material to fabricate MOX fuel for FUGEN. It was one of the epochs in the ATR line to fabricate FUGEN fuel by the utilization of plutonium recovered from FUGEN spent fuel in 1987 with closing the fuel cycle through FUGEN.

5. SUMMARY

Through the MOX fuel fabrication for FUGEN in the ATR line for this quarter century, the following key techniques have been demonstrated and confirmed to be applicable for 10 tons of MOX fuel fabrication per year.

- One step blending with ball mill
- Pelletizing without granulation
- Sintering with batch type furnace

These key techniques are essential ones to fabricate MOX fuel economically on this scale. They are also easily applied for the expanded scale of MOX fuel fabrication.

CURRENT DEVELOPMENTS OF FUEL FABRICATION TECHNOLOGIES AT THE PLUTONIUM FUEL PRODUCTION FACILITY, PFPF

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Abstract

The Japan Nuclear Cycle Development Institute, JNC, designed, constructed and has operated the Plutonium Fuel Production Facility, PFPF, at the JNC Tokai Works to supply MOX fuels to the proto-type Fast Breeder Reactor, FBR, "MONJU" and the experimental FBR "JOYO" with 5 tonMOX/year of fabrication capability. Reduction of personal radiation exposure to a large amount of plutonium is one of the most important subjects in the development of MOX fabrication facility on a large scale. As the solution of this issue, the PFPF has introduced automated and/or remote controlled equipment in conjunction with computer controlled operation scheme. The PFPF started its operation in 1988 with JOY0 reload fuel fabrication and has demonstrated MOX fuel fabrication on a large scale through JOY0 and MONJU fuel fabrication for this decade. Through these operations, it has become obvious that several numbers of equipment initially installed in the PFPF need improvements in their performance and maintenance for commercial utilization of plutonium in the future. Furthermore, fuel fabrication of low density MOX pellets adopted in the MONJU fuel required for a complete inspection because of difficulties in pellet fabrication compared with high density pellet for JOYO. This paper describes new pressing equipment with a powder recovery system, and pellet finishing and inspection equipment which has multiple functions such as grinding measurements of outer diameter and density, and inspection of appearance to improve efficiency in the pellet finishing and inspection steps. Another developments of technology concerning an annular pellet and an innovative process for MOX fuel fabrication are also described in this paper.

1. INTRODUCTION

The PFPF adopted a concept of fully automated fuel fabrication and contact maintenance for process equipment by glove operation to reduce personal radiation exposure during fabrication of the MOX fuel containing about 20 ~ 30 % of plutonium on the scale of 5 tonMOX/year. (See Ref.[l]) In order to realize this concept in the PFPF, structure of process equipment in a glove box become more complex than conventional one. In addition to this, the batch size used for MOX fuel fabrication was also increased up to 38 kgMOX from 15 kgMOX used in the Plutonium Fuel Fabrication Facility, PFFF. Because the MONJU fuel adopted low density pellet different from JOY0 fuel, two kinds of MOX pellets such as low density pellet and high density one have to be fabricated alternately in the PFPF. In the beginning of operation, the PFPF encountered difficulties in fuel fabrication caused by unaccustomed operation of fully automated equipment, troubles in automated equipment and low density pellet fabrication. However, those difficulties have been overcome by the improvement of process equipment and operational conditions in the PFPF and it resulted in 3 tonMOX of annual production for MONJU fuel recorded in 1992. Taking account of the rejected ratio in low density pellet fabrication for MONJU, tbis throughput of pellet fabrication process achieved almost designed annual production capability.

Up to the present, the PFPF has fabricated 401 MOX fuel assemblies in total, 116 assemblies for JOY0 and 285 assemblies for MONJU including 205 assemblies of initial core fuel, and the cumulative amount of MOX fuels has reached to about 11 tons of MOX in this decade.

It is the mission given for JNC to develop and demonstrate the FBR fuel cycle including MOX fuel fabrication technology for commercial use of plutonium in the future. In order to realize this mission, the technologies concerning the FBR fuel cycle that can compete with LWR fuel cycle in economics need to be established. Under this recognition, new technologies for the improvement of performance of MOX fuel fabrication equipment and introduction of safeguards system compatible with the automated MOX fuel fabrication have been developed and demonstrated along the MOX fuel fabrication in the PFPF. (See Refs.[2-51)

2. OPERATIONAL EXPERIENCES IN THE PFPF

2.1. Automated MOX fuel fabrication for F'RR

In order to realize the concept of fully automated fuel fabrication, the structure of process equipment in the PFPF becomes more complex than conventional ones in the PFFF, and it increases the difficulties in maintenance operation. Especially, in the glove box handling bulk MOX powder, it disperses into the glove box and is trapped in the process equipment. This trapped powder had been accumulated in the process equipment along the fuel fabrication in the PFPF. It resulted in decreasing life time of process equipment and increasing personal radiation exposure during maintenance operation because the dispersed MOX powder promoted the deterioration of cables and the wear of sliding surface and shaft parts in the process equipment.

In case of pressing equipment in the first generation used in the PFPF, it adopted a withdrawal type press designed to be compatible with fully automated operation on the basis of the pressing machine used in the PFFF. This equipment employed a reversal hopper that is used to feed the granulated MOX powder being accommodated in a transfer container into pressing machine by turning the container upside down. This hopper caused falling and scattering of MOX powder, and generated hold up in the glove box. The other main sources of dispersing the MOX powder into the glove box were moving components of the equipment handling the MOX powder directly such as a powder shaker to supply the MOX powder into die, die set and green pellet transfer system. Figure 1 shows the locations where MOX powder scatters and disperses in this equipment. In addition, there was a lack of consideration to easy maintenance by glove operation in the design stage for the automated equipment in the past. Although the press machine itself had a sufficient performance to pelletize the MOX powder into green pellets with stable quality, the dispersion of MOX powder and maintenance by glove operation were not fully taken into consideration in the design step for this equipment. With reflecting these experiences obtained through the operation of automated equipment in the design, new equipment in the second generation has been developed and demonstrated in the PFPF.

2.2. Low density pellet fabrication

The specification for density of MONJU pellet is 85 %T.D. (Theoretical Density) with 2 % of allowance, and this nominal value is almost 10% lower than that for JOY0 pellet. In this low density pellet fabrication for MONJU, about 10 volume % of organic material, herein after referred to as pore former, needs to be added into the blended MOX powder to drop the pellet density by 10 % lower than JOY0 pellet in addition to binder and lubricant. These organic materials such as binder and lubricant are usually decomposed and removed from the matrix of pellet completely during dewaxing step prior to sintering in the pellet fabrication for JOYO. However, in case of low density pellet fabrication, certain content of carbon was still remained in the matrix of pellet after dewaxing and it was substituted

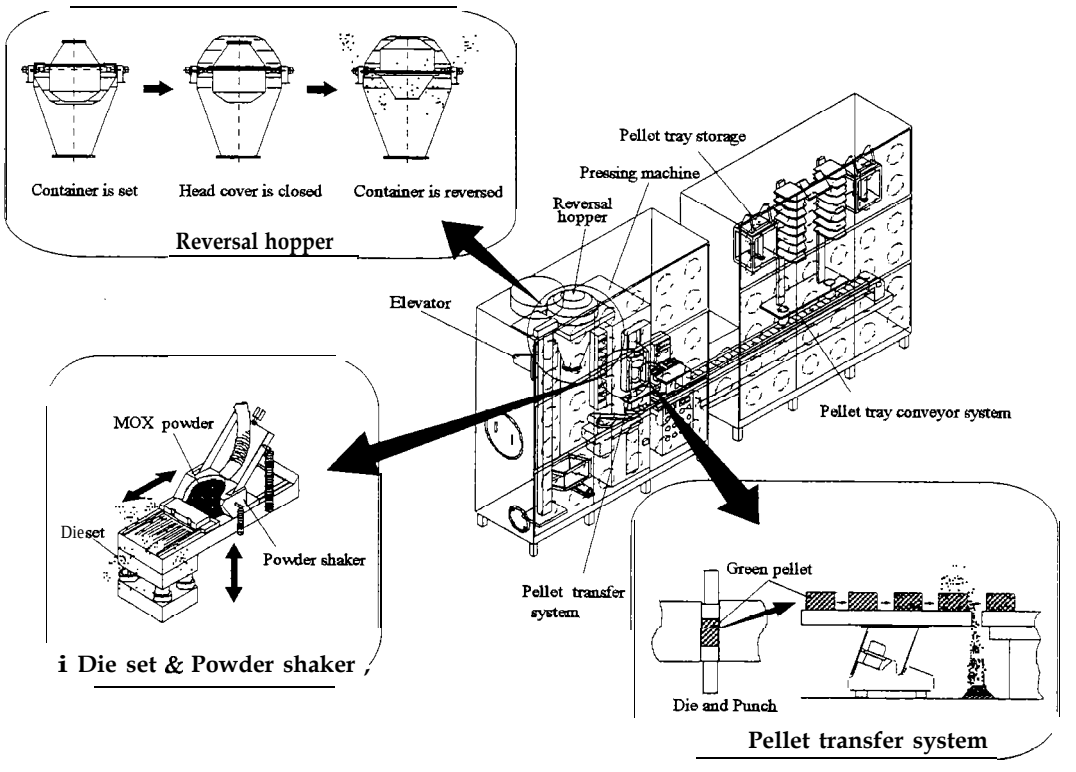

Figure I. The locations where the MOXpowder scatters and disperses in the conventional pressing equipment

for nitrogen contained in the atmospheric gas. Consequently, the nitrogen content in the sintered pellet for MONJU was over the specification limit. It was caused by the utilization of nitrogen and hydrogen mixture gas for the atmospheric gas in the furnaces as same as high density pellet fabrication for JOY0 without utilization of pore former. (See Ref.[6]) Therefore, this atmospheric gas was changed from nitrogen and hydrogen mixture gas to argon and hydrogen mixture gas to restrain the nitrogen content in the sintered pellet. The analytical data of nitrogen content in the sintered pellet for MONJU initial reload fuel is shown in figure 2. After adoption of argon gas for the atmospheric gas in the furnaces, the nitrogen content in the sintered pellet has been restrained into the values lower than specification limit.

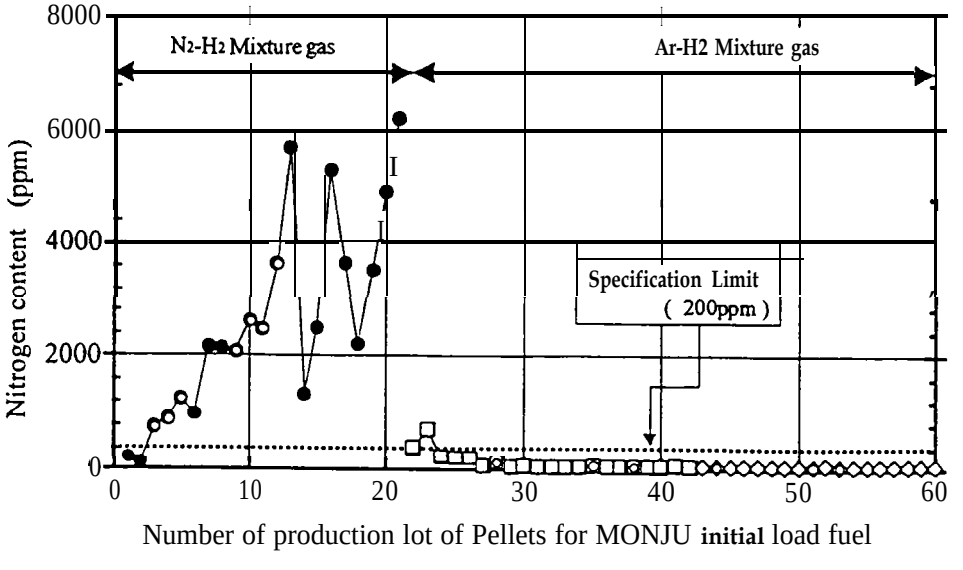

Figure 2. Analytical data of nitrogen content in the sintered pellets

The addition of pore former also resulted in wide spreading sintered density of pellet compared with high density pellet. (See Ref.[6]) It is very difficult to make 10 volume % of pore former distribute into the blended MOX powder uniformly because the densities of MOX powder and pore former are far different from each other. This non-uniform distribution of pore former in the blended MOX powder also gives affects on the diameter and surface appearance of sintered pellet. Figure 3 shows the average densities of each production lot of sintered pellets for JOY0 and MONJU with their standard deviations. In general, the PFPF has to handle a numerous number of MOX pellets due to smaller size of FBR pellet compared with LWR fuel fabrication. In addition to that, a complete inspection for diameter and density was caused by this wide spread of the density. Therefore, improvement and optimization of pellet finishing and inspection steps were essential to increase the efficiency in low density pellet fabrication.

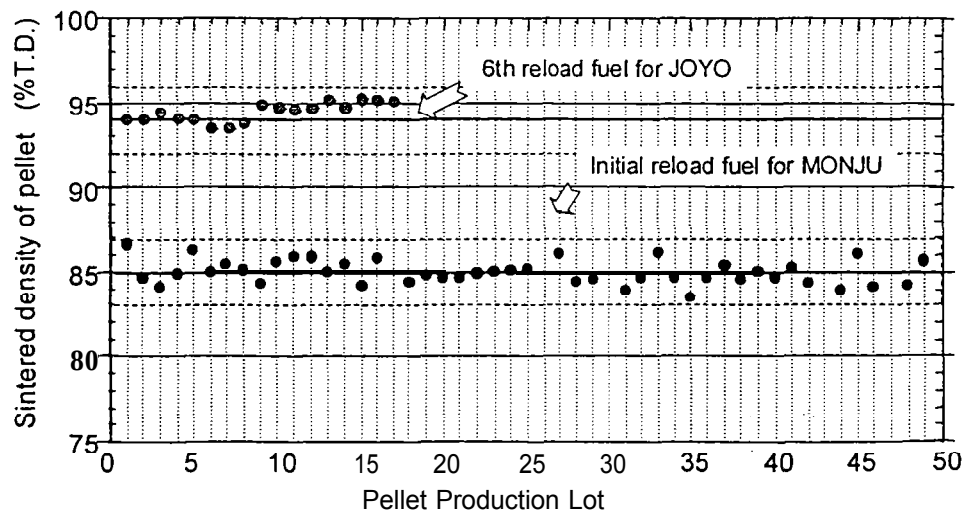

Figure 3. Statistic of sintered pellets for JOY0 and MONJU

3. DESCRIPTION OF CURRENT DEVELOPMENTS IN THE PFPF

3.1. New pressing equipment

The development of new pressing equipment was started in 1992 with the purpose of overcoming the difficulties such as hold up, maintenance and productivity arising from the operation of conventional ones in the first generation. Concerning the hold up, in order to prevent the scattering and dispersing of MOX powder which had been caused in the conventional one, a knife gate was attached to the powder container during powder supply operation by turning the container upside down as shown in figure 4. For collecting the scattered and dispersed MOX powders, the sucking heads connected to the powder recovery system were applied for all locations where the MOX powder may scatter and disperse.

Regarding the maintenance, layout of main components in the glove box was designed fully taking contact maintenance with globe operation into consideration. For realizing this concept, several parts that needs frequent adjustments such as oil pressure unit, compressed air control unit and clutch for speed control unit were arranged outside of the glove box different from conventional one. In addition, a small crane was introduced in the glove box to replace major components for maintenance operation. Prior to the installation of equipment in the PFPF, it was confirmed thoroughly that the equipment was able to be repaired by glove operation easily in cold test operation.

For the improvement of productivity in pressing step, the die set and feeder that were to able to pelletize 4 green pellets simultaneously with variable pressing speed from 5 to 20 strokes per minute were introduced. This performance of pressing equipment was also confirmed completely witb ferrite powder before its installation in the PFPF.

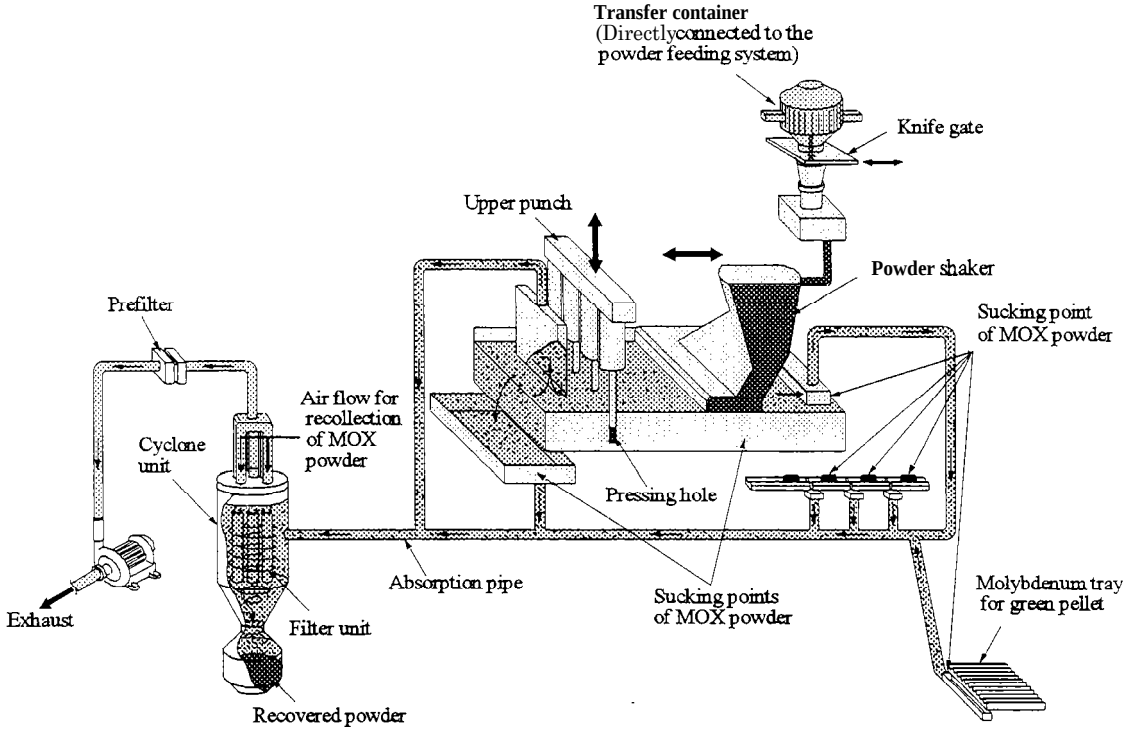

Figure 4. The main structure of new pressing machine

This new developed pressing equipment started its operation in 1994 with MONJU fuel fabrication and has pelletized more than 2.6 million pellets up to the present. By the introduction of this equipment, the amount of hold up has been kept in lower level than before. At the same time, the amount of radiation exposure during maintenance operation for it decreased by one eighth of before. Through the operation so far, it has been demonstrated that the powder recovery system is indispensable technology to the automated fuel fabrication equipment in the glove box.

3.2. Powder recovery system

A specially designed cyclone unit is the most important component in the powder recovery system developed by the JNC for automated MOX fuel fabrication as shown in figure 5. This system consists of sucking head applied for the location where the MOX powder may disperse, three filter units made of polyester fiber laminated by polytetrafluoroethylene and blower. This material utilized for filter units is selected after many cold tests with simulated MOX powders to confirm the capability of trapping the MOX powders in operational condition and recollecting them by reversed air flow through the filter unit. In the cold test operation, it is also verified that the filter unit has a enough capability to trap more than 99.9 % of simulated MOX powders. In addition, of course, it is confirmed that the material has sufficient resistance for radiation damage caused by at particles emitted from plutonium by hot tests with plutonium. The MOX powers collected through the sucking head are trapped on the surface of filter unit and they are recollected into the powder recovery container in reverse operation. In ordinary operation, the first filter unit is used to trap the collected MOX powders while the second one is in reverse operation to remove the trapped MOX powders from this unit. The last one is back up for the first one.

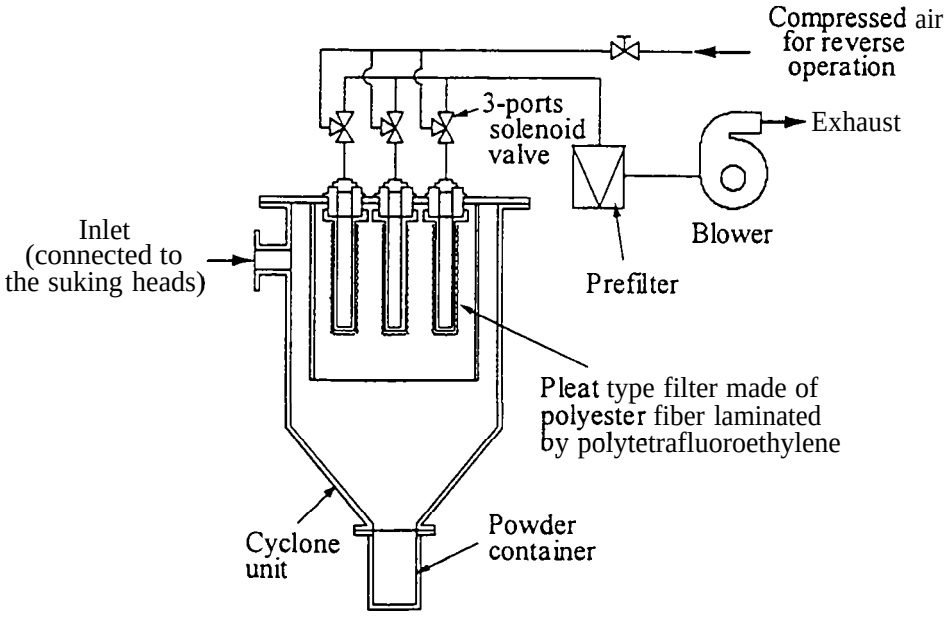

Figure 5. Detailed structure of powder recovery system

3.3. Pellet finishing and inspection equipment

It is necessary to improve the pellet finishing and inspection steps in the low density pellet fabrication because a complete inspection for diameter and density was needed. The pellet finishing and inspection steps originally consisted of three process steps such as grinding the sintered pellets, measuring their dimensions and visual inspection. For each of these steps, one unit of process equipment installed in two standardized size, 3m x 3m x 1 m, of glove boxes was prepared in the past. However, in order to save the manufacturing cost of equipment and increase the processing speed in these steps as well, the development of new pellet finishing and inspection equipment was started in 1995 by unifying and down sizing the structures of equipment, and optimizing the layout of devices including pipes and cables in the design. These improvements resulted in the new equipment in which three units of equipment were able to be integrated into one unit of equipment with realizing continuous processing as shown in figure 6. By the introduction of this equipment, reduction of space required for installation of equipment was successfully done, and substantially reduction of the time required for the transfer of pellets between these process steps was also achieved. In addition, by reducing a tact time in each process step of this equipment and introducing a series of line consisted of outer diameter measurement, density measurement and visual inspection, it has been succeeded to reduce the total time required for processing in this equipment.

For reducing hold up in the new pellet finishing and inspection equipment, a compact and high-performance cyclone unit for powder recovery system has been also developed. For collection of the MOX dust generated in course of operation such as grinding the sintered pellets and handling the pellets, these cyclone units have been installed at the locations where hold up might occur.

In addition to these improvements, grouping of equipment control software related to remote and automatic operation (sequence programs) has been introduced, and control software that corresponds to operation flow of equipment has been developed. A system was also developed for taking the progress of status of program into the computer on real time to identify the cause of abnormality and recover from abnormal stoppage through the operations on a CRT screen. By the introduction of this new system, man-machine interface in the operation of process equipment has been substantially improved from conventional ones.

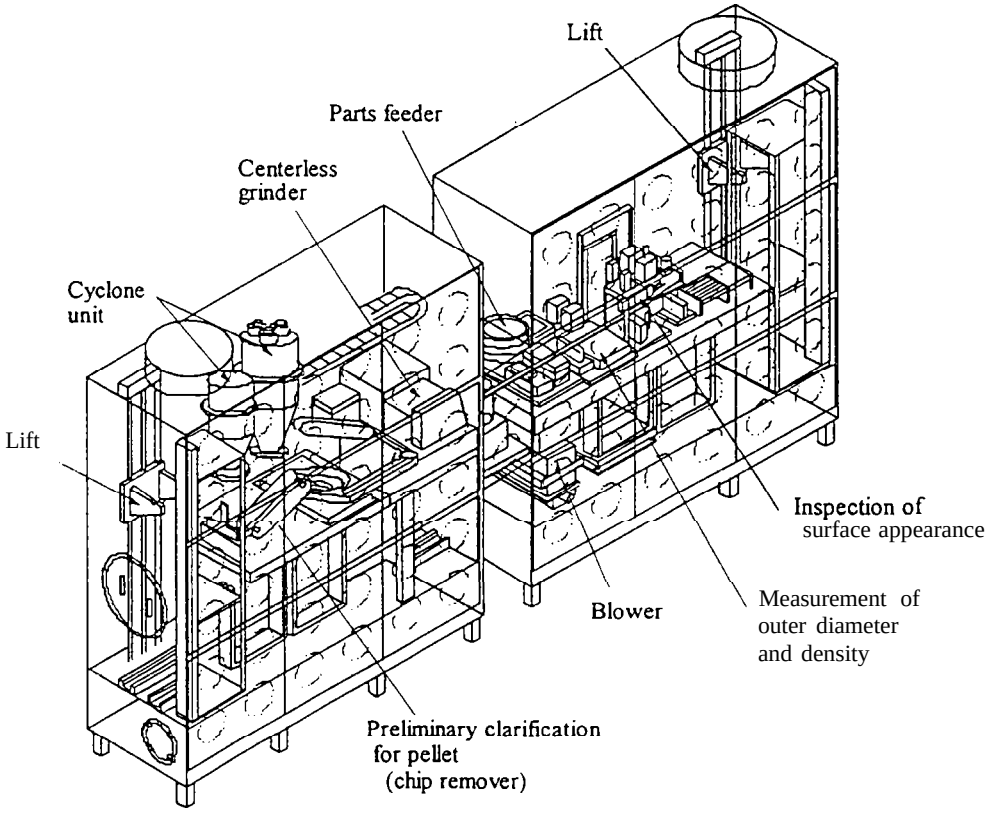

Figure 6. Overview of pellet finishing and inspection equipment

Now, this equipment has more than twice as high processing capability as a conventional one, occupies only half the space, and has about four times higher processing performance per installation space as is shown in table 1.

TABLE 1. THE COMPARISON OF PERFORMANCES BETWEEN NEW EQUIPMENT AND CONVENTIONAL ONE

Items	Conventional Equipment	Newly Developed Equipment
Numbers of Equipment	-Measurements of pellet diameter and density d d : 2 -Grinding : 1 -Inspection of appearance : 2 Total : 5	- Finishing and Inspection : 1 Total : 1
Numbers of Glove Box (Standardized Size:9m^3)	6	2
Areas for Required for Installation of Equipment	18 m^2	6 m^2
Performance of Processing	-Measurement of pellet diameter and density : 2-3 sec./pellet -Grinding : 2 sec./pellet -Inspection of appearance : 4~6 sec./pellet	1.7 sec./pellet
	-Measurement of pellet diameter and density : 5 shifts/lot -Grinding : 4 shifts/lot -Inspection of appearance : 6 shifts/lot	3 shifts/lot

4. DEVELOPMENTS FOR FUTURE

4.1. **Pressing equipment for annular pellet**

In order to decrease the cost of FBR fuel cycle, it is necessary to increase the burn up of MOX fuel in the reactor. Introduction of high density annular pellet with larger diameter is one option for realizing this objective. In the PFPF, the development of pressing equipment for annular pellet is now undertaken since 1997. The prototype pressing equipment was already completed in 1998, and its performances including durability of center rod equipped with die set have been confirmed with simulated MOX powder. This pressing equipment can pelletize 6 annular pellets per stroke with variable pressing speed up to 15 strokes per minute. It is scheduled to install this equipment in the test facility with uranium in this autumn.

4.2. **Short process**

It is the mission given for JNC to develop and demonstrate the FBR and its related technologies for commercial use of plutonium in the future. Along this mission, the JNC has carried out the developments of new MOX fuel fabrication technologies described in previous section. In order to meet the final goal of this mission, further development, which can be achieved by the introduction of an innovative technology, is still necessary.

Under this recognition, basic research and development have been started to realize the innovative fabrication process, called short process, as is shown in figure 7. In this concept, it is possible to skip the number of process steps and also achieve same quality level in the products compared with the conventional process. After the realization of short process, the plutonium content is adjusted in mixing step of plutonium nitrate and uranium nitrate at the conversion facility with the

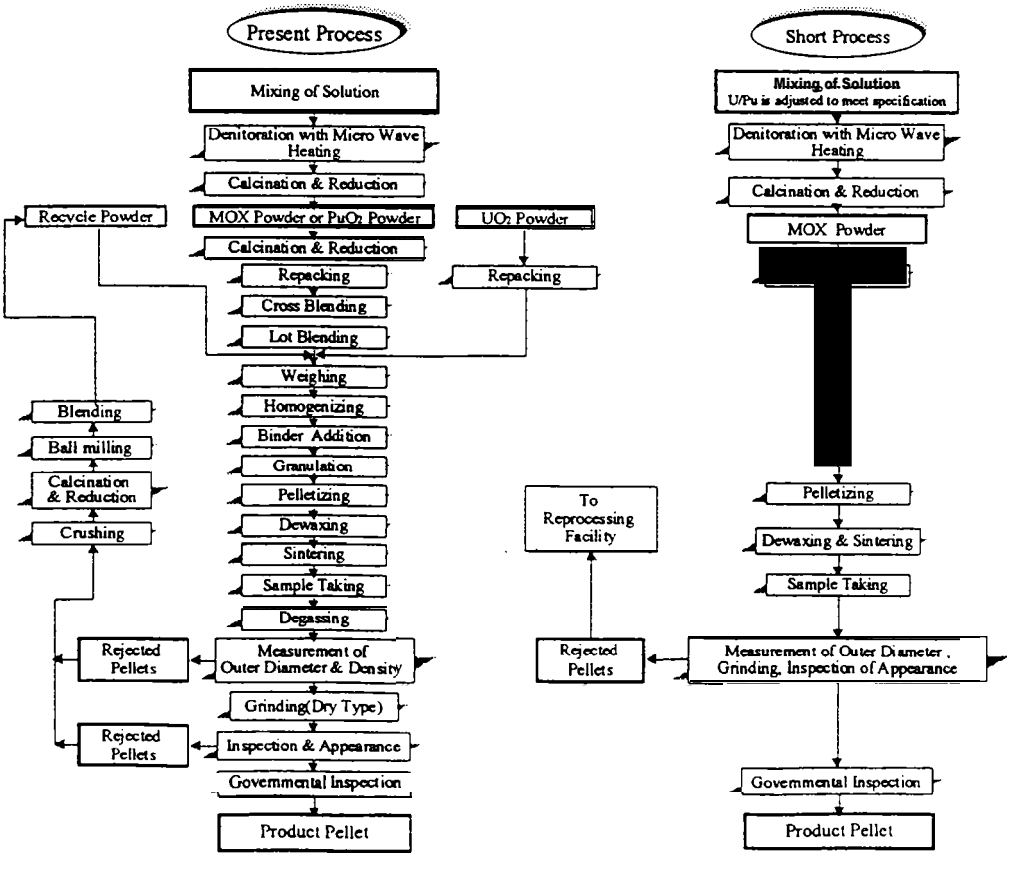

Figure 7. The flow of short process

microwave direct denitrationmethod. Followed by the conversion from mixed solution to the MOX powder, the property of MOX powder is prepared by the calcination and reduction to have certain flowability that can be pelletized into green pellets without granulation. By the introduction of short process, it is possible to omit the powder preparation steps in the conventional one. It is scheduled to find fundamental process conditions to realize the short process by laboratory scale of experiments in four years from 1999.

SUMMARY

Although the PFPF encountered many difficulties such as hold up, low density pellet fabrication, maintenance and productivity being peculiar to the FBR MOX fuel fabrication on a large scale, the PFPF has overcome these subjects and continued the MOX fuel fabrication as discussed in this paper. Through the operation of PFPF, the fundamental technologies required for MOX fabrication on a large scale have been being developed and demonstrated by the JNC. It is recognized that further development efforts are also needed to achieve FBR fuel cycle being capable of competing with LWR fuel cycle even in economics. In case of MOX fuel fabrication, it is essential for the realization of commercially acceptable cost to introduce innovative technologies into the current MOX fuel fabrication through the developments of annular pellet and short process.

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INTEGRATED SOFTWARE PACKAGE FOR NUCLEAR MATERIAL SAFEGUARDS IN A MOX FUEL FABRICATION FACILITY

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Abstract

Since computerized data processing was introduced to Safeguards at large bulk handling facilities, a large number of individual software applications have been developed for nuclear material Safeguards implementation. Facility inventory and flow data are provided in computerized format for performing stratification, sample size calculation and selection of samples for destructive and non-destructive assay. Data is collected from nuclear measurement systems running in attended, unattended mode and more recently from remote monitoring systems controlled. Data sets from various sources have to be evaluated for Safeguards purposes, such as raw data, processed data and conclusions drawn from data evaluation results. They are reported in computerized format at the International Atomic Energy Agency headquarters and feedback from the Agency's mainframe computer system is used to prepare and support Safeguards inspection activities. The integration of all such data originating from various sources cannot be ensured without the existence of a common data format and a database system. This paper describes the fundamental relations between data streams, individual data processing tools, data evaluation results and requirements for an integrated software solution to facilitate nuclear material Safeguards at a bulk handling facility. The paper also explains the basis for designing a software package to manage data streams from various data sources and for incorporating diverse data processing tools that until now have been used independently from each other and under different computer operating systems.

I. INTRODUCTION

Several kinds of advanced high technology Safeguards systems have been implemented and are under developnlent in MOX fuel fabrication plants. During the past decade the International Atomic Energy Agency (IAEA), national nuclear safeguards offices and facility operators have extensively used new tools to improve the implementation of Safeguards. New tools such as advanced accountancy systems (AAS), advanced containment and surveillance systems (AC/S), improved non-destructive assay systems (NDA), and advanced accountancy verification systems (AAVS) were developed.

The basis of an improved Safeguards system at a MOX fuel fabrication facility is formed by an advanced accountancy system that is an on-line real time material accounting system. Such a system can produce declarations of nuclear material inventory and flow within the facility on the basis of data acquired at the time of material transfer. The advanced accountancy verification system (AAVS) makes use of the near real time material accountancy (NRTA) for the purpose of the continuous knowledge and statistical evaluation of the nuclear material in the process area. Improved containment and surveillance systems (AC/S) consist of several kinds of‘ sensors. radiation detectors, monitors, and cameras to detect and register any movement of material, personnel and equipment in an observed area. Non-destructive assay (NDA) systems are mostly based on high-level neutron coincidence counting technique and high-resolution gamma-ray spectrometry. Several specific detector systems have been developed for the verification of nuclear material in the form o!‘ small samples of powder, pellets and solutions, large powder containers, holdup in glove-boxes, scrap outside glove-boxes, fuel pills, fuel assemblies and waste in drums or in large cubic containers. AC/S and NDA systems in combination are used to verify

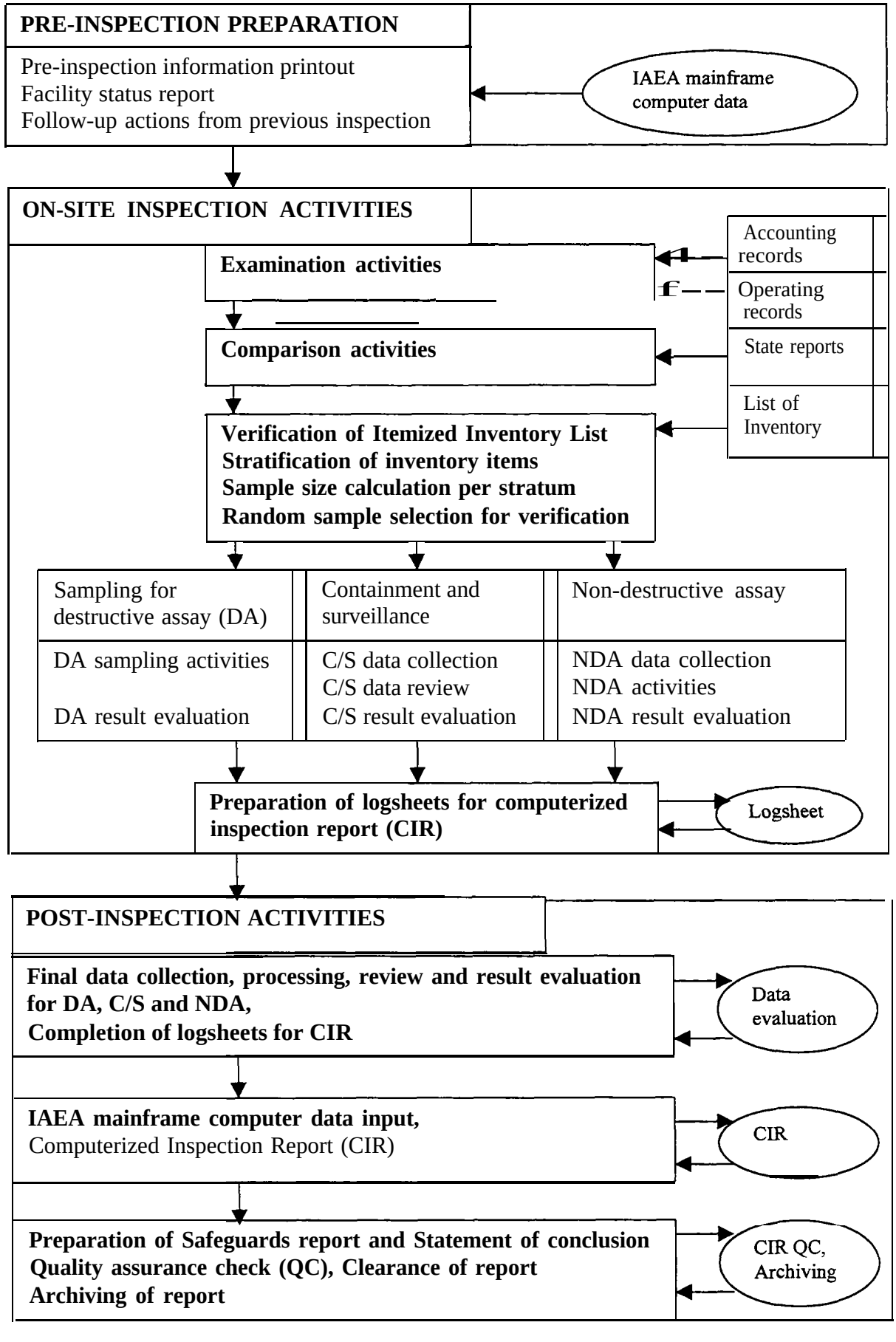

Figure I. Flow diagram of Safeguards inspection activities with a relation to computerized data processing.

automatic nuclear material transfers. The robotics used for automation at large fuel fabrication plants requires that the verification systems are operated continuously in unattended mode or remote controlled.

The IAEA collects and evaluates data at a MOX fuel fabrication plant during inspections in a monthly basis. Most of the on-site data are available in computerized format. The IAEA prepares the statement of conclusion derived from the evaluation results and from reports that were received from State System of Accountancy and Control (SSAC). In-between, an extensive effort is dedicated to processing data for review, evaluation and report the large amount of data collected from various sources before, during and after the inspection. At IAEA headquarters improved measures are applied to confirm that all verification activities met the Safeguards criteria requirements. Performances of the nuclear material verification, nuclear material balances, transfer matching data are evaluated and a thorough quality control is applied on the computerized inspection report. Figure 1 gives an overview of the Safeguards inspection activities. It shows that data in computerized format have to be processed at almost every stage of the inspection until the Safeguards inspection report is completed.

2. SAFEGUARDS INSPECTION DATA PROCESSING

2.1. Safeguards Inspection Data Sources

Besides the General Ledger (GL) and the itemized inventory listing (IIL), the containment and surveillance equipment (C/S) and the non-destructive measurement (NDA) systems are the main sources of Safeguards inspection data in various computerized file formats. Figure 2 shows lists of equipment that is typically used to perform Safeguards verification activities at the feed storage, process area and product storage of a MOX fuel fabrication facility. The sources of Safeguards inspection data can be categorized as follows:

(1) *Information system data:*
At the IAEA headquarters the preparation for any Safeguards inspection comprises collection of historic and recent information about the facility to be inspected. Safeguards criteria, Safeguards Manuals (SM), procedures for non-destructive assay (NDA) as well as containment and surveillance (C/S) instruments, working papers and forms can be retrieved through the IAEA local area computer network from IAEA internal Web sites. The IAEA spends effort on building up an Inspection Reference System (IRS) that provides information related to Safeguards activities summarized for a country.

(2) *Mainframe data:*
The IAEA Safeguards mainframe computer system is currently utilized to prepare a pre-inspection summary information report, a briefing document (RS-1) together with a facility status report (FSR), containment/surveillance (C/S) status and performance report on seals and surveillance systems, performance values for non-destructive assay (NDA) as well as regarding destructive assay (DA).

(3) *State reports:*
Inventory change reports (ICR), material balance reports (MBR), physical inventory listings (PIL) are prepared by the state authorities. The IAEA receives the reports in computerized format and provides the data also in computerized format to the Safeguards inspectors for records and reports comparison purposes and for further evaluation.

(4) *Operator data:*
Usually, the facility operator provides at the beginning of a Safeguards inspection for every material balance area the General Ledger with material balance information,

itemized inventory listing (IIL), near real-time accountancy data (NRTA), material data (like plutonium isotopic data, item location data),and operational records. The format of the data files was negotiated and agreed with between the IAEA and the facility operator. The data is usually provided on floppy diskettes.

(5) *Instrument setup data.*
During a Safeguards inspection several software program system files, configuration data, calibration data are needed to operate all the measurement equipment for nuclear material verification. Support files like operating system setup and configuration files, library files, software drivers, etc. are usually already installed on instrumentation computer hardware, but they must also be held available if needed for re-installation in case a software or hardware problem occurs.

(6) *NDA raw data and C/S review and evaluation data:*
This kind of data is collected from NDA measurement equipment or C/S systems. In case of remote controlled or unattended mode operation the data is automatically collected and stored by the equipment over a certain period of time. Raw data measurement results are retrieved from that equipment periodically. When a measurement is performed in attended mode then the raw data is collected immediately after the measurement. Regarding C/S instruments, a computer file is generated when the C/S information is reviewed and evaluated using computer equipment. Several nuclear material verification systems exist where C/S equipment is used to complement NDA measures for material verification and item identification or moving direction indication. Currently, for Safeguards material verification purposes the IAEA is using about 25 different NDA and advanced C/S systems at a MOX fuel fabrication plant (Figure 2). To be able to operate the various instruments in an easy and comfortable way efforts are needed to find a uniform and standardized approach for a common data format and simple system operation.

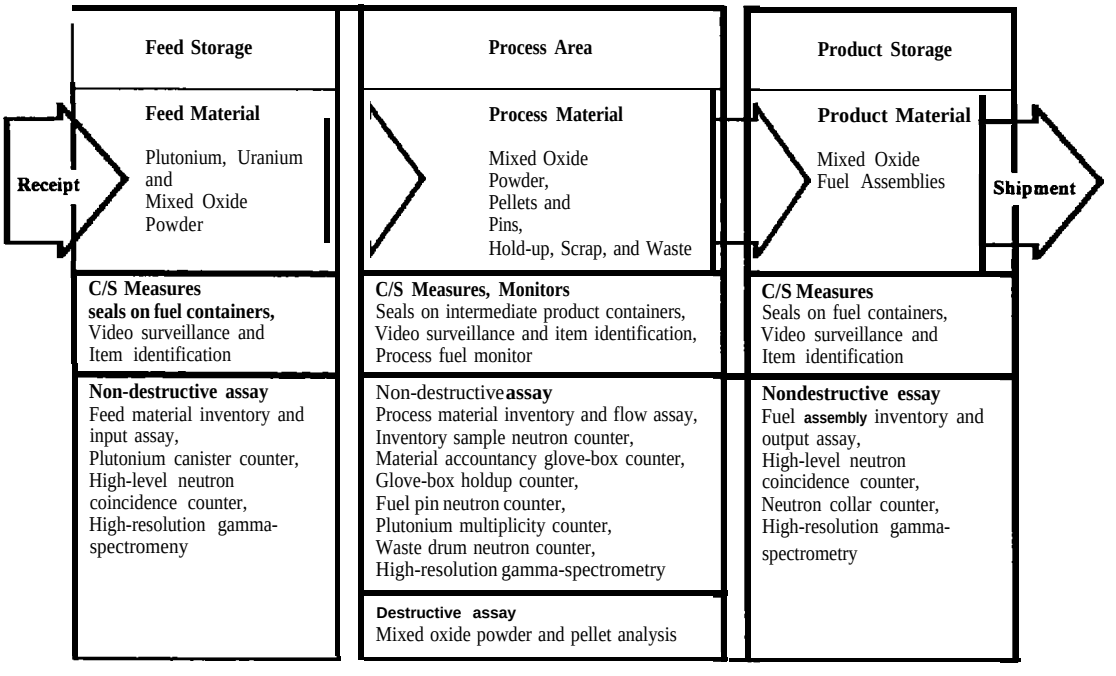

Figure 2. Typical C/S measures and NDA equipment used to perform verification activities at a MOX fuelfabrication facility.

2.2. Safeguards Inspection Data Processing Tools

The term "Safeguards inspection data processing tool" refers to those software applications that have been developed during the past decade to process the data received from the sources that are mentioned under paragraph 2.1 of this paper. While in the past a wide variety of individual software applications was developed over the time and in response to separate needs, the current workload and the complexity of the inspection tasks have reached such a critical level that it is today an essential requirement to develop a software package that integrates the different data components. Inspection data processing tools relevant to the MOX fuel fabrication facility can be categorized as follows.

(1) *Accountancy examination and comparison software:*
This category covers software used for accountancy examination purposes as to validate the nuclear material amount declared by the facility operator. To a certain extent it can check the consistency of data, arithmetical correctness and calculate totals from the accounting records. Comparison software, as the name says, compares the state reports against examined records. An important tool for a large bulk handling facility is the near real time accountancy system software. Typical examples for these kinds of software applications are the In-field support system (IFSS) program and the near real time accountancy (NRTA) software. Material balance evaluation software is used at the IAEA headquarters to derive a conclusion on the nuclear material unaccounted for (MUF) per material balance period.

(2) *Material stratification, sample size calculation, sample selection software:*
Software tools like IFSS, SPAN, NMAX are used for grouping the itemized inventory, so-called material stratification, and calculating the sample size for every verification method. SPAN software is also used to automate announcing items to the facility operator that were selected for verification.

(3) *Instrumentation software, measurement result and C/S review evaluation software:*
NDA equipment works with computer software like HLNC, INCC (LANL software, Reference l), SR-Collect/MIC/SR-Review (LANL software, Reference 1) for neutron measurement devices, and PUIS for high-resolution gamma-ray systems. Containment and surveillance equipment makes use of MOSS software, while review and data evaluation are performed using MORE software. Remote monitoring requires a new type of software that works fully automatically.

(4) *Other IAEA Safeguards mainframe, NDA, C/S and DA data collection, review, processing, evaluation, reporting, archiving and retrieval software systems:*

- IAEA mainframe software system
- Interface to and from the IAEA mainframe computer system
- Logsheet software for IAEA mainframe data input
- Equipment inventory and performance software
- Non-destructive assay (NDA) data review and evaluation software
- Containment and surveillance data review and evaluation software
- Destructive assay (DA) data review and evaluation software
- NDA and DA performance evaluation software
- Data archiving and retrieval software
- Generic and facility specific instrument and software procedures and manuals
- IAEA Safeguards intranet Web site.

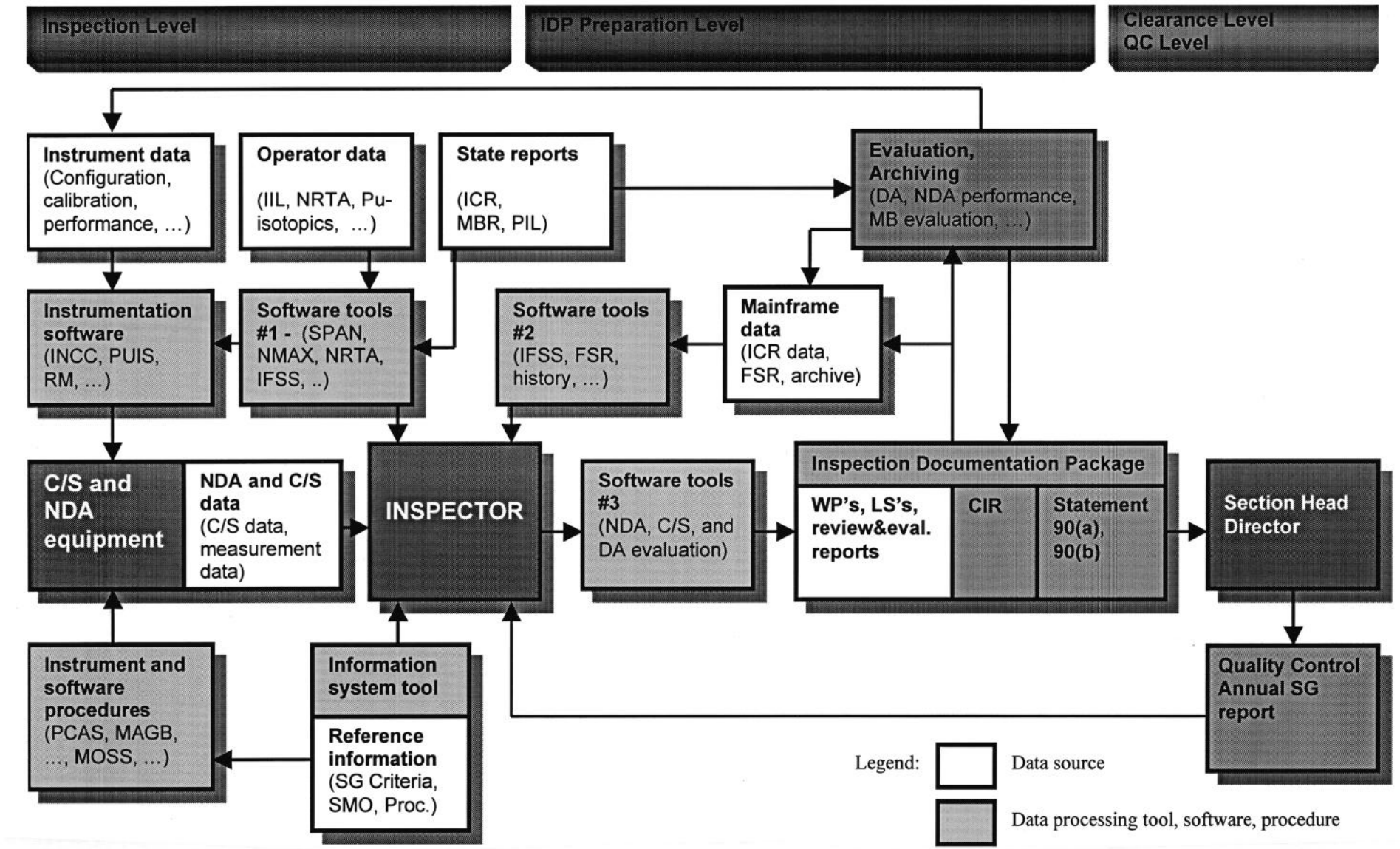

Figure 3: Safeguards inspection data sources, data processing tools and data streams

2.3. Safeguards Inspection Data Streams

Several individual software applications were implemented for routine Safeguards inspection use at MOX fuel fabrication plants. Usually, individual software versions work independently from each other. Often, lack of data transfer between different computer programs requires duplicate and sometimes even repeated manual data entry. Different user interfaces increase the effort that has to be spent on training for Safeguards inspectors operating the software. To improve the situation the data streams were analyzed to identify the relations between data sources and data processing tools that have to be dealt with in the course of a Safeguards inspection.

For a fuel fabrication facility Figure 3 gives a simplified view of how complex the relations between the various data sources and software tools appear. The Safeguards inspector collects data that has to be processed using software applications as mentioned under paragraph 2.2 of this paper. Mostly, direct data streams exist from the data source through the processing tool to the inspector, but also often part of certain data sets are used for input to more than one software application. For example, individual material data is needed for accountancy and stratification purposes, but also as input to the verification measurement systems. Data streams can split and at certain points they merge again. Data streams exist in parallel before they merge. At a large bulk handling facility the Safeguards inspectors as members of a team perform several verification activities at the same time.

Verification data is collected at different locations in the facility and ideally it is moved to a database management system for further evaluation. A specific software tool controls the timing of individual software applications to be invoked. It also guides and coordinates collection, transfer and processing of data. Feedback of data from the IAEA Safeguards mainframe has also to be taken into account for the preparation of software tools regarding follow-up actions to be carried out at a future inspection. With a quality assurance system incorporated in the software package an important data stream is characterized by re-analysis, re-evaluation and measures to correct errors in the reports.

2.4. Safeguards Inspection Data Reporting and Archiving

Computerized Safeguards inspection data is archived on the IAEA Safeguards mainframe computer. An efficient integrated Safeguards inspection software package for a fuel fabrication facility will also include a convenient system regarding data reporting, archiving and a comfortable method to retrieve archived data. An inspection documentation package (IDP) is prepared that incorporates all the data that cannot be computerized as well as reports printed from the database.

3. APPROACH FOR INTEGRATED SAFEGUARDS INSPECTION DATA PROCESSING

In early 1997, the IAEA initiated a project to develop software tools integrated into a software package that assist the inspectors in performing their Safeguards inspection activities. The software shall use a common data repository to avoid duplications, cumbersome exchange of data between different tools, and re-entry of the same information. All the tools shall be implemented through a common user interface. Where applicable commercial software packages shall be integrated into the software project. By means of an integrated software package it is intended to further automate the inspection activities and to reduce the manpower effort. In the year 1998 a review was performed by L&T Information Technology Limited (Reference 3) concluding the following guidelines for the development of an integrated Safeguards inspection data processing package.

3.1. Common Database

A unique format has to be defined for data processing and storage in the mainframe computer of the Safeguards Department. Modules programmed following the standard will accept data in a consistent format. Data output from a module will also follow the standard format as close as possible. A database server shall run a central relational database management system for the integrated software package. Software that is loaded onto portable computers to perform a certain part of the integrated software package can cover a reduced database system which is sufficient to run the specific task.

3.2. Modular Software Structure

A standard modular design shall be used to facilitate integration of software components from different developers. Also maintenance of the new software product will be easier for software that has a modular design.

3.3. Standard User Interface

An integral part of the software package will be a user interface that links individual modules together. The user interface should be easy to learn and easy to use. It shall guide the user through the various tasks and inspection activities. The inspection data shall be processed mostly automatically with a minimum of user interference. In addition, a wide range of software menu options shall be provided for experts to resolve individual problems, if needed.

4. SUMMARY

Computer hardware and software is utilized to a large extent for performing Safeguards inspection activities at all stages. Many data in computerized format have to be collected and processed starting with pre-inspection preparatory activities through on-site nuclear material verification and book examination until post-inspection activities and finishing with a Safeguards statement of conclusion. The current collection of Safeguards software applications confines mostly stand-alone versions. Some of the programs are partially working under old operational systems, like DOS, without data transfer options between individual applications and lacking convenient user interfaces. The implementation of advanced systems for nuclear material accountancy, improved material verification equipment and enhanced data evaluation methods require a new, standardized and uniform approach regarding Safeguards data processing. The IAEA is currently developing a complex computer software model for a MOX fuel fabrication facility that will integrate data collection, evaluation and reporting for Safeguards inspection purposes in a modular structure using a database system combined with a comfortable user interface. This new Safeguards tool is to meet also the requirements from the Additional Safeguards Protocol.

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SOME ASPECTS OF A TECHNOLOGY OF PROCESSING WEAPONS GRADE PLUTONIUM TO NUCLEAR FUEL

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The concept of Russia to use fissile weapons-grade materials, which are being recovered from nuclear pits in the process of disarmament, is based on an assessment of weapons-grade plutonium as an important energy stuff intended for the usage in view of fuel of nuclear power facilities. This valuable raw-material is the national property of Russia and its effective usage on behalf of people is a significant statuary task.

However, at the path of involving plutonium excessive from the purposes of national safety into industrial power engineering there are a lot of problems, from which effectiveness and terms of its disposition are being dependent upon. Those problems have political, economical, financial and environmental character.

In this report we would like to outline several technological problems of processing weapons-grade metallic plutonium into MOX-fuel for reactors based on thermal and fast neutrons. In particular, we will touch the issue of conversion of the metal into dioxide from the viewpoint of fabrication of pelletized MOX-fuel.

The processing of metallic weapons-grade plutonium into nuclear fuel is rather complicated and multi-stage process, every stage of which is own production. Some of the stages is absent while production of MOX-fuel, for instance a stage of the conversion, i.e. transferring of metallic plutonium into dioxide of the ceramic quality.

At this stage of plutonium utilization some tasks must be resolved as follows:

- As a result of the conversion, a material purified from ballast and radiogenic admixtures have to be obtained. This one will be applied to fabricate pelletized MOX-fuel going from morphological, physico-mechanical and technological properties.

- It is well known that metallic gallium, which is used as an alloying addition in weapons-grade plutonium, actively reacts with multiple metals. Therefore, an important issue is to study the effect of gallium on the technology of MOX-fuel production, quality of the pellets, as well as the interaction of gallium oxide with zirconium and steel shells of fuel elements depending upon the content of gallium in the fuel.

The rate of the interaction of gallium oxide, containing in MOX-fuel, with zirconium alloys and stainless steels is not known.

The knowledge of its acceptable content in fuel will permit to choose an optimum variant of plutonium purification from gallium. Therewith, it is possible to guess that the acceptable content of gallium in plutonium dioxide used for fabrication of the fuel for the reactors on fast and thermal neutrons can be different.

To furnish the environmental safety of MOX-fuel during its storage, transportation and production, plutonium dioxide must be refined from americium-241, which is being accumulated in weapons-grade plutonium on the account of decaying plutonium-24 1.

At present, in Russia there are several technologies under study, which principally allow to resolve the tasks mentioned above.

All technologies have advantages and disadvantages, but neither of them can be considered complete since in each one there are fields not to be studied sufficiently, and mainly in many processes a final result, the quality of plutonium dioxide, is not assessed.

I would like to tell you more in detail about some technologies of the conversion, which are under study by scientific centers of MINATOM.

Aqueouschemical technology

The technology is separated upon several variants hinging upon the way of the dioxide fabrication.

Oxalate precipitation (Appendix I). This technology has been studied better; its merits and limitations are given in the Table 1.

Merits and limitations of the technology of oxalate precipitation

Merits	Limitations
1. Long-term experience of dioxide plutonium fabrication at a full scale for the eventual metallurgical production (the presence of equipment). 2. The availability of experience of using such plutonium for fabrication of MOX-fuel for the reactors BN-350 and BN-600 by a selective choice of the material parties. 3. The feasibility of correcting properties of an initial dioxide powder at the stage of fabrication of a press-powder.	1. Not sufficient amount of R&D works with the purpose of selecting modes of fabrication of a powder of the ceramic sort. 2. High dispersity, dust formation, high adhesive ability. 3.Instability of the powder features that is defined by the complexity of parameters of the technological modes (composition of solutions, temperature, duration of the process, intensity of blending, etc.) 4. Liquid waste enriched by plutonium (up to 200 mg/l) must be processed additionally.

Ammonium precipitation (Appendix 2a) or co-precipitation of plutonium and uranium *(Appendix-t* 2b) with parallel involving of surface-active substances into the solution. This method allows to obtain a prepared master-blend with a regulatory ratio of PuO_2 and U02.

The technology of ammonium precipitation and especially co-precipitation from our point of view is more favorable because it allows to obtain the prepared master-blend. On the account of adding the surface-active substances, the powders do not raise dust. Basic merits and drawbacks of the technology of ammonium precipitation are presented in the Table 2, but the technology of ammonium co-precipitation – in the Table 3.

Table 2.

Merits and limitations of the technology of ammonium precipitation with the surface-active substances

Merits	Limitations
1. Obtaining of agglomerates of plutonium dioxide with the given dimension and having insignificant dust-formation, good fluidity, low adhesion and technological characteristics according to technical requirements. 2. Intensification of the process of powders fabrication due to formation of quickly filtrating sediments. 3. The possibility of using regular radiochemical equipment. 4. Low content of plutonium in the master taps (<5 mg/l) 5. Preliminary positive experience of using such powders to fabricate pellets by the technology of mechanical blending.	1. Not sufficient amount of R&D works with the purpose of selecting modes of fabrication of a powder of the ceramic sort. 2. The absence of special equipment for the stages of precipitation and granulation. 3 The necessity of a stage of preliminary reduction of plutonium in the solution by the tree-valent condition before precipitation.

Table 3.

Merits and limitations of the technology of ammonium co-precipitation with the surface-active substances

Merits	Limitations
1. The availability of using such a type of the material to fabricate MOX-fuel for the reactors BN-350 and BN-600 at the PA "Mayak". 2. A great volume of R&D works to define technological modes providing fabrication of the powders with required features. 3. Obtaining of agglomerates of plutonium dioxide with the given dimension and having insignificant dust-formation, good fluidity, low adhesion and technological characteristics according to technical requirements. 4. Intensification of the process of the powders fabrication due to formation of quickly filtrating sediments. 5. The possibility of using regular radiochemical equipment. 6. Low content of plutonium in the master taps (<5 mg/l). 7. The presence of positive laboratory experience of using the co-precipitated master-blend to fabricate fuel for the reactors VVER. 8. Reduction of a technological process of pellets fabrication.	1. The absence of special equipment for the stages of precipitation and granulation. 2. Some difficulties of correcting properties of an initial granulate of the mixed oxides at the stage of pellets fabrication.

The aqueouschemical technology in all variants permits to purify plutonium from gallium, americium and considerable amounts of ballast admixtures almost completely. It is the effective, but rather expensive technology. Along with a complicated process of metallic plutonium dissolution, its main limitation is the formation of considerable amounts of waste basically in view of aqueous tailings. But, one should note that the technology of their disposal is developed and mastered by the Russian atomic industry.

A pyrochemical technology (Appendix 3) is more reliable from the viewpoint of the absence of such waste. It is proposed to use hydration of metallic plutonium and the conversion of hydride into dioxide. Therewith, the hydride can be transferred to nitride and then to dioxide.

However, the pyrochemical process has a lot of uncertainties ("white blots") related to the absence of purification from americium and ballast admixtures, ineffective purification from gallium and high potential explosive danger caused by the presence of hydrogen and oxygen in the same process. Properties of the dioxide obtained by means of a dry technology must be studied more diligently. Our Institute does the very subject. In particular, we began to study the behavior of gallium at all stages of MOX-fuel production with the aim to work off the technological regimes that allow to disregard gallium from the fuel and on the other hand to provide therewith the formation of the dioxide of the ceramic sort.

By the way, **a combined method of the conversion of metallic plutonium,** in which merits of the pyrochemical and aqueouschemical technologies are being used, is under research (Appendix +). A poor place of the aqueouschemical technology is the preparation of metallic plutonium to its dissolution (the necessity of plutonium grinding) and also a labor-intensive process of the dissolution.

Finally dispersed products of the pyrochemical process is easily dissolved in nitric acid with an addition of fluoride ion. In the forth, a merit of the aqueous processes as effective extraction of plutonium from gallium, americium and the ballast admixtures and warranty fabrication of plutonium dioxide of the ceramic sort is used.

A pyrochemical technology of the conversion of metallic plutonium in molten salts (Appendix 5) is being developed. This technology is worked off by RIAR (Dimitrovgrad) well in order to obtain vibro-pressed fuel, which is applied in the reactor BOR-60 and has been tested in the industrial reactor BN-600. However, the technology of fabrication of the dioxide of the ceramic sort for pelletized MOX-fuel whilst is absent. Works are under way in RIAR to realize this technology. A complicated problem of the pyrochemical technology of the conversion is to develop a technology of the complex utilization of wastes of this process from the standpoint of its economical and environmental optimization.

MOX-fuel obtained by different technologies of weapons-grade plutonium proessing must pass pre-reactor tests, a reactor verification in a research reactor, and post-reactor investigations. Without this complex of the works it is impossible to get a license of GAN to load the fuel into commercial reactors. By this time, tests have been made in breeders of pelletized MOX-fuel, fabricated by the aqueouschemical technology (oxalate precipitation, ammonium co-precipitation), and vibro-pressed MOX-fuel, fabricated by the pyrochemical technology. Preparation to test pelletized MOX-fuel for light-water reactors is in progress.

The technologies, which are under study and will furnish fabrication of the dioxide met to the requirements of the ceramic sort with purification from gallium, americium and the ballast admixtures, will be assessed as potentially applicable for industrial aims. However, to select one or two of the technologies it will be necessary to make comparable objective appraisals of the technologies upon the following parameters:

- Economical efficiency of the process;
- The presence and complexity of equipment;
- The volume of waste and difficulty of its reprocessing;

- Environmental safety of the technology, -including nuclear, radiation, explosive and fire ones;
- The availability of engineering infrastructure needed to realize the technology at industrial sites;
- The necessity and volume of investments needed to create an industrial production of the technology;
- A series of other aspects, including social.

Going from those deliberations, before making any decision about enacting MOX-fuel production it is necessary to scrutinize variants of the technologies, to assess the quality of the product, a possibility and terms of licensing the fuel, to implement engineering feasibility studies of the technologies and finally upon the results of these works to accept an optimum decision.

We believe that it is advisable to tell a few words about another aspect of plutonium reprocessing into the nuclear fuel. To utilize plutonium more reasonably there is a need to develop new types of the fuel, which would allow to decrease reproduction of plutonium dramatically or rather completely. A technology of the fuel with the content of plutonium up to 45-50 wt% (and without uranium) with inert matrices (diluants) of the ceramic or carcass type is being studied in our institute.

The fuel of the ceramic sort is plutonium mononitride or monocarbide. Zirconium mononitride or monocarbide can be used as a diluant.

A laboratory scale technology of fabrication of fuel cores from 54.5 % wt PuC and 45.5 % wt ZrC has been developed. An experimental set of the cores was produced; reactor tests of experimental fuel elements in the reactor BOR-60 at the linear output of 400-500 Wt/cm up to burn-up equals to 8 % wt. All elements retained hermetically and were permissible for eventual exploitation. Swelling did not exceed 1 % wt per percent of the burn-up. The yield of gaseous products of decaying was less then 20 % wt of the forming quantity. Only local fields of carbonizing the shell were identified.

At present, an experimental set of fuel elements made from plutonium mononitride and zirconium nitride (40 o/o wt PuN, 60 % wt ZrN) for reactor tests in the BOR-60 are being fabricated.

A material on the base of a porous carcass from different refractory high-temperature compositions (zirconium carbide or nitride, or its solid solutions with incorporation of plutonium) is the other variant of the fuel with the high content of plutonium intended for its use in reactors-burners.

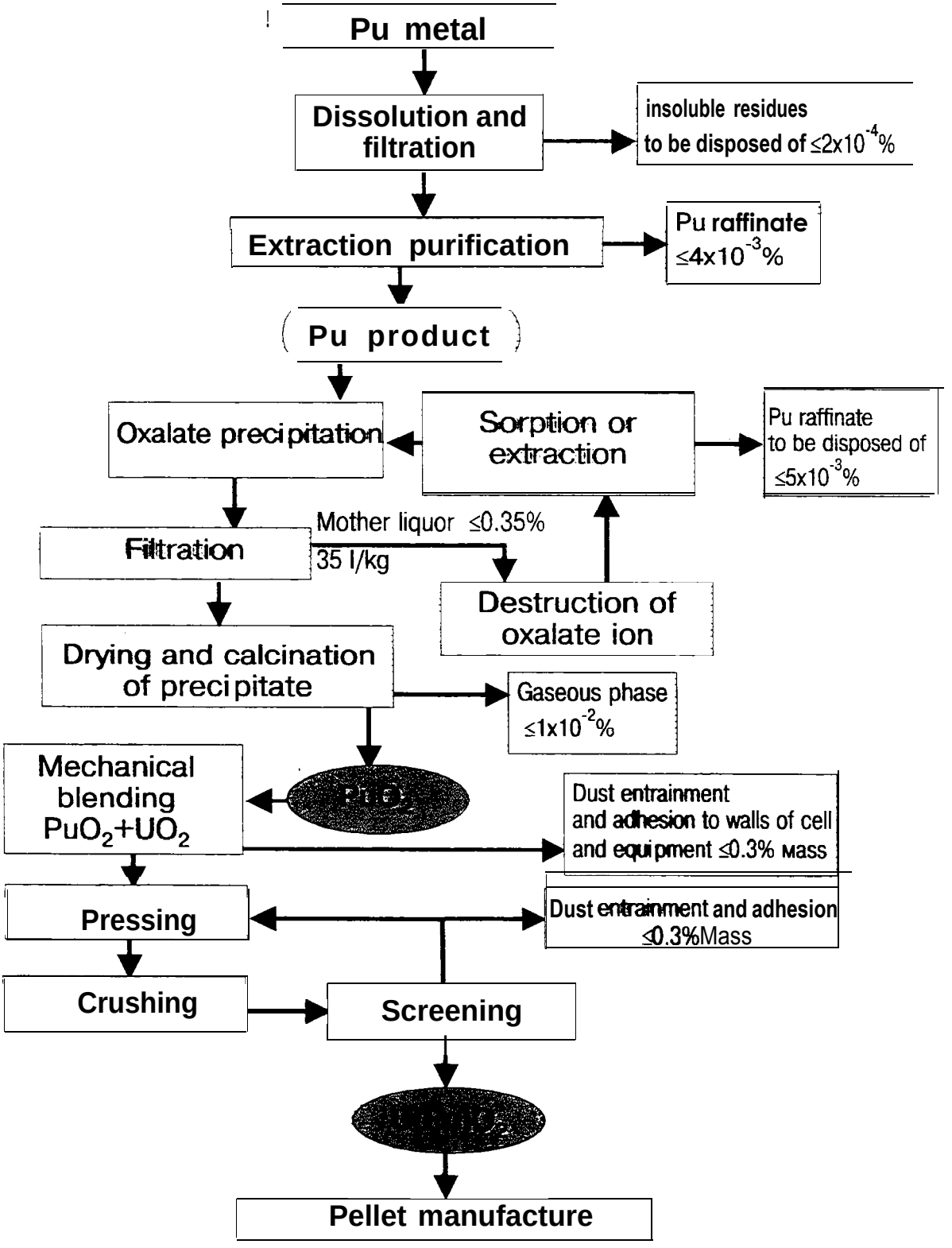
The porous carcass from zirconium carbide or nitride (porosity is within 20-80 % vol) is impregnated by plutonium-containing solutions, from which plutonium dioxide is being formed during the thermal destruction.

This way can also be used for transmutation of long-term high-radioactive wastes, containing minor actinides (neptunium, americium, and curium). It permits to obtain in porous of the cores a mixture of oxides of different nuclides. By the way, there is a possibility of profiling of the content of a fissile component upon the radius and longevity of the core, therewith accurate dosing of the fissile material is provided by parameters of the technology.

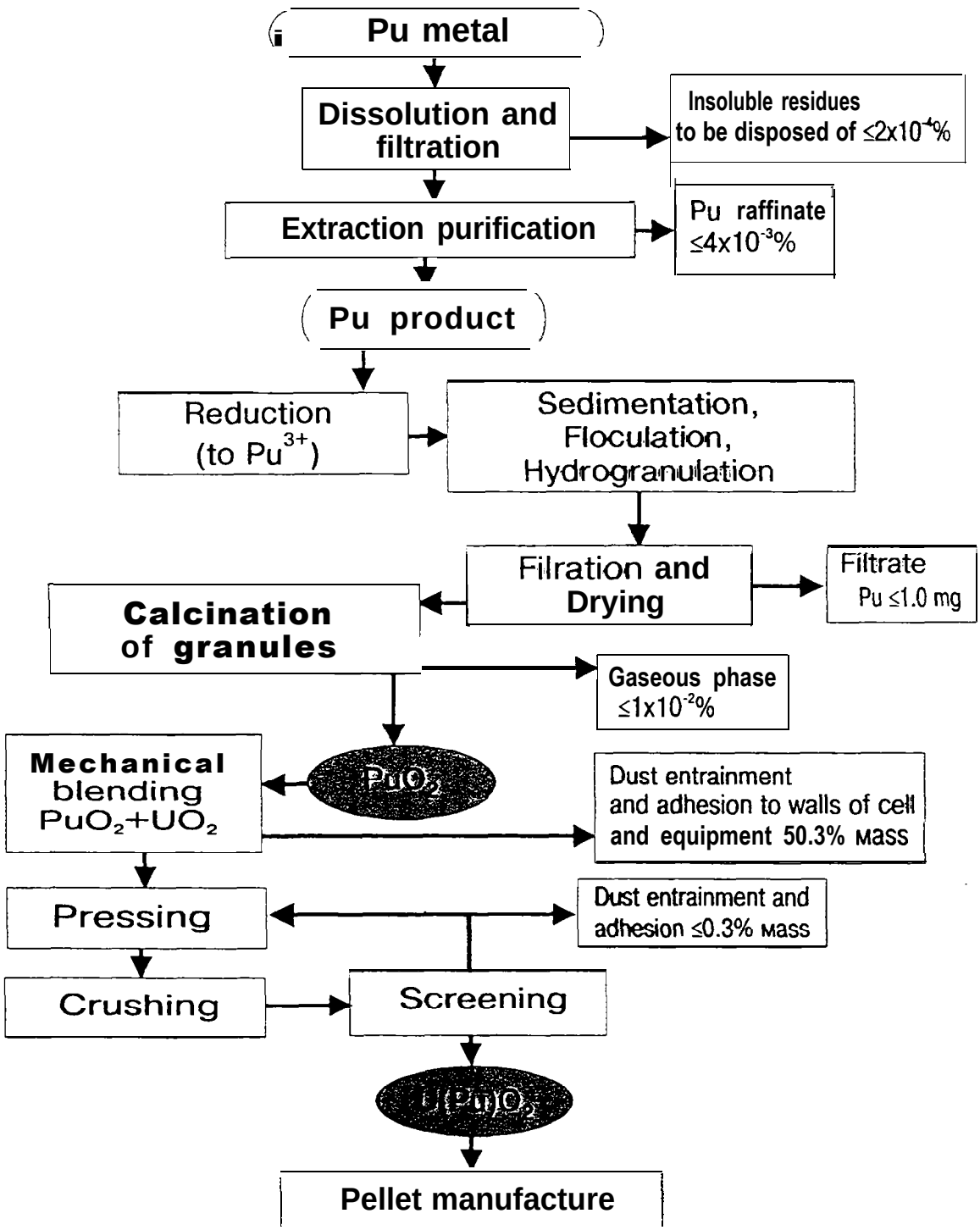
Right now we are conducting the works on the post-reactor study of these compositions.

Appendix I

OXALATE PROCESS SCHEMA OF WEAPON'S PU METAL CONVERSION TO MOX-FUEL

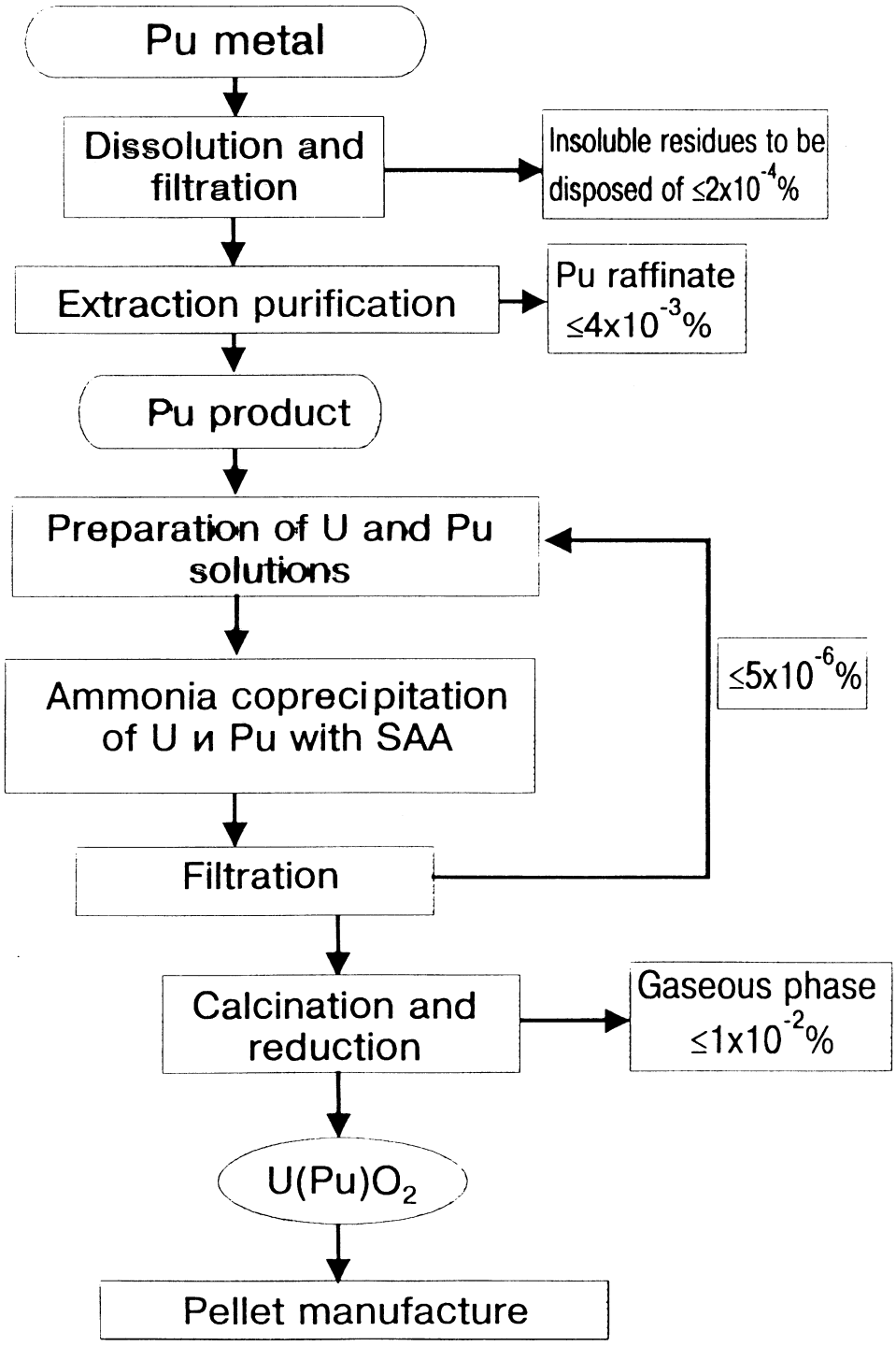


AMMONIUM PROCESS SCHEMA OF WEAPON'S PU METAL CONVERSION TO MOX-FUEL



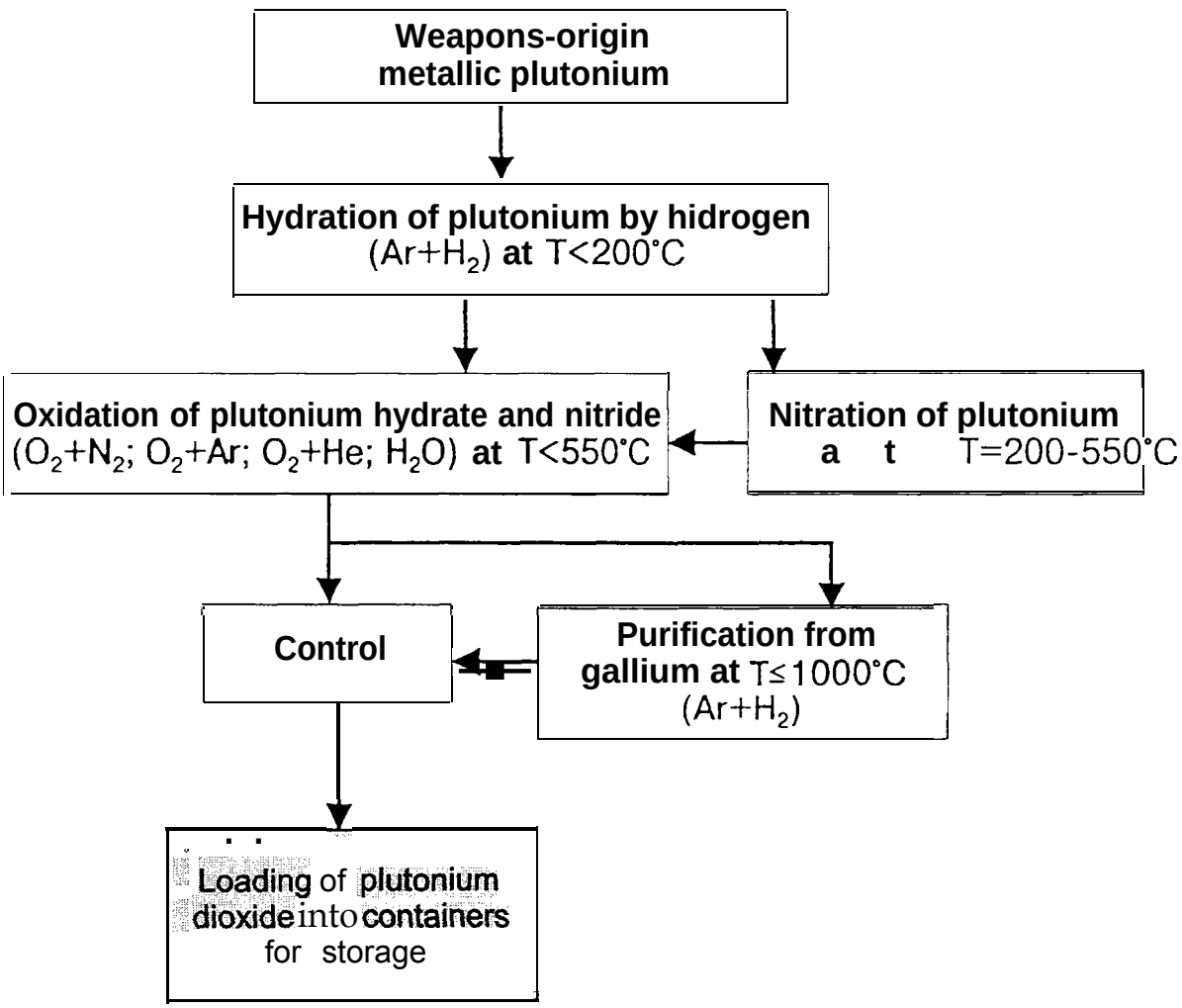
Appendix 2b

«GRANAT» PROCESS FLOW SHEET OF WEAPON'S GRADE PU CONVERSION TO MOX-FUEL (ammonia coprecipitation of U and Pu with SAA)



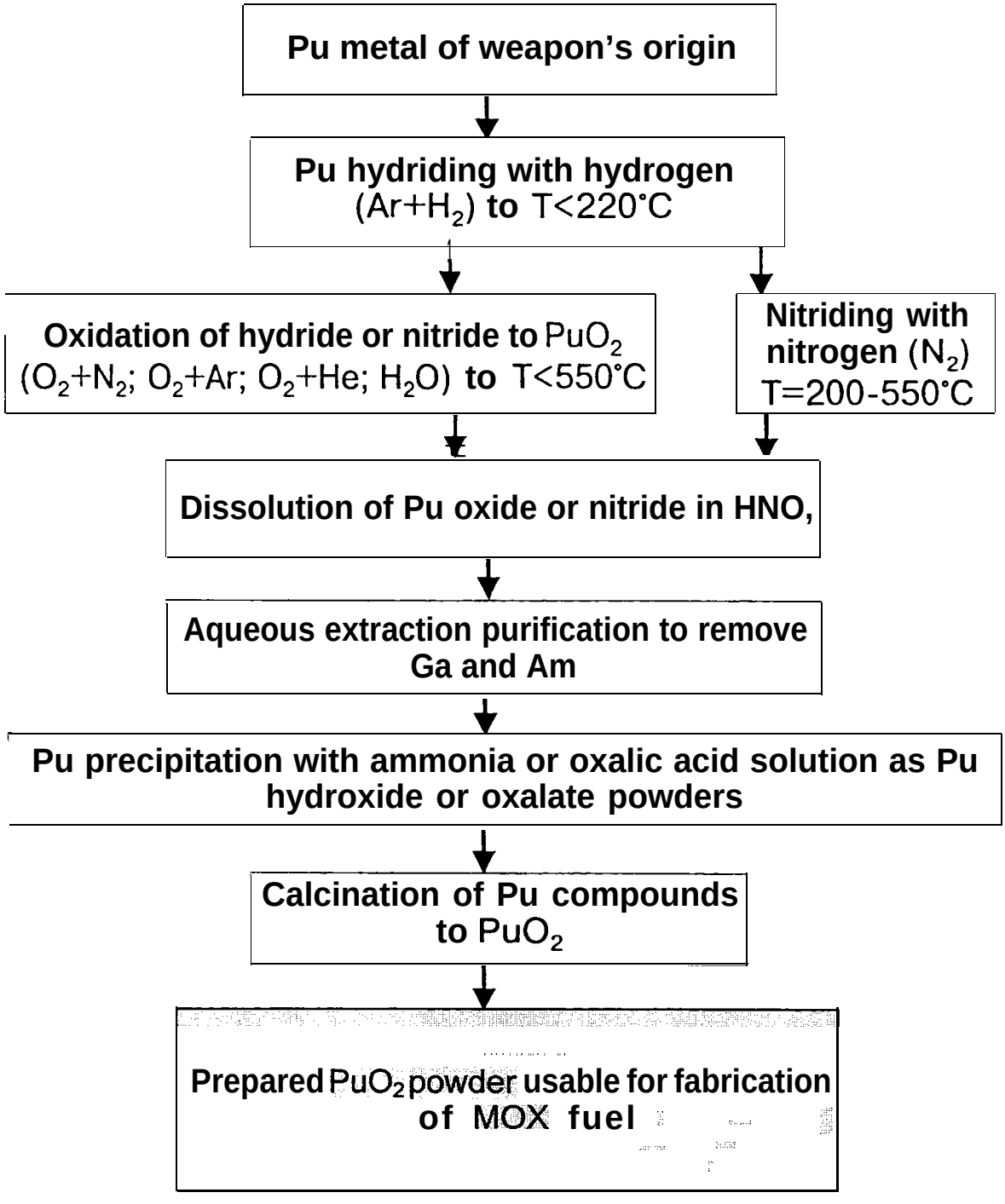
Appendix-3

A FLOW SHEET OF A PYROMETALLURGICAL PROCESS OF TRANSFERRING WEAPONS-GRADE PLUTONIUM INTO PLUTONIUM DIOXIDE

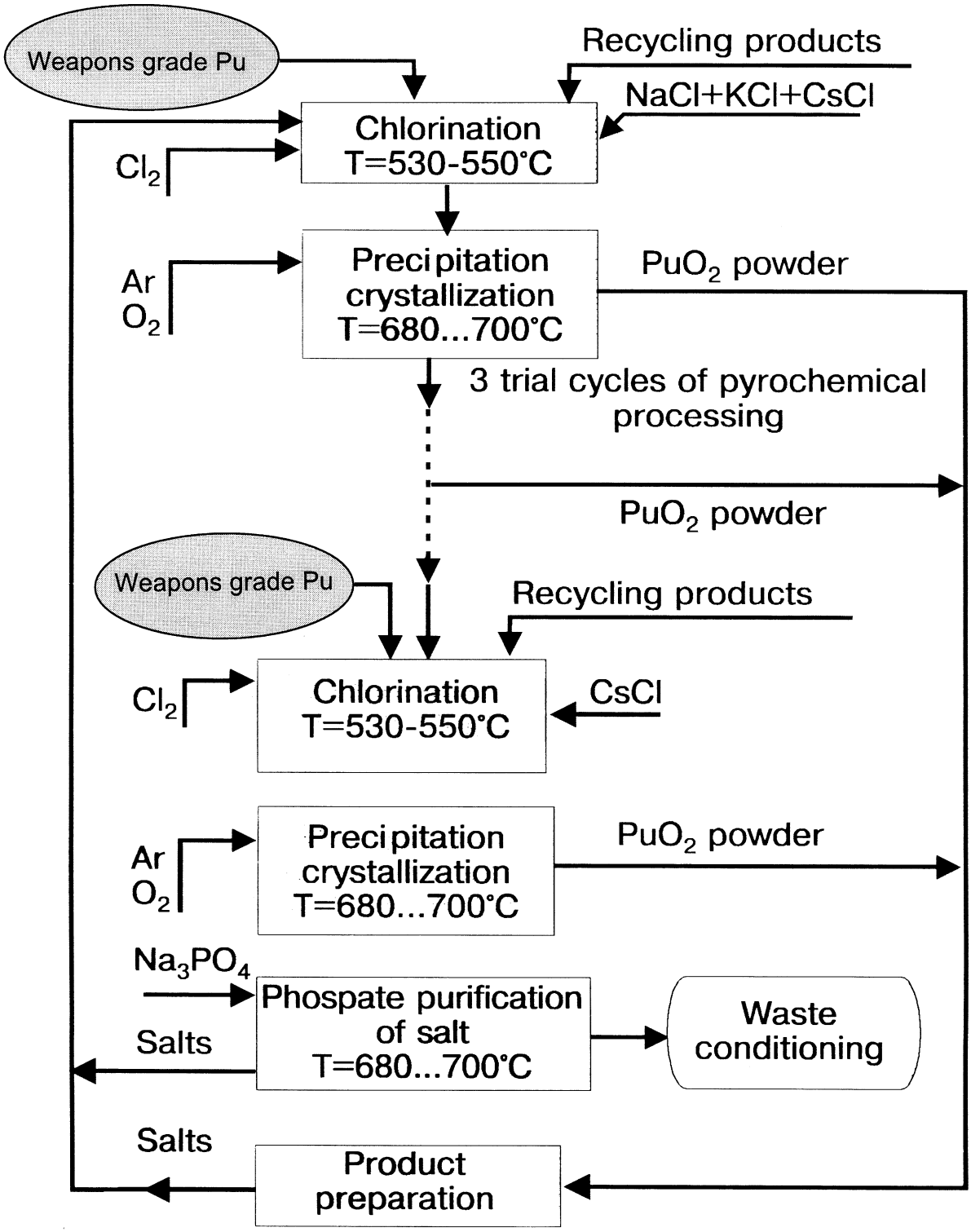


Appendix 4

PROCESS FLOW SHEET of PuO_2 preparation from weapon's crade Pu (combined schema)



Appendix 5


Weapons grade Pu
Recycling products
NaCl+KCl+CsCl
Chlorination
T=530-550°C
Cl_2
Precipitation
crystallization
T=680...700°C
Ar
O_2
PuO_2 powder
3 trial cycles of pyrochemical processing
PuO_2 powder
Weapons grade Pu
Recycling products
Cl_2
Chlorination
T=530-550°C
CsCl
Precipitation
crystallization
T=680...700°C
Ar
O_2
PuO_2 powder
Na_3PO_4
Phospate purification
of salt
T=680...700°C
Salts
Waste
conditioning
Salts
Product
preparation

FURTHER DEVELOPMENTS OF "GRANAT" TECHNOLOGY OF GRANULATED U-Pu FUEL

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Abstract

The «GRANAT» technology based on the uranium and plutonium hydroxide co-precipitation and granulation with surfactants present followed by heat treatment was subjected to a representative large-scale trial by fabricating U and Pu oxide powder pellets and fuel rods for BN-350 and BN-600. The acquired results corroborated the high quality of the powders and pellets. Several advantages of the process were revealed in comparison to the traditionally used processes of dioxide mechanical stirring. Primarily they are the elimination of the fine dispersion operation, reduced dust formation, high degree of U and Pu homogenization, high (96% mass) yield of specified quality pellets. The further investigations were aimed at increasing the competitiveness of the «GRANAT» process via simplifying the equipment used, reduced number of process operations and lower waste arising. The most important results are one-stage heat treatment of granular precipitate in a reducing atmosphere that excludes individual operations of drying and calcining; mother liquor reprocessing by the catalytic decomposition of ammonium nitrate and the complete utilization of the products of this operation within the «GRANAT» process flow sheet. It is demonstrated that the introduced modifications do not result in the property degradation of oxide powders and pellets.

1. INTRODUCTION

The main three criteria that are to be used when choosing the technology of the mixed U and Pu oxide preparation were formulated more than 20 years ago, they are: completeness of fuel solubility i n HNO_3; minimal dust formation by intermediate products; feasibility of process automation [1]. Currently, those criteria as applied to the restructured and newly emerging U-Pu fuel production facilities have not only become obsolete but are more urgent.

The «GRANAT» technology based on the U and Pu co-precipitation with ammonium hydroxide from HNO_3 solutions and the granulation of the precipitate immediately in the mother liquor with polyacrylamide (PAA) as a binder conforms to the requirements to a greater extent:

The fuel manufactured from granulated U-Pu dioxides is completely solvable in HNO_3 since the homogeneous distribution of U and Pu is provided at the stage of mixing U and Pu solutions prior to the precipitation;

U-Pu dioxides prepared as granules practically eliminate dust produced by the intermediate product while the proper ceramic properties of the granules eliminates the need in operations that mostly produce dust, namely, pre-pressing, grinding and screening that are typical of individually processing the oxide powders;

The fractional composition of the prepared oxides consists to 94-95% of granules more than 200 pm (10^{-10} m) in size, having proper flowability, adequately high bulk density (e.g., at the Pu mass fraction of 27.5% the flowability of granular dioxides is $3.2\pm$ 0.2 g/s and the bulk density is 1.5 ± 0.1 g/cm^3); the operations dealing with such a material are readily automatic.

Aside from this, also other merits of this technology are to be mentioned.

The precipitated finely dispersed particles of U and Pu hydroxides PA.4 bound to agglomerates promote a drastic increase in the phase separation rate. The granulated precipitate is easily separable from the mother liquor. The sizes of the granules may be controlled via varying the PAA introduction and the intensity of suspension mixing. The quantitative precipitation of U and Pu with ammonium hydroxide ensures the specified composition of the U-Pu fuel without any other adjustments.

However, the investigations carried out by us give an evidence that not all the potentialities of this technology have yet been exhausted.

2. DESCRIPTION OF INVESTIGATIONS

2.1. Instrumentation of operations

At first the conditions of the precipitated flocs granulation were worked out and optimized in rotary drum-type equipment. The semi-commercial batches of fuel were produced in a specially designed for the purpose nuclear safe slotted apparatus with pulsed mixing. Its design allowed the reproduction to some extent the hydrodynamics of the drum-type equipment.

Further instrumentation studies of the precipitation, flocculation and granulation were aimed at choosing the standard apparatus. As a result reactors-precipitators of different designs were tested. The experiments were implemented not only under the laboratory conditions. The favorable results were also acquired at a radiochemical plant using the standard reactor-precipitation.

The results gave an evidence that for the implementation of precipitation, flocculation and granulation in the «GRANAT» process essentially any standard reactor with the controlled mixing intensity is usable. In progress are developments of precipitation-granulation process and equipment to be continuously operated.

2.2. Direct Reduction of Granular Precipitate

According to the realized «GRANAT» process flow sheet the operations of granular precipitate drying, calcining and reducing were implemented successively in three stages. First, the granular precipitate was dried with hot air on the Nutch-filter. Then the granules were calcined in air. After the calcination uranium and plutonium were reduced to dioxides in an argon-hydrogen atmosphere. Initially this structure of the process was reasonable. The multistage mode provides the flexibility in controlling the properties of the resultant product, particularly, it ensures the complete removal of carbon the source of which is PAA.

The implemented investigations, and the chosen conditions of the reductive heat treatment revealed that those three operations might be replaced by a single one, namely, the heat treatment of the precipitate in $Ar\text{-}H_2$, the so-called «direct» reduction. It was established that under those conditions at 700-800 °C the destruction of PAA to form volatile components proceeds essentially at the same rates as in air and the carbon content of the final powders is 10-2% mass which complies with the established requirements.

It is known that the direct reduction option is used to produce UO_2 by the ADU process /2/. However, the final press-feed characteristics needed for pelletizing are afforded by an additional operation of dry granulation. This operation is not available in the «GRANAT» process; the prepared dioxide powders are immediately subjected pelletizing.

2.3. Utilization of waste

Two types of waste arise from the «GRANAT» process, namely, filtrates from U and Pu hydroxide precipitation that contain some 1.4 mole/l NH_4NO_3 and less than 5 mg/l U and Pu acid solutions arising from the periodic cleaning of the equipment.

As for as the main type of waste, i.e., mother liquors from hydroxide precipitation - the method of catalytic decomposition of nitrites to nitrogen gas in the closed process cycle was developed and tested using actual materials. The method was described in our paper submitted to «GLOBAL» [9]. The evaporator bottoms of this operation does back to the head end of the «GRANAT» process.

Acid rinsing solutions rich in U and Pu are not large in the volume. After rather simple conditioning (boiling in presence of H_2O_2 to destruct PAA) they are added to the U and Pu solutions in the initial solution preparation unit of the «GRANAT» process.

2.4. Modified NGRANATH Process Flow Sheet

The results of the implemented investigations give grounds to recommend the «GRANAT» process flow sheet illustrated in the figure below.

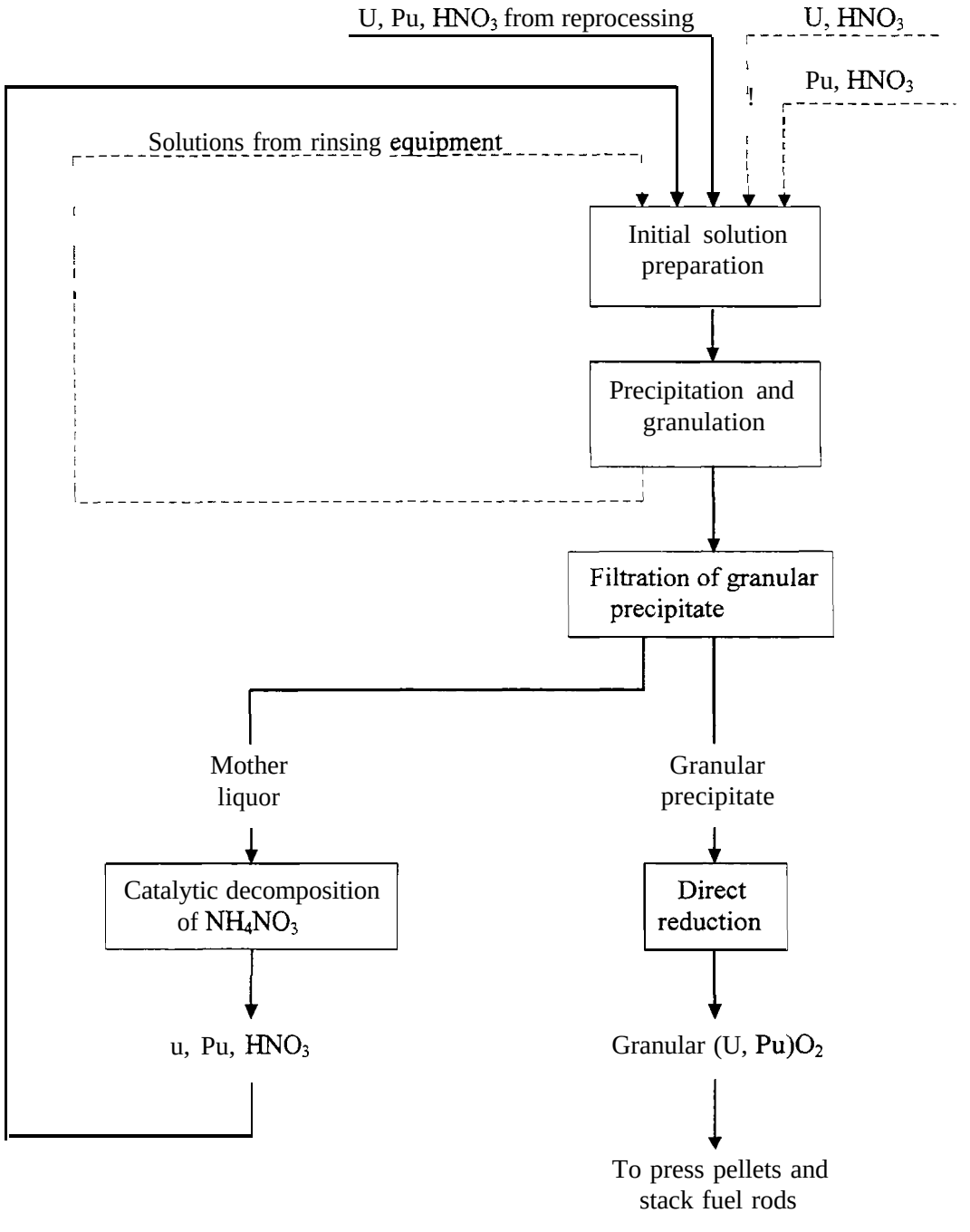

FIG. Modifed «GRANAT» Process

The forthcoming objective consists in the complex testing and mastering the process in the demonstration semi-commercial facility. However, to-day the «GRANAT» technology is conceived to be a compact efficient method of the U-Pu fuel fabrication with the utilization of waste within the

cycle characterized by the mentioned above advantages over the processes presently in operation. The technological and environmental merits of the «GRANAT» process show up as a single structure together with spent U-Pu fuel reprocessing.

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VALIDITY OF USING $UPuO_2$ VIBROPACK EXPERIMENTAL FUEL PINS IN REACTORS ON FAST AND THERMAL NEUTRONS: FIRST EXPERIMENTS ON CONVERSION OF WEAPONS GRADE PLUTONIUM INTO NUCLEAR FUEL

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Abstract

Extensive scope of scientific and technological work has been carried out in SSC RF RIAR to substantiate usage of vibropack oxide fuel pins in fast and thermal neutron reactors. In fulfilling of the work there have been studied physical-mechanical and technological characteristics of granulated fuel, carried out radiation tests and material science investigations of mock-up, experimental and research fuel pins of BN-type (in BOR-60 and BN-600 reactors) and VVER-1000 type (in SM-2 and MIR reactors). Total quantity of fabricated fuel pins is about 30 000 pieces. In BOR-60 reactor maximum burn-up attained 30% h.à. for regular SA and burnup was of 32,3% h.a. for experimental fuel pins of the dismantled SA. In testing $UPuO_2$ vibropack fuel pins in BN-600 reactor there was attained maximum burn-up of- 10,8% h.a. Post irradiation examinations of fuel pins have revealed, that since the problems both of chemical and thermo-mechanical fuel-cladding interactions were solved, the resource of the fuel pins like these would only depend on the choice of cladding material. Vibropack fuel pins, containing $UPuO_2$ under conditions of MIR reactor attained burn-up more than 30 MW day/kg U both under nominal operation and under load-following modes.

The experience in designing, manufacturing and operating the facilities on fabrication of granulated uranium and MOX fuel and fuel pins is gained. There is created the data bank and calculation codes, describing vibropack fuel pin behavior under different operation modes.

According to the Concept of RF Minatom on recovery of surplus weapon-grade plutonium, resulted from disarmament, the State Scientific Center of Russian Federation RIAR (Dimitrovgrad) has begun a practical realization of the technology on conversion of metal weapon-grade plutonium into mixed uranium - plutonium oxide fuel. There has been carried out processing and is obtained granulated $UPuO_2$ fuel for BOR - 60, BN - 600 reactors and experimental batches of granulated fuel for mock-up and experimental fuel pins of VVER type, which are intended for testing in SM and MIR reactors.

I. INTRODUCTION

The main fuel kind of energetic facilities for the last 50 years is oxide fuel. It is unlikely to be substituted for either metallic fuel or nitride one in the near future. That's why the technologies, enabling to increase greatly oxide fuel possibilities and to upgrade considerably technicoeconomic factors of fuel cycle as a whole as well as taking into account up-to-date requirements to the process safety and ecological compatibility are of special interest.

For the last 25 years SSC RF RIAR has been carrying out the complex investigations to validate the possibility of implementing short close fuel cycle for nuclear reactors. In contrast to the conventional aqueous method fuel reprocessing SSC RF RIAR activities are based on the principles of «dry»technology, including:

- pyroelectrochemical methods of reprocessing high-active nuclear fuel in the molten salts followed by obtaining granulate, which is suitable for fuel pin fabrication by vibropacking;
- vibropacking technology for fabricating fuel pins of granulated fuel;
- automated remote-control processes to produce granulated fuel, fuel pins and SA.

By the moment the first stage of scientific-research activities on development and experimental validity of fast reactor fuel pins, containing vibropack MOX fuel has largely been completed. Developed fuel pin design and carried out comprehensive study on radiated stability of vibropack fuel and the pins, containing this fuel, as well as worked out automated remote-controlled technology for fuel pin fabrication perfectly meet the requirements to the closed fuel cycle concept and show high technicoeconomic and operational characteristic. There is gained the experience in designing, manufacturing and operating the facilities for the production of granulated uranium and uranium plutonium oxide fuel as well as fuel pins. There is created the data bank and calculation codes. Since 1981 BOR-60 Core has been used vibropack $UPuO_2$ fuel. At present there is attained maximum in the world burnup of - 30,3 % for regular design SA, and -32,3 % for individual experimental fuel pins. By pyroelectrochemical way there has been reprocessed about 10 kg of weapon grade plutonium, from which 16 SA have been manufactured and 12 SA have been tested in BOR-60 reactor. In BN-600 reactor 6 SA with $UPuO_2$ fuel have been tested successfully up to the burnup of 9,6 % (maximum burnup for individual fuel pins ~10,8 %). The report presented the main results to substantiate vibropack oxide fuel pin serviceability for fast and thermal reactors.

2. **Technology of vibropack fuel pin fabrication**

Vibropacking technology is always considered as the way of fabricating fuel column, which allows one to cut substantially costs for nuclear reactor fuel pin fabrication and to improve fuel pin operational performance. The main merits of the vibropacking technology and vibropack fuel pins are the following:

- simplicity and reliability of the technologic process attributed to less the number of technologic and check operations. This fact simplify automation and remote control of the process, therefore the vibropacking technology can be employed to make fuel pins of recovered fuel in the shielded hot cells;
- the possibility of fabricating fuel column , having easily variable parameters and on the basis of multicomponent compositions;
- the possibility of using any kind granulate: both of homogeneous composition and of mechanical blend;
- . less thermal-mechanical fuel effect on the cladding in comparison with pellet fuel
- diminished requirements to the inner diameter of fuel pin claddings.

Vibropacking consists in creating dense packing of the particles for determined granulometric composition powder under the effect of mechanical vibrations. Vibration application makes it possible to impart quasiliquid properties to a dispersive powder medium. At that fuel particles are becoming movable and able to form optimum packed structure in accordance with the particles size and composition.

For the particles of irregular form there is usually used the granulate of polydispersive composition, allowing one to attain necessary smear density value, which is determined as the ratio of $UPuO_2$ granulate full mass to its volume. Application of optimized composition enables to orient to the planned final smear density as early as the fuel portion preparation stage. As a result of vibropacking the fuel column smear density for fast reactor fuel pins is 9,0 ÷9,4 g/ñm^3. For comparison, «smear» density, in other words the one with the allowance for clearance and pellet central hole, of pelletized fuel column is 8,5-8,6 g/ñm^3.

Filling and vibropacking technology provide fuel column smear density and plutonium distribution along the axis within the limits of not more than ±5 %of the nominal level.

To correct O/M ratio and eliminate the influence of technologic impurities, metal uranium granules as a getter are introduced in the granulated fuel at the portion preparation stage. The form of metal uranium getter particle is close to the spherical one with the maximum diameter up to 100 mcm. In exercising the technology there is provided the evenness of getter distribution over the fuel column length in the range of ±5 %. As a result of complex investigations, which included out-pile and in-pile tests (BOR-60, BN-350, BN-600), and postirradiation material science examinations there was upgraded getter contents with the allowance both for thermodynamic characteristic and for the necessity of attaining the required hypostoichiometric fuel **O/M** ratio.

The possibility of fabricating fuel pins by vibropaking is shown in two versions: manual - in the glove boxes and remote - in the shielded cells (Table2. 1). Semi-industrial technology of fuel pin and SA manufacture for BOR-60, BN-3.50 and BN-600 reactors under remote conditions was implemented when working at «ORYOL» facility(1977 - 1986, about 300 SA for BOR-60 were manufactured), SIC (1989-1997, 26 SA of BN-800 type were produced) and «Kolibry» mockup facility (1992-l 997, 63 fuel pins were manufactured).

Table 2.1

VIBROPACK OXIDE FUEL PIN FABRICATION

Fuel kind	**Reactor**	**SA quantity, pieces**	**Fuel pin quantity, pieces**
$UPuO_2$(RG)	BOR-60	426	15762
$UPuO_2$(RG)	BOR-60	16	592
$UPuO_2$(WG)	BN-350	2	254
UO_2(reg)	BN-350	7	889
UO_2(reg)	BN-600	6	762
$UPuO_2$(WG)	BN-600	6	762
$UPuO_2$(RG)	BN-600	4	508
$UPuO_2$(WG, RG)	Physical Stand	8	1016
UO_2(reg) + PuO_2(irr.)	BOR-60	1	6
UO_2(reg)	BOR-60	235	8695

R.G. - Pu of reactor grade;
W.G. - Pu of weapon grade;
reg. - ((regenerated)) -product of irradiated fuel reprocessing at RT- 1.
irr. &radiated)) - product of BOR-60(24 %) and BN-350 (4.9%) irradiated fuel reprocessing
Total (state at the end of 1998): SA - 731 pieces, fuel pins - 30000 pieces

At present there are operated two chains of glove boxes to fabricate fuel pins by vibropacking, which are also used for carrying out different experimental programs, as a well as for producing BOR-60 fuel pins and regular loading SA. Operating technological equipment of Semi-Industrial Complex (SIC) exhibited its high reliability. Serviceable production yield was more than 98 %. In fabricating fuel pins manually at the technologic line, located in the shielded glove box (-1300 BN-350 fuel pins, -300 BN-600 fuel pins, -9000 BOR-60 fuel pins), the serviceable fuel pins yield was 100 % almost in all the quality parameters, being under check.

At the end of 1998 there was started the program of the weapon grade plutonium recycling by pyroelectrochemical way with simultaneous purification of gallium down to 7 - 10 ppm. About 20 kg of plutonium was reprocessed into MOX fuel, which was used to manufacture 16 SA by vibropacking, and 12 SA are currently being tested in BOR-60 reactor.

3. The results of vibropack MOX fuel tests in BOR-60, BN-350, BN-600

Tests of vibropack MOX fuel pins occurred in BOR-60, BN-600, BN-350 reactors with the aim of substantiating the design and upgrading both technologic and operational parameters. Main parameters of fuel pins for these reactors are presented in table 3.1.

Table 3.1

Main parameters of fuel pins for fast reactors

N	Parameter	Value		
1	Reactor	BOR-60	BN-350	BN-600
2	length of, mm			
	• fuel pin	1082	1790	2400,2440
	• gas plenum	273	300	600
	• bottom blanket zone	150	400	350
	• fuel column	450	1060	950
	• upper blanket zone	100	-	350
3	Cladding diameter, mm	6.0; 6.9	6.9	6.6
4	Cladding thickness, mm	0.3; 0.4	0.4	0.4
5	Spacer wire dimension , mm			
	• round	1.05	1.05	1.15
	• ellipsoidal	0.6×1.3	0.6×1.3	0.6×1.3
6	Smear density of fuel column, g/ñm^3	8.3 ÷ 9.5	8.4 ÷ 8.8	8.8 ÷ 9.2
7	Average mass ratio of PuO_2 in granulate, %	15 ÷ 40	20	30
8	Granulate enrichment in ^{235}U, %	45 ÷ 90	10	-
9	Unevenness of distribution over fuel column length, %			
	• of smear density	± 5	± 5	± 5
	• of plutonium	± 5	± 5	± 5

3.1 Tests of mixed vibropack fuel in BOR-60 reactor

Since the end of 1981 BOR-60 reactor Core has used vibropack MOX fuel. In the last three years recovered uranium dioxide has also been introduced in the reactor fuel cycle. By the moment about 500 SA have already been tested and are currently under test in the reactor. Their main technologic and operational characteristic are given in table 3.2 . The results of complex investigations on fuel pin design optimization exhibited, that the most radical way of improving fuel pin serviceability consists in introducing metal uranium powder as a getter in the fuel column composition. Getter addition of not more than 5 % wt gave an increase in burnup up to 11,5 % h.à. (maximum burnup is 15÷16 % h.à.). The getter content elevation from 5 up to 10 % and the increase of fuel column density enabled to attain burnups of the same depth, when thermal loading and cladding temperature were higher. Histogram, showing attained burnups for SA with getter, is given in Fig. 3.1.

Post irradiation material science examinations of SA verified the information, obtained before for experimental mixed vibropack fuel pins, concerning their behaviour under irradiation: off-gas, the change of cladding OD (outer diameter), formation of fuel column structure, FP s distribution (Zr, Ce, Ru) and etc. Mechanical properties and swelling of the fuel pin claddings were close to those of the experimental fuel pins, containing homogeneous fuel (UPu)Î $_2$, mechanical blend UO_2+PuO_2, and uranium dioxide pellets as well.. It was shown, that in the fuel pins with getter mass ratio more than >3 % there was no sign of physicochemical fuel cladding interaction even at the maximum burnup, thermal loading and cladding temperature. The single limiting cause, preventing us from attaining burnup of more than 15 % h.à. for the fuel pin design in question, is an enhanced swelling of austenitic cladding material under damaging doses of more than 80 dpà.

In-pile tests and material science investigations of base design fuel pins, which contain metal uranium powder in fuel composition as a getter and have the cladding made of advanced ferrite-martensite steels, confirmed their high serviceability up to the burnup of -30 % h.a. (Table 3.2). Indi

Table 3.2

Tests of SA, containing mixed vibropack oxide fuel in BOR-60 reactor

Parameter	Value
1. Fuel composition	UO_2+PuO_2, UO_2+PuO_2+U, $(UPu)O_2$+U
2. PuO_2 mass ratio, %	20-28
3. getter mass ratio, %	3-10
4. Fuel pin cladding diameterxthickness, mm	6,0×0,3; 6,9×0,4
5. Maximum linear power generation, W/ñm	510
6. Maximum cladding temperature	722
7. Maximum fuel burnup, % h.a. • regular SA • experimental SA • experimental fuel pins	 18.0 30,0 32,3
8. SA quantity, pieces total which attained the burnup of: • 10...15 % h.a. • 15...20 % h.a. • > 20 % h.a.	 442 279 11 10

vidual experimental fuel pins attained the burnup of more than >32% h.a. In PIE progress there was no sign either of thermomechanical or physicochemical fuel-cladding interaction in any of the fuel pin cross-sections under analysis. The typical microstructure of vibropack $(UPu)O_2$+U fuel upon testing in BOR-60 up to the burnup of 30 % h.a is given in Fig.3.2.

Analysis of radiation characteristic of vibropack oxide fuel pin serviceability has shown that:

- using fuel composition, containing $(UPu)O_2$ + U getter, makes it possible to exclude corrosion processes due to cesium and halogens availability as well as possible technologic impurities, and therefore to remove restrictions in burnup because of physicochemical interactions;
- availability of the initial structure circumferential layer results in the fact, that under transient modes cladding stresses are many times as low, and stress relaxation comes much earlier, than those of pellet fuel pins. As a result, fuel pin changes in SA Core under power ramp (Table 3.3) don't effect fuel pin serviceability; elevated smear density of fuel column (>9,0 g/cm') allows us to possess sufficient temperature reserve up to the melting temperature;
- integral value of swelling for vibropack $(UPu)O_2$+U fuel is (0,6±0,1) %/ % of burnup;
- there is no revealed an influence of granulated fuel $(UPu)O_2$ or UO_2+PuO_2 production technology upon fuel pin serviceability.

3.2 *Tests of mixed vibropack* fuel *pins in BN-350 reactor*

In the period from 1985 till 1986 in BN-350 reactor there were irradiated two experimental SA, containing mixed vibropack oxide fuel (MVOF) and 7 SA with uranium vibropack oxide fuel (UVOF). Their design features and test conditions are given in Table 3.4. According to the data of reactor CIC (Checking integrity of cladding), in operating one of SA, containing MVOF (S-585), were considered to have been failed and even have contacted with coolant. Upon exhibiting failure signs on the 45 operation day (burnup of 1,2 % h.a.), SA was irradiated for 110 more effective days. When cutting SA there was found one failed fuel pin. The other fuel pins didn't have external failures. Cladding leakage of the failed fuel pin took place along the Core center as a cross crack. Fuel was

available at the region of the cladding failure. There were no signs of defect propagation, fuel washing out and fuel-coolant interaction. The fuel pins with axial fuel mass transfer were found by gamma-scanning and masstransfer correlation with the fuel column initial smear density was fixed. Investigations determined, that, cause of axial fuel mass transfer and, therefore, occurrence of cladding through defect in the same fuel pin, consists in exceeding actual test parameters over the design ones in combination with the minimum fuel smear density (~8,4 g/ñm^3) in some fuel pins. Fuel pins with smear density of more than >8,6 g/ñm^3

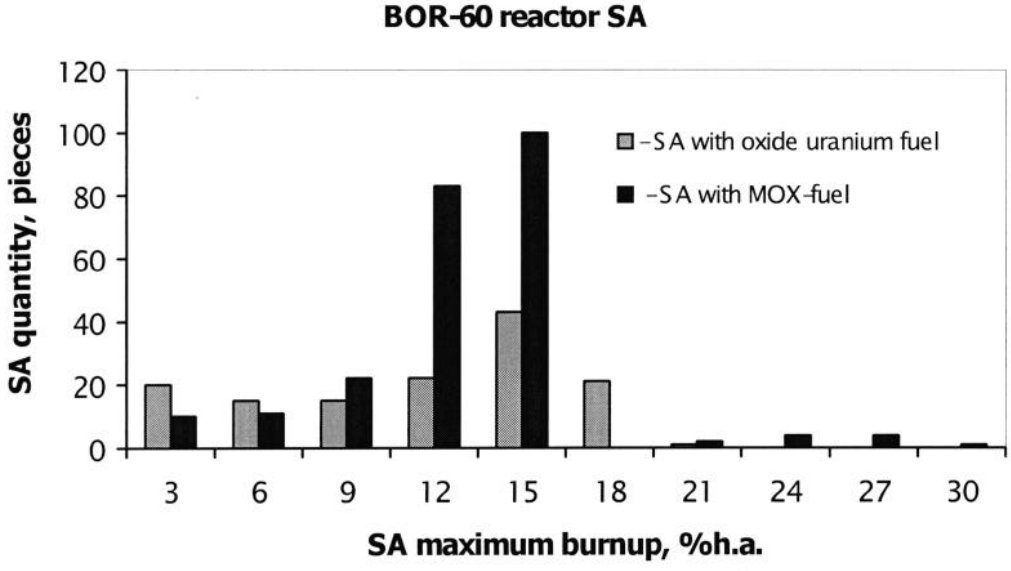

Fig.3.1 SA histogram of BOR-60 reactor

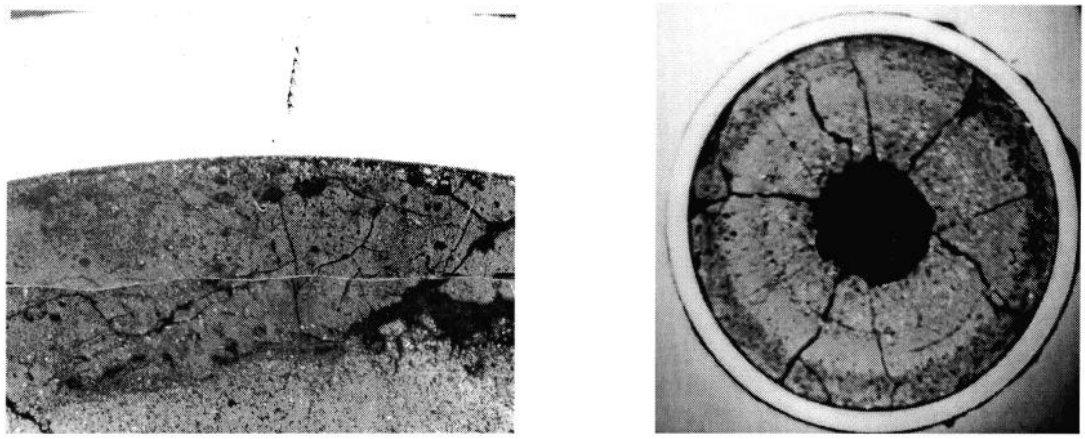

Fig.3.2.Micro- and macrostructure of fuel pin cross section
with burnup 32 % h.à.for BOR-60 reactor

Table 3.3

Test parameters for those BOR-60 SA, which are rearranged in operating

Rearrangement type	Pellet fuel			Vibropack fuel		
	SA quantity, pieces	Burnup h.a.	Failed SA quantity	SA quantity, pieces	Burnup, h.à.	Failed SA quantity
Within the same row	4	10÷12	2	16	8÷24	2*(in 2 Core runs after rearrangement)
From the center toward the circumference	18	10÷12	-	57	7÷22	-
From the circumference toward to the center	4	12÷17	3	85	8÷25	1*(in 5 Core runs after rearrangement)

remained serviceable even at the elevated operational parameters, there were no revealed any anomalies of either fuel or cladding. SA S-586, which attained the burnup by 36 % more than the design one, was unfailed according to the reactor CIC, but after transport procedures of unloading there was registered gas release. Material science investigations showed, that two fuel pins exhibited non-gastightness. Their unsealing was attributed to the excessive ovalization of the cladding, made of EI-847 steel, as a result of enhanced swelling and embrittlement in combination with high level of the stresses due to mechanical interactions of the swelling fuel pin bundle both with each other and with wrapper tube. Additional to the plan parameters of SA tests in BN-350 in conjunction with the variation of initial fuel column smear density enabled to verify experimentally the estimation of limit operational parameters for mixed vibropack fuel pins. From the results of in-pile tests and material science investigations there were made changes in design and in fabrication technology of mixed vibropack fuel pins with the purpose of increasing reliability and their life-time, namely:

- there has been increased minimum value of fuel column smear density up to 8,6 g/ñm^3 and getter mass ratio up to 10 %;
- there has been used more resistant to irradiation construction materials.

All the design upgrade was implemented in manufacturing six experimental SA for BN-600 reactor.

3.3Tests of mixed vibropack fuel pins in BN-600 reactor

In BN-600 reactor there were tested 6 SA containing vibropack MOX fuel, and 4 SA with uranium-oxide fuel as well. In-pile tests of 6 SA under design parameters were successfully completed. Maximum burnup was 9,8 % h.à.(for individual fuel pins -10,9 %), table 3.4.

There were no revealed failed fuel pins in SA at the CIC facility. The results of complex material science investigations of the subassemblies fuel pins with burnup 6,8 % h.à. made it possible to find out, that:

- general condition of all the SA and fuel pin components was satisfactory;
- maximum changes of the cladding diameter and the operating dimension of the wrapper tube didn't exceed 2%, which provided evidence for unexhaust of SA serviceability resource according to the admissible deformation criterion;
- there were no signs of failed fuel pins, the failure of fuel column uniformity and fuel axial masstransfer.

While investigating by metallography there were no revealed any anomalies, overheat signs and cladding corrosion damage both from fuel and from coolant. The comparison of fuel column

Table 3.4

Tests of vibropack fuel SA **in** BN-350 **and** BN-600 **reactors**

Reactor	BN-350			BN-600			
SA No	P1....P7	S-585	S-586	WG-0187	WG-0287	VPC1-VPC4	WG03 - WG0(
the year of fabrication	1982	1984	1984	1987	1987	1989..1990	1990..1991
Fuel composition	UO_2	$(UPu)O_2$		$(UPu)O_2$		UO_2	$(UPu)O_2$
Getter contents (U met.), %	5	5		10		10	10
Plutonium contents, %	-	20		22....28		-	~ 30
Enrichment in U^{235}, %	27	10		-		-	-
Smear density, g/ñm^3	8,8	8,3...8,8		8,9...9,1		8,8...9,2	8,8...9,2
Material of fuel pin cladding	EI-847			EP-172		CHS-68	
Material of SA wrapper	Cr16Ni11Mo3Ti			05Cr12Ni2Mo		05Cr12Ni2Mo	
Thermal loading, êW/m	48	51	48	45	45	42	42...44
Cladding temperature, °Ñ	710	740	690	670	670	680	680
Damaging dose, dpa	45	30	42	53	77	27	76...77
Burnup, % h.à.	7,2	4,7	6,8	6,8	9,6	3,6	9,6 (10,9)
Availability of failed fuel pins: in gas	1*	0	2*	0	0	0	0
in fuel	0	1	0	0	0	1+1	0

* - in carrying out transport operations

structural alteration from metallographic analysis results with those of thermophysical calculation showed a good agreement, which allowed verifying the fuel pin calculation program .

Thus, positive test results both of 6 experimental SA (2 SA - manufacture in the glove boxes, 4 SA- a remote, automated manufacture), containing vibropack MOX fuel, in BN-600 reactor and of fuel pin material science investigations confirmed the efficiency of the technologic decisions and the concepts, which were taken as principles in the mixed vibropack oxide fuel pin design, but in order to substantiate the results and to compile the set of statistical data the tests should be prolonged with the aim of accumulating irradiated SA massif not less than 30 SA in number.

3.4 *Emergency overheat of irradiatedfuelpins*

To study the serviceability of fuel pins under transient cladding overheat conditions in accordance with the operational regulation of BN-600 reactor there was carried out a series of experiments, including an isothermal heat of irradiated fuel under out-pile conditions, as well as the experiment on heat of the cladding up to its melting in the capsule-loop.

Main parameters of irradiated in BOR-60 fuel pins, having different fuel compositions, modes and results of emergency tests are presented in table.3.5.

From the tests and investigations made there were found out the features and regularities of irradiated fuel pin behavior under the emergency overheat. It is shown in particular, that mixed oxide fuel (UO_2-PuO_2-U-getter) pin, irradiated up to the planned and additive to planned burnups, remain sealed under rather severe modes of emergency overheat (for example, temperature of 800-850 °Ñ for 1-

1,5 hrs). Under these modes there was no revealed any corrosion damage of cladding inner surface, as well as signs of plutonium radial masstransfer, but the radial masstransfer of metal FPs took place.

Table 3.5

Main fuel pin parameters, modes and results of overheats

SA index	Material of fuel column	Material of cladding	Protective layer	Maximum burnup, %	Overheat mode: temperature, °Ñ	Overheat mode: Time, min	results: Cladding corrosion, mcm
ÎG-73	$(U,Pu)O_2$ vpc	E I - 8 4 7	is absent	7	800	90	
ÎG-73	$(U,Pu)O_2$ vpc	EI-847	is absent	7	850	90	
ÎG-115	$(U,Pu)O_2$vpc	EP-450	is absent	21	850	60	
I-805	U	EI-847	UO_2	2,0	770	90	
I-805	U	EI-847	UO_2	2,0	800	90	270
AZ-14i	U-15Pu	EI -847	W or Cr	6,2 - 6,8	740-830	90	24 - 160
ÀZ-15Ì	U-15Pu	EI -847	W or Cr	6,8	740	90	110
AZ-15i	U-15Pu	EI -847	W or Cr	6,8	800	90	160
ÎP-25	UO_2 -tab.	EP-450	is absent	12,6	740	90	200

4.Main results of vibropack fuel pin tests in thermal reactors

An extensive scope of technologic activities, in-pile tests and material science investigations was performed to study the possibility of applying vibropack oxide fuel in thermal reactors. The investigation results also give evidence for a real possibility of upgrading technocoeconomic figures of an advanced fuel cycle for thermal reactor, based on “dry” irradiated fuel reprocessing and vibropacking technology. The principal directions of the investigations are given in table 4.1. In carrying out work physicomechanical and technologic characteristic of granulated fuel were studied, mockup, experimental and pilot fuel pins of VVER type were tested in MIR and SM-2 reactors and then were investigated with material science aims.

Table 4.1

Characteristic and test conditions of pilot mixed vibrooack fuel pins

Kind of investigation	quantity, pieces	ρ_{eff}, g/ñm^3	$Q_{L,max}$ W/ñm	Duration, eff.davs	burnup, Ì W·dav/t	Reference
Exercising vibropacking technology	40 25 12	10,6...10 ,8				l_{ac}=(0,2...4) m u =(0...30) % Gd =(0... 15) %
forming structure, thermophysics	**18**	10,3	65	250		
Thermocycling	**35**	9,8	56	535	43000	4300 cycles
Resource tests: MIR						
MIR (water boiling loop)	24 6	9,8 10,2	**55** 40	550	32000 47500	Tests are completed
MR	12	10,4	50		35900	
MIR (UPuÎ $_2$ +U, UPuÎ $_2$ +Gd)	6	9,8	48		35000	in the reactor
Failed fuel pins : UO_2, $(UPu)O_2$	1	10,3	85	500		Initial defect, unsealing at power, endplug defect

5. Conclusions

By the moment in SSC RF RIAR there has been fulfilled the extensive scope of scientific and research activities to validate vibropack oxide fuel pin application in fast and thermal reactors. In carrying out the work physico-mechanical and technologic characteristic of granulated fuel were studied, in-pile tests and material science of mockup, experimental and pilot fuel pins of BN-type (in BOR-60, BN-350 and BN-600 reactors) and WER type (in SM-2 and MIR reactors) were implemented. Mass tests of vibropack MOX fuel pins in BOR-60 reactor in combination with their successful tests in BN-600 reactor, reliable operation of Semi-Industrial Complex Facilities enable to come to the conclusion on a real possibility of using vibropack oxide fuel pins to develop safety, profitable uranium-plutonium fuel cycle on the basis of «dry» technologies as well as to recover plutonium both of reactor and weapon grade in nuclear reactors.

THE MICROSTRUCTURE OF UNIRRADIATED SBR MOX FUEL

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Abstract

At Sellafield, BNFL has been producing MOX fuel assemblies for use in thermal reactors since 1994 in its MOX Demonstration Facility (MDF) using the Short Binderless Route (SBR). The SBR is a compact, simple and rapid dry powder process for blending IDR UO_2 and PuO_2 feed powders and for granulation prior to pressing.

A scaled-up version of this process will be used in the larger capacity Sellafield MOX plant (SMP). One of the advantages of this manufacturing route is that only a single direct powder blending stage is employed and there is no requirement for precompaction or other granule pre-treatment prior to pressing other than spheroidisation.

The finished fuel structure is uniformly free from internal cracking and the overall quality is excellent. The PuO_2 is distributed through the UO_2 matrix homogeneously by the high energy 'attritor' mill, the sintered pellet grain structure is uniform and the porosity, controlled by the Conpor additive is bimodal and stable to thermal densification. Typical production fuel pellet macro and micrographs, pore size distribution, grain structure and autoradiograph are presented.

1. INTRODUCTION

BNFL's MOX Demonstration Facility (MDF) began producing Short Binderless Route (SBR) PWR MOX fuel assemblies in 1994. BNFL has over 30 years' experience of manufacturing MOX fuels for a range of reactor designs but MDF represents BNFL's first commercial venture into the thermal MOX market. It is a small scale semi-automatic demonstration plant built around the SBR (originally developed for the manufacture of Fast Reactor MOX) which employs attritor milling and spheroidising technologies for blending and granulation of the feed powders prior to 'green' pellet pressing.

Soft pellet handling techniques developed by BNFL are used for loading green pellets to furnace boats and for handling sintered pellets through buffering, grinding and inspection prior to rod production.

The result is a compact manufacturing route generating a MOX nuclear fuel product which has excellent characteristics. PWR MOX fuel assemblies have been manufactured for 4 thermal reactors.

A scaled-up version of MDF's MOX fuel manufacturing route forms the basis for BNFL's full commercial scale Sellafield MOX Plant (SMP).

2. BNFL SBR MOX PELLETING PROCESS DESCRIPTION

2.1 Attritor Milling and Granulation

The first stage in the process is the attritor milling of the feed powders. These comprise UO_2 from BNFL's Integrated Dry Route (IDR) process and PuO_2 from the oxalate precipitation process at Sellafield. These powders are weighed into the attritor mill and are then

blended together with a small quantity of stearate lubricant and Conpor porosity control agent. The blended MOX powder is discharged automatically into **the** spheroidiser which granulates the powder and lubricates it with stearate before transferring to the hydraulic press.

2.2 Pressing And Sintering

The granules are pressed into compacts ('green pellets') of geometric density greater than 6g/cm3 which are transferred on BNFL's soft pellet transfer system to an automatic furnace boat loading 'pick and place' unit. Samples of the green pellets are taken, visually inspected to check physical condition and measured for density. The green density data are used as part of the overall Statistical Process Control (SPC) system for the plant process. The design of the green pellet transfer and loading equipment is such as to minimise damage to the fragile compacts. Furnace boats containing fuel from each production 'lot' are logged into and from the furnace with lot to lot segregation ensured using empty boats.

The fuel is sintered at a maximum temperature of 1650°C in a reducing atmosphere of 4%Hz/Ar to which is added a small quantity of CO_2 to control grain growth *The* furnace treatment takes at least 24 hours to complete before discharging the cooled sintered pellets prior to grinding. The pellets at this stage are visually assessed and measured for length, diameter and taper. This data is used to control both the hydraulic press and also the selection and condition of the press tooling.

2.3 **Grinding And Inspection**

Each production 'lot' is segregated and has an individual identity. When all of the boats of fuel recorded as containing a particular 'lot' of fuel have been discharged from the furnace, the pellets are dry centreless ground to a customer specified diameter before transfer to the pellet inspection area.

All of the ground pellets are measured for diameter at various positions along the length, the data collected and the geometric density determined on a sample. All of the pellets are then inspected for physical defects before being consigned to a store. The fuel is bonded until chemical analysis has confirmed that it meets the specification then the 'lot' of fuel is released for loading to rods.

3. THE MACRO AND MICROSTRUCTURE OF BNFL SBR MOX FUEL PELLETS.

3.1 Macrostructure

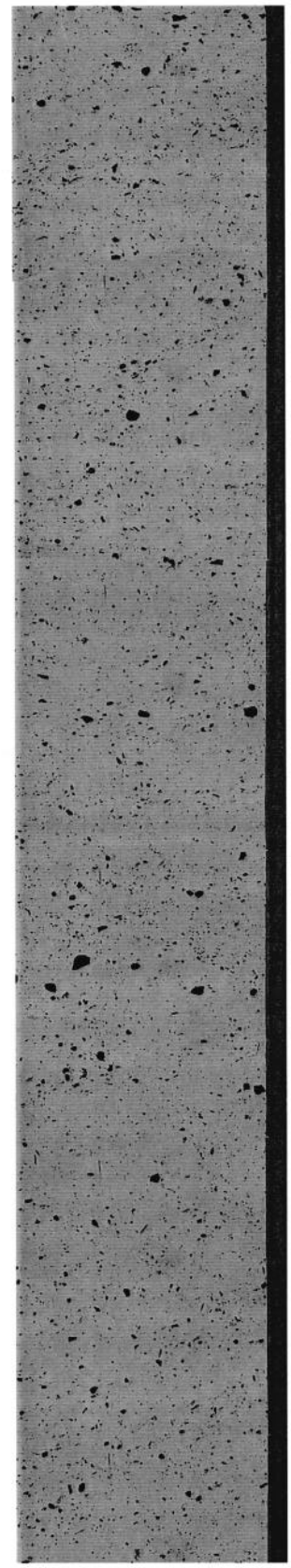
The macrostructure of typical production fuel covering several fuel manufacturing campaigns is illustrated in Figure la. It can be seen that the pellet structure is uniform and that there are no inclusions or significant physical flaws. Importantly the visible porosity is not linked (see Figure lb), there is no evidence of internal cracks or fissures and the surface condition is excellent.

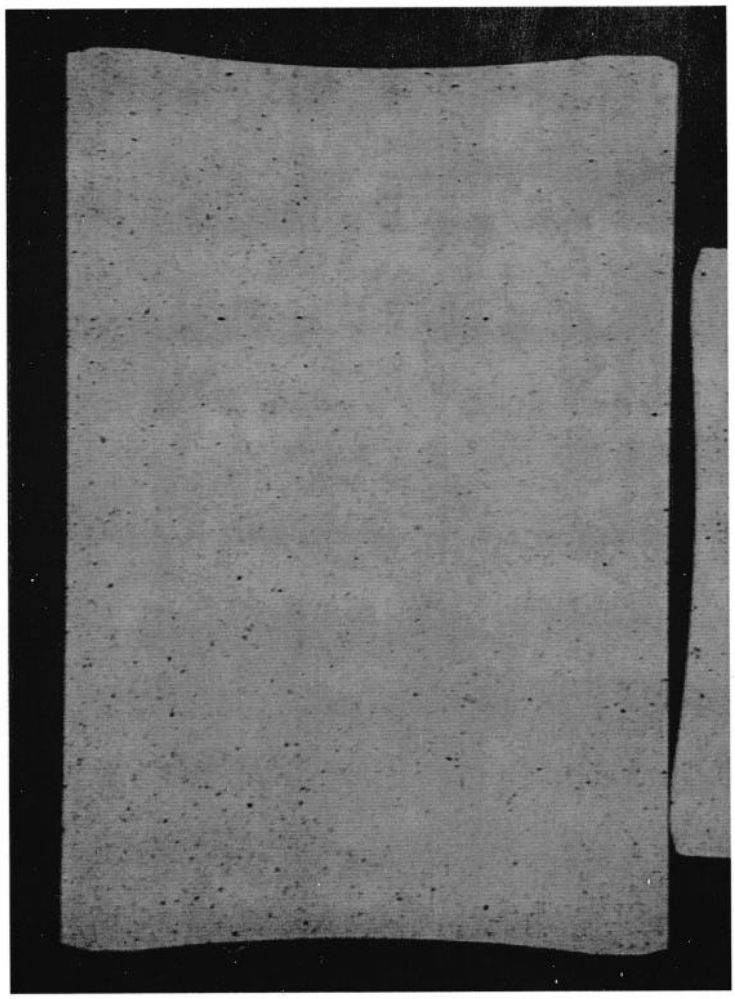
3.2 **Microstructure**

3.2. I *Porosity*

The porosity distribution appears bimodal, (see Figure 2) and consists of two well-defined peaks, one centred at about 2μm diameter which is due mainly to the sintering process and a second peak centred between 10 and 30μm diameter which is mainly due to the burning out of the additives. There is virtually no porosity above 1 00μm.

(1b- part of the side x45)





(1a- whole pellet x10)

Figure 1
Macrographs of BNFL SBR MOX Fuel Pellet.

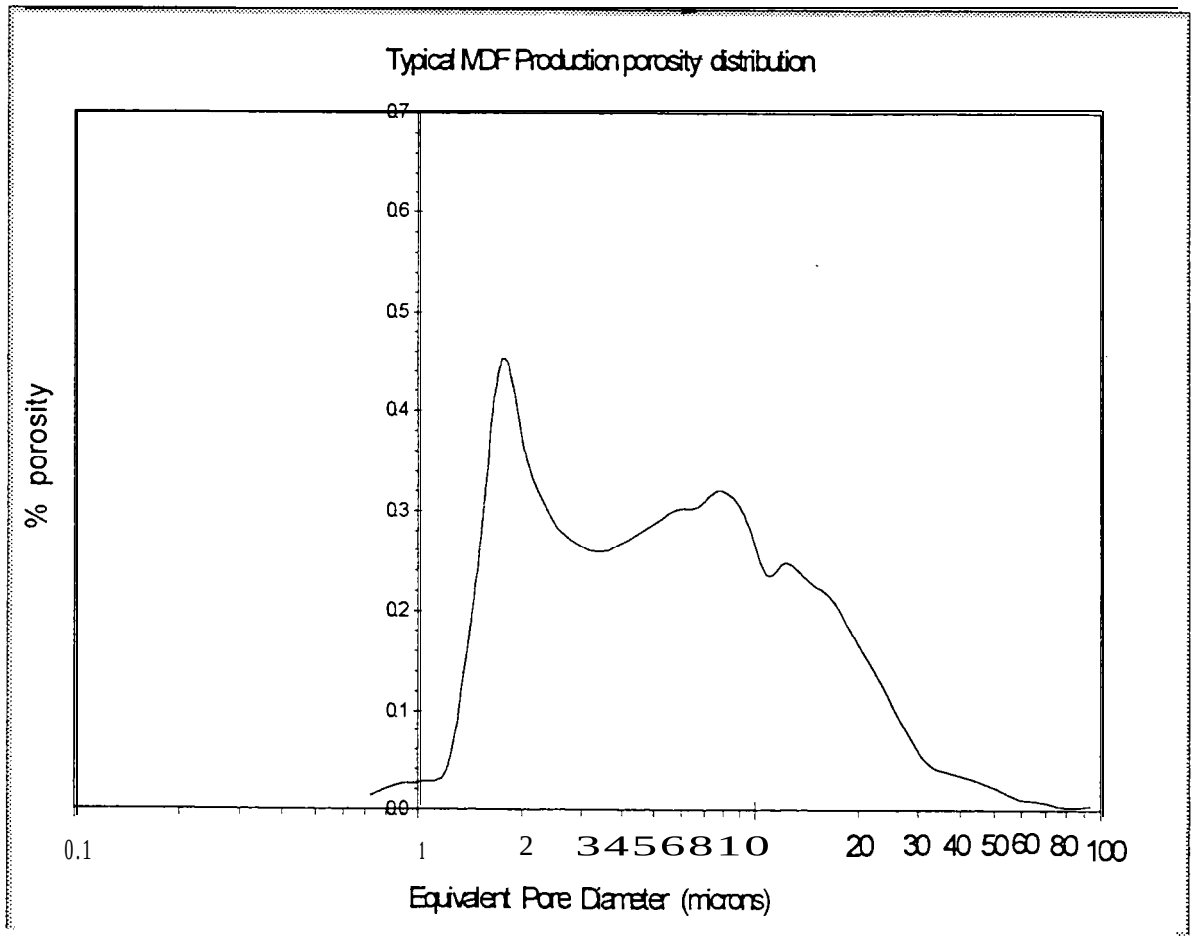


Figure 2

Typical Porosity Distribution of **BNFL** SBR **MOX** Fuel.

The first peak represents about 1 to 2% porosity. The second peak represents between 3.5 and 4.5% porosity. A summary of the quantitative porosity measurements is given in Table 1 below.

TABLE 1. QUANTITATIVE POROSITY

Campaign No.	Total % porosity	% porosity <3 microns	% porosity <5 microns	% porosity >3 microns	% porosity >5 microns	% porosity >30 microns	% porosity >50 microns	% porosity >100 microns
1	4.65	1.11	2.05	3.55	2.60	0.08	0.03	co.01
2	5.01	1.68	2.62	3.33	2.40	0.04	co.01	nd
3	5.74	1.68	2.35	4.06	3.40	0.30	0.08	co.01
4	5.06	1.52	2.20	3.55	2.87	0.30	0.08	<0.01
5	5.03	1.37	2.20	3.69	2.85	0.44	0.03	0.01

The pore size range and profile can be broadly predicted from the particle size of the Conpor. The use of Conpor of the same particle size distribution ensures a uniform porosity distribution. A macrograph of the pores in a typical production fuel pellet showing the even structure, and the absence of both interlinked and open porosity is shown in Figure lb.

3.2.2 *Grain Structure*

SBR MOX fuel produced in MDF has a uniform grain structure. An example is shown in Figure 3. Grain size is controlled by adjusting the oxygen potential of the sintering furnace atmosphere. This is achieved by adding a small quantity of carbon dioxide to the reducing atmosphere of 4% hydrogen in argon. Grain size is required as part of fuel certification and is measured by the linear intercept method. A series of grain size results is presented in Table 2 below.

3.2.3 Pu Homogeneity

BNFL routinely uses colour alpha autoradiography for assessing the macrohomogeneity of their MOX fuel. In this method, a polished section of a fuel pellet is held in contact with a colour slide film. The PuO_2 in the sample causes the film to produce a range of colours which depend on the Pu concentration of the Pu rich region. Assessment of the Pu concentration is made by comparison with a series of standards exposed at the same time (see Figure 4).

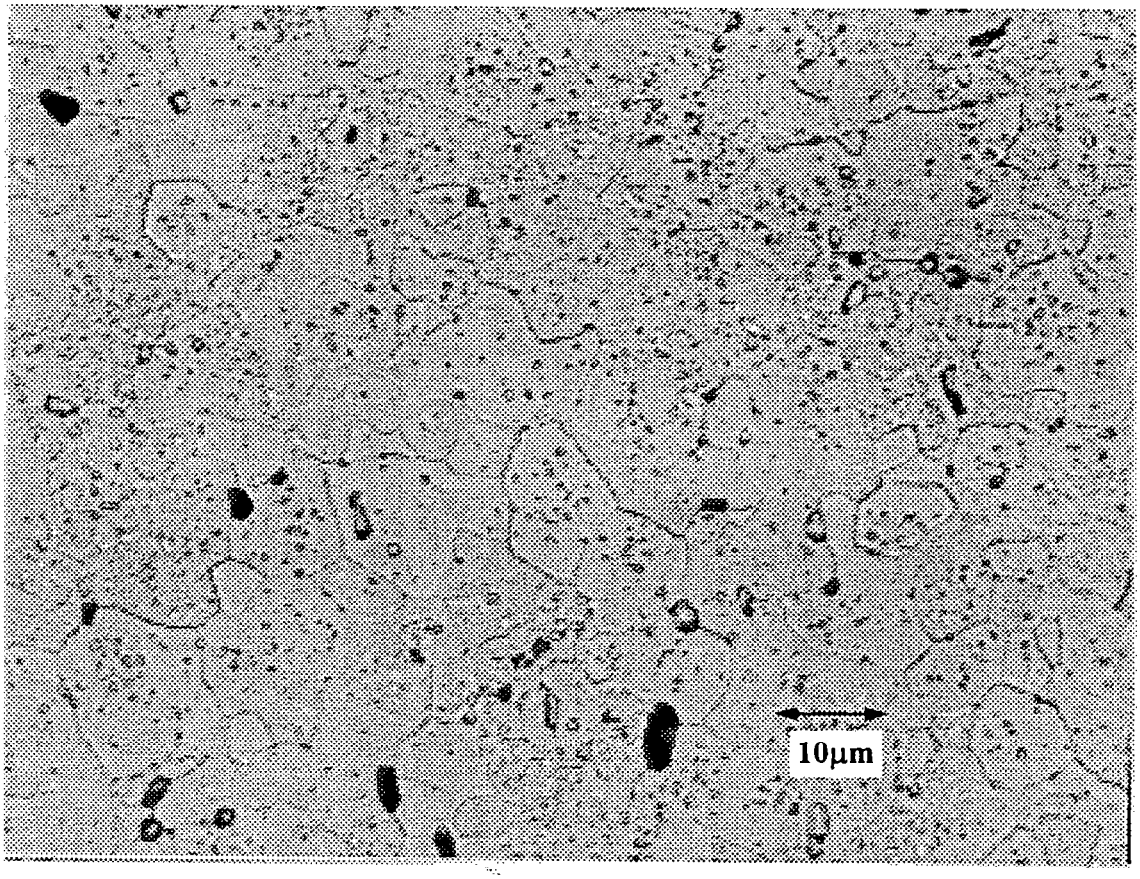
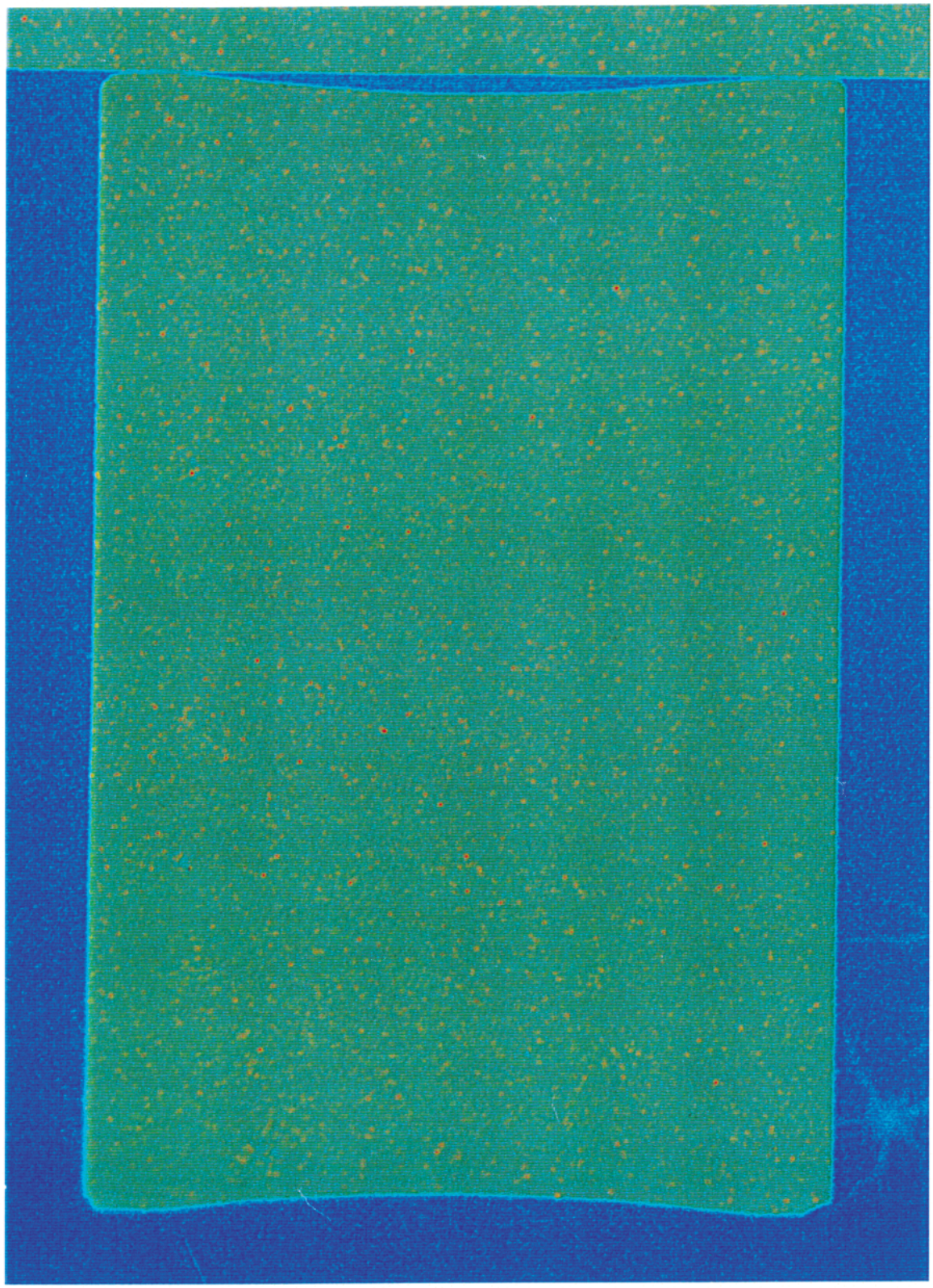


Figure 3
Grain Structure of Standard BNFL SBR MOX Fuel
(x 1400 magnification)



5	10	15	25	30	35	45	55	60	70	90	100

Plutonium standard (% Pu/U+Pu)

Figure 4
Autoradiograph of BNFL SBR MOX Fuel Pellet (x15).

TABLE 2. GRAIN SIZES FOR A RANGE OF %Pu IN MOX

Enrichment (% Pu)	Mean Grain Size (microns)	Standard deviation (microns)
2.6%	7.6	0.2
3.2%	8.0	0.2
3.5%	6.9	0.3
4.4%	8.9	0.4
4.9%	6.6	0.4
6.3%	8.5	0.4
6.9%	7.2	0.6

The average grain size for MDF fuel is about 7.5 microns,

A typical autoradiograph of production fuel containing approximately 8% Pu is shown in Figure 4. There are no visible zones of pure PuO_2 present in the fuel pellet. The small areas showing a higher level of Pu concentration are typically up to 20μm in diameter and the average Pu concentration is less than 50%.

In addition to the above routine assessments, a sample SBR MOX fuel pellet has been subjected to energy dispersive X-ray (EDX) analysis to produce a chemical 'map' of Pu rich areas, [2]. The EDX data reveals information on the Pu rich areas within the MOX (represented by size range) and the maximum Pu contents within those areas. The size ranges are shown below in Figure 5.

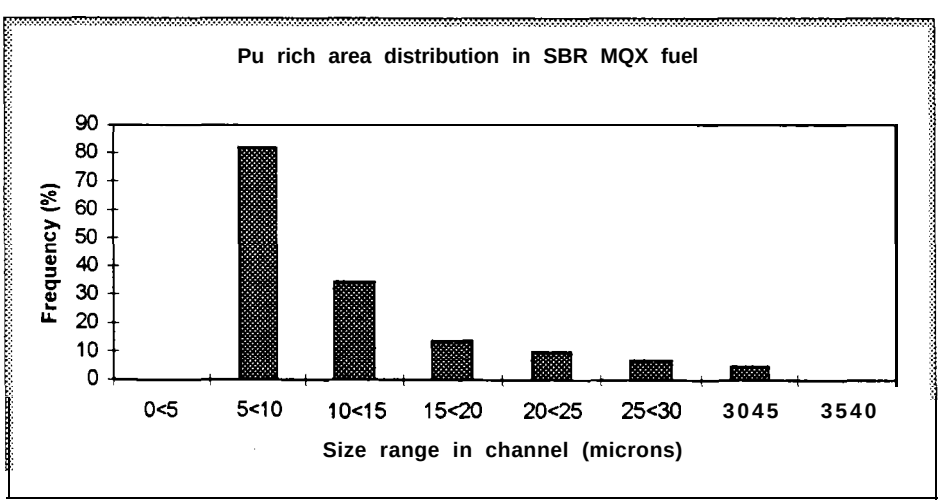


FIGURE 5. HISTOGRAM OF PuO_2 RICH AREAS IN MOX

All of the Pu rich areas are smaller than 40 microns diameter with greater than 90% below 25 microns. The volume fraction with a Pu concentration above 20 wt% was only 2%. The highest Pu concentration (50 to 60 wt%) within any area was negligible at 0.01%.

SBR MOX fuel therefore has a uniformly mixed oxide matrix with a Pu content very close to the nominal enrichment with only a few small areas enriched in Pu. The EDX work confirms the alpha-autoradiography and shows that the homogeneity of SBR MOX fuel is excellent.

3.2.4 Immersion And Geometric Density

SBR MOX fuel made in MDF has generally been manufactured to a target geometric density of 95% +/- 1.5% Theoretical Density (TD) or 10.45g/cm3 +/- 0.17g/cm3. Fuel certification is carried out on a statistical basis on a 20 pellet sample where the density is measured using a geometric technique. Both immersion and geometric densities have been performed on samples of sintered fuel pellets. Data are listed in Table 3.

TABLE 3. COMPARISON BETWEEN GEOMETRIC AND IMMERSION DENSITIES

Mean Geometric Density (GD) (g/cm^3)	Mean Immersion Density (ID) (g/cm^3)	Difference (ID-GD) (g/cm^3)
10.430	10.437	0.007
10.420	10.426	0.006
10,401	10.420	0.019
10.43 1	10.441	0.010
10.403	10.442	0.039
Mean 10.417	Mean 10.433	Mean 0.016

Generally the immersion density (carried out using water to which is added a small concentration of wetting agent) is higher than the geometric densities by about $0.015g/cm^3$ or 0.15% by volume. The observed positive bias in favour of immersion densities is due to the water penetrating other physical features which are not identified by geometric measurement. The residual open porosity is practically negligible (see Figure lb).

3.2.5 Thermal Resinter Stability

The test for thermal resinter stability is carried out at between 1700 and 1720°C over a 24 hour period in an atmosphere of 4% hydrogen in argon. The results are corrected for any adjustment to the oxygen to metal ratio which might occur. Every MDF fuel pellet tested has passed the thermal stability test so far specified by any customer. Table 4 presents a series of typical results taken across the full production history of MDF.

TABLE 4. THERMAL STABILITY TEST RESULTS

Nominal enrichment (%Pu)	Mean of 10 results (% TD)	Standard Deviation (% TD)
2.6	0.430	0.095
3.1	0.323	0.098
3.5	0.214	0.065
4.4	0.291	0.079
6.3	0.128	0.073
8.2	0.372	0.068

4. SUMMARY

The BNFL Short Binderless Route has been employed for 5 years in MDF at BNFL, Sellafield for the manufacture of thermal reactor MOX fuel. This dry manufacturing route is a

fast efficient method for producing press feed granules froiFi the starting feed powders. The fuel produced has proved to be of excellent quality and its macrostructure confirms its physical consistency. Micrographs are presented which demonstrate the absence of physical flaws and the homogeneity of the PuO_2 distribution. Data collected over the 5 year period of operation confirm the consistent grain structure, the near absence of open porosity and the characteristic bimodal porosity and thermal stability of the fuel pellets.

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EXPERIENCE AND TRENDS AT THE BELGONUCLEAIRE PLANT

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Abstract

The BN Dessel plant started operation in 1973. During the initial period (up to 1984), the plant was equipped to fabricate as well FBR as LWR fuel (5 tHM/year) within the framework of LWR demonstration and FBR programmes.

The first ten years of operation have laid down the bases for all modifications or improvements in the fields of fuel fabrication and control processes, waste management, safety and safeguards which were implemented in the 1984-1985 refurbishment. On this occasion, the MIMAS fabrication process has been introduced to make MOX fuel reprocessable in the operating conditions of the modern reprocessing plants (COGEMA in La Hague and BNFL in Sellafield) and the capacity of the plant has been upgraded to the current nominal capacity of 35 tHM per year (with a maximum licensed of 40 tHM per year).

The nominal fabrication capacity was achieved in 1989, maintained consistently since then at least at that level, approaching even the license limit. The process has proved its high flexibility, in particular in terms of the large variety of MOX manufactured for both PWRs (14 x 14, 15 x 15, 16 x 16, 17 x 17, types) and BWRs (8 x 8, 9 x 9, 10 x 10 types), of the size of the campaign (from less than 4 tHM to 28 tHM), of the origin of the PuO_2 (from COGEMA and BNFL reprocessing plants, Pu from second generation), of the Pu content (up to 8.6 w/o in HM). From 1986 up to the end of March 1999, more than 21 t Pu as PuO_2 have been processed into more than 388 tHM of MIMAS fuel, delivered or to be delivered for use in 796 PWR assemblies and 184 BWR assemblies for 21 large power reactors (17 PWRs and 4 BWRs) in 5 countries (France, Germany, Switzerland, Japan, Belgium). The MOX fuel produced has been demonstrated to reach at least the same performance as the UO_2 fuel used simultaneously in the same reactors. An increasing number of the MOX assemblies are discharged at a burnup of at least 45 GWd/tHM assembly average. One assembly reached 5 1 GWd/tHM assembly average after 6 irradiation cycles. No failures due to the MOX were noticed.

Since the mid 1990's, the plant is being backfitted without interruption of the fabrication, to incorporate improvements resulting from accumulated experience to improve still further the flexibility of the plant while meeting the more challenging future requirements in particular in terms of radioprotection regulation (degrated plutonium isotopic composition, higher burnup design fuel assemblies, ICRP 60) as well as in terms of economics (recycling of the scrap, reduction of the fabrication generated waste).

1. Introduction

MOX fuel has been fabricated at the BN Dessel plant, for LWR demonstration, and Fast Breeder Reactor Programmes since 1973.

In 1984, 1985, the Mimas fabrication process has been implemented to make MOX fuel soluble in nitric acid, in the operating conditions of the modem reprocessing plants (COGEMA in La Hague, BNFL in Sellafield). Simultaneously, the plant has been refurbished and its fabrication capacity increased to the current nominal capacity of 35 tHM per year.

Today, thanks to the experience gained and the excellent result obtained with the MIMAS process, the goals have been met : the process is mature and has proven its flexibility. The nominal fabrication capacity achieved in 1989 has been at least maintained consistently since then. The MOX fuel produced has been demonstrated to reach at least the same performance as the UO_2 fuel used in the same reactor. No failures due to the MOX were noticed and the slight behaviour differences compared to the UO_2 fuel fuel are adequately taken into account by the fuel designers.

Current investment programmes aim to increase still further the flexibility of the plant and to take into account the more severe constraints to be met in the future in terms of radiation protection regulation (degrated plutonium, isotopic composition, MOX fuel assemblies designed for increased discharge burn-up, ICRP-60).

This paper presents successively:

- the fabrication process;
- the status of our fabrication;
- the performance reached by our fuel;
- the trends in the evolution of the applied technology.

2. Mox fabrication process

The actual MIMAS process invented by BN in the early 1980s was implemented in 1985. It is the result of the evolution of four successive fabrication techniques (see table 1) developed by BN to produce pelletized MOX fuel.

TABLE 1. EVOLUTION OF THE MOX FUEL FABFUCATION TECHNIQUES OF BELGONUCLEAIRE

Fuel type	period	advantages and inconveniences
Granulated (UOz + PuO_2) blend	1960-1962 and 1965-1969 (laboratory)	Assumed best similarity to UOX fuel. Contamination levels, personal exposure and waste arisings resulting from complex handling of fine powder
PuO_2 blended into granulated UO_2	1967-1975 (pilot facility)	Significant departures from UO_2 fuel behaviour. Simplified handling of fine powder. Large plutonium-rich agglomerates. Unfavourable thermal conductivity. Higb fission gas release.
"Reference", i.e. PuO_2 blended into free flowing UO_2	1973-1984 (fabrication plant)	Fuel microstructure governed by the UO_2 matrix microstructure. Occurrence of plutonium-rich agglomerates. Too large proportion of the plutonium in insoluble residues (reprocessing problem).
"MIMAS", i.e. mixing of free flowing UO_2 and a micromised (UO_2+PuO_2) primary blend	Since 1986 (fabrication plant)	Same advantages as the reference MOX and applicability of its data base. Disappearance of the plutonium-rich agglomerates issue due to dilation. Resolution of the reprocessing issue

2.1 The reference process was applied commercially from the BN Dessel Plant start up in 1973 up to 1984. It consisted in a single blending of PuO_2 powder with free flowing UO_2 powder, resulting in a blend of adequate flowability to feed the pelletizing press. This reference process was used for the partial reloads and fuel supplies for the following thermal power reactors and facilities : Garigliano (Italy), BR3 and VENUS (Belgium), CNA-Chooz (France), Dodewaard (Netherlands), Oskarshamn (Sweden), and NPD (Canada). It was also applied to fuel supplies for the following fast

breeder reactors and facilities: SNR and KNK (Germany , Rapsodie and Phenix (France), DFR and PFR (UK). The MOX fuel produced according to this route was of adequate quality. However for use in LWRs the interdiffusion of Pu and U during sintering was not preventing the presence of high Pu content agglomerates, not acceptable with regards to RIA behaviour experience, and with regards to the reprocessing capability in commercial reprocessing plants currently in operation.

To meet these 2 acceptance requirements, the single blending step of the reference process was replaced by a two-step blending approach:

- in the first step, the pure PuO_2 feed and some UO_2 are co-micronized, resulting in a primary (master) blend of UO_2-PuO_2, the Pu content being typically in the range of 25 to 35 w/o. MIMAS is the BN proprietary acronym derived from this Micronized MASter blending step.
- in the second step, the primary biend is biended down with free-flowing UO_2 to the specified Pu content of the MOX fuel.

The intimate contact between the micronized UO_2 and PuO_2 of the primary blend provides for adequate interdiffusicn during sintering and good solubility in conditions representative of the current reprocessing plants. Larger contact area between the more abundant fine powder of the primary blend and the free-flowing UO_2 powder, feed of the secondary blend results in a less heterogeneous MOX structure than the Reference Process. The data base accumulated in the fabrication and irradiation experience for MOX fuel fabricated according to the Reference Process was therefore conservatively fully applicable to support the design and license of MIMAS MOX fuel.

2.3 The MIMAS process is schematically shown in Figure 1.

One line of production of primary and secondary blends is feeding two lines for pellet and rod production, interconnected to eachother which allows the bypass of some parts of the equipment under maintenance or repair with a reduced impact on the output of the plant.

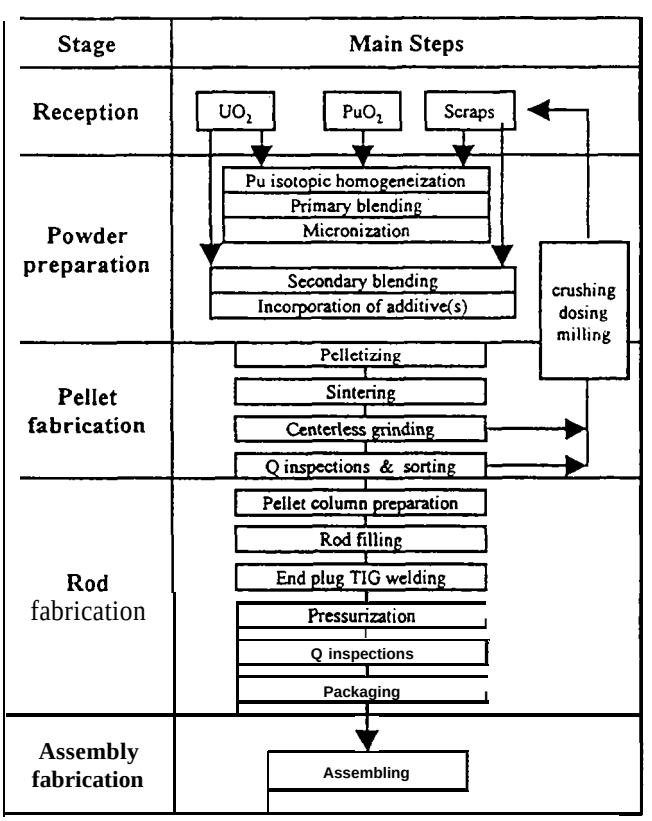

FIG. I. The MIMAS process.

The mean advantages of MIMAS regarding fabrication quality, flexibility and throughput are:

- the micronization step concerns only about 15% of the powder. It leads to a reduced Pu milling time and a small Pu rich dust production;
- the very high flexibility of usage is due on the one hand on the possibility of intermediate storage of the primary blend, and on the other hand to the easiness of cross blending of powders for isotopic homogeneization;
- the process allows a high percenta ge of scrap which can be dry recycled on a routine basis (over 20% in the final product, a.o. by incorporating more than 70% in the primary blends). The scrap issue has been extensively evaluated in terms of the Pu contained in the materials rejected during the fabrication process (various types of powders, pellets, filters....). Currently the BN plant is reaching an equilibrium situation between scrap production and recycling;
- the fuel rod filling and welding can be performed with a minimum intrusion of the cladding into the glove-box, which minimizes the fuel rod surface contamination, decontamination and contamination monitoring;
- the type and limited number of equipment used allow a minimal powder retention level.

3. Fabrication status and records.

3.1 The MIMAS process was implemented during the 1984-1985 refurbishment, taking also into account the returns of experience and improvements from the first 10 years of operation, in the field of fuel fabrication and control process, waste management, safety and safeguards.

The process was progressively qualified by the production of some fuel for irradiation in BR2 MTR and BR3 PWR, and also by the production of 16 MOX assemblies loaded in the French reactor CNA in Chooz. However (see table 2) the most important quantitative step was the start of use of MOX in the French EdF 900 MWe reactors in 1987. Formal qualification from FRAMATOME took place in 1986 with the first EdF MOX reload. Up to 1991, the Dessel plant produced for EdF reactors, with a small fraction for the Swiss power plant of Beznau 1. From 199 1 on, production started for Germany : the plant was qualified for SIEMENS and a first reload was manufactured for UNTERWESER. Diversification went on with, in 1995, the first BWR MOX reloads for Gundremmingen and the first Belgian MOX reloads for Doe1 3 and Tihange 2 respectively. The plant was qualified in 1996 for the Japanese BWR fuel vendors Toshiba and Hitachi. The rods for 2 reloads have been manufactured up to now. The most recent development consists in the current fabrication of ATRIUM 10x10 BWR MOX fuel.

3.2 The refurbishment of 1984-1985 brought the capacity of the plant to a nominal capacity of 35 tHM per year, with a maximum licensed of 40 tHM.

Thanks to the experience gained and the excellent results obtained with the process, these goals have been met (see figure 2)

The nominal fabrication capacity was achieved in 1989, maintained consistently since then at least at that level, approaching even the license limit making available additional fabrication capacities for sale, although the number of fabrication campaigns increased from 2 to 5 or 6 per year in order to meet the delivery requirements of the different customers.

A large number of qualifications were performed, as shown in table 3, confirming the capacity of the MIMAS process to manufacture MOX fuel of any type, with different feed materials. The BN plant is today the only MOX plant producing for BWRs : by the end of the current fabrication campaign, 20 tHh4 of MOX fuel will be produced in 1999 for BWRs.

TABLE 2. MIMAS FUEL COMMERCIAL DELIVERIES

DELIVERY YEAR	POWER PLANT	MOX DELIVERIES
1987	EdF	1
	CNA	2
1988	EdF	2
	Beznau 1	1
1989	EdF	4
	Beznau 1	1
1990	EdF	4
	Beznau 1	1
1991	EdF	4
1992	EdF	2
	Beznau 1	2
	Unterweser	1
1993	EdF	2
	Grafenrheinfeld	1
	Philippsburg	1
1994	EdF	5
	Brokdorf	1
	Electrabel	2
1995	Gundremmingen B	1
	Gundremmingen C	1
	Electrabel	2
1996	Brokdorf	1
	Philippsburg	1
	Electrabel	1
1997	K.Goesgen Daniken	1
	Electrabel	1
	Gundremmingen B	1
	Gundremmingen C	1
1998	TEPCO	1
	Brokdorf	1
	Electrabel	2
	K.Goesgen Daniken	1
	Beznau 1+2	2
1999	TEPCO	1
	Brokdorf	1

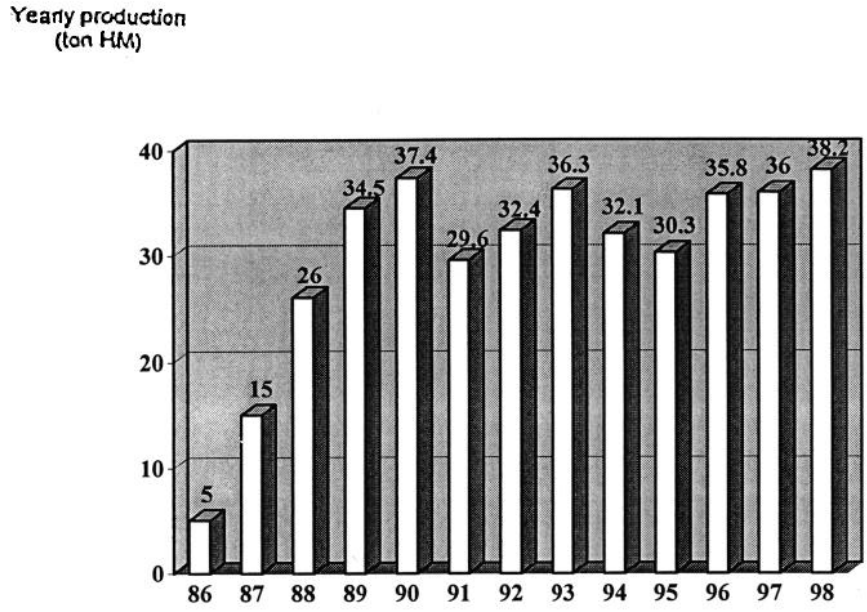

Fig. 2. MOX fabrication record.

MIMAS FABRICATION RANGE OF TESTED PARAMETERS

Cumulated MOX tonnage	**382 tHM**
Fuel rod types (PWR)	**14 x 14, 15 x 15, 16 x 16, 17 x 17**
Fuel rod types (BWR)	**8 x 8, 9 x 9, 10 x 10 now starting**
Qualified MOX fuel designs	**FRAGEMA, SIEMENS, BN, TOSHIBA/HITACHI/JNF**
Size of a fabrication campaign	**4 to 29 tHM**
Number of Pu contents per campaign	**3 to 6**
Pu tot. content in the pellet (over Putot + U)	**up to 8.6%**
Am content (over Putot)	**up to 20,000 ppm**
Putot content in primary blend (over Putot + U)	**25 to 35%**
PuO_2 material	**COGEMA, BNFL**
Cladding materials	**SS304, Zr4, Zr4 Duplex, Zr2, Zr2/Zr linear, Zr2/Fe doped liner**

By April 1999, more than 21 t Pu have been processed in more than 204 000 rods, containing about 388 tHM for use in 796 PWR fuel assemblies and 184 BWR fuel assemblies, loaded, or to be loaded soon, in 17 PWRs and 4 BWRs located in 5 countries (see figure 3).

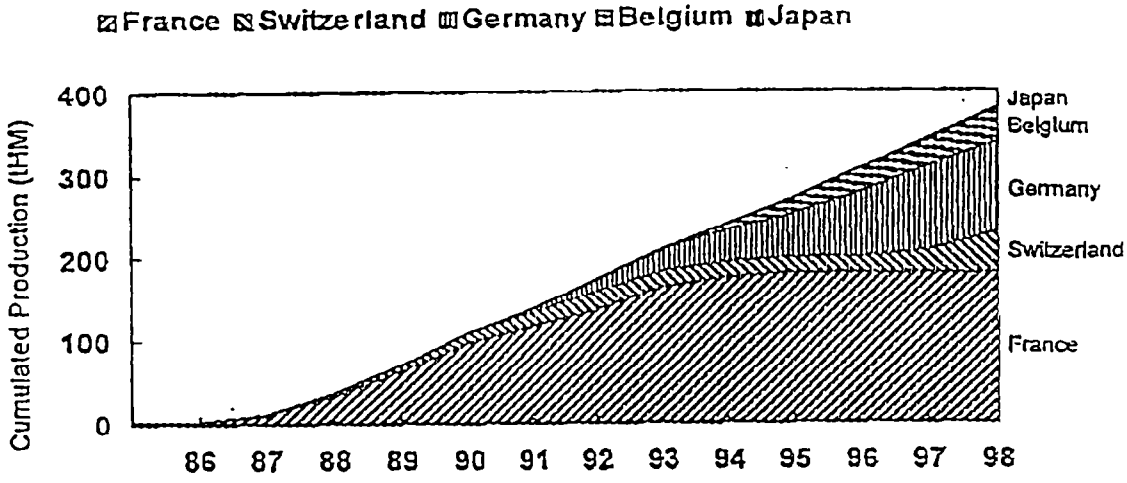

FIG. 3. MIMAS production at Belgonucleaire.

4. MOX fuel inreactor performance

Today 19 LWRs (17 PWRs and 2 BWRs) located in France, Germany, Switzerland and Belgium have been or are in operation with industrial MIMAS MOX fuel delivered by BN. (see figures 4 and 5).

The present burnup performance of MOX fuel is comparable to that of uranium fuel : industrial MOX fuel was unloaded in April 1999 from a Belgian PWR with assembly burnup exceeding 46 GWd/tHM. An increasing number of MOX fuel is discharged in Germany with an assembly burnup of or more than 48 GWd/tHM. The maximum burnup achieved so far was in Beznau 1 : more than 51 GWd/tHM assembly average or 60 GWd/tHM peak pellet, after 6 irradiation cycles

No failures due to the MOX were noticed.

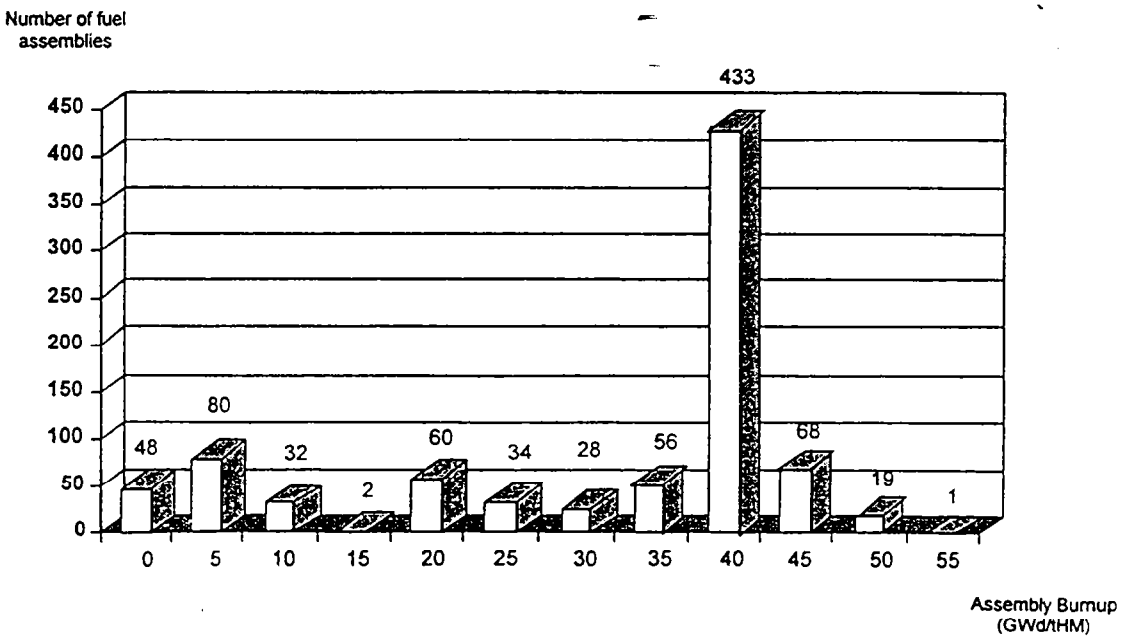

FIG. 4. MIMAS fuel irradiation experience by October, 1998.

	France PWRs SS clad	France PWRs Zr4 clad	Belgium PWRs Zr4 clad	Germany PWRs Zr4 clad	Switzerland PWRs Zr4 clad	Germany BWRs Zr2 clad
6					1 (51 Gwd/t)	
5		1 (51 Gwd/t)			16	
4		3 (44 Gwd/t)	8 (46 Gwd/t)	35 (48 Gwd/t)	43	16 (40Gwd/t)
3	8 (27.5 Gwd/t)	376	20 (44 Gwd/t)	65		16
2	8		20	20	8 (35 Gwd/t)	32
during 1° cycle			8	16	20	36
			24	16	36	24

FIG. 5. Irradiation experience of BN MIMAS fuel in LWRs (commercial deliveries) anticipated by October 1999 number of assemblies per irradiation cycle.

5. Evolution of the Applied Technology

As a result of the decreasing quality of the PuO_2 and of the increasing quantities of PuO_2 being processed, programmes are continuously conducted aiming at decreasing the occupational doses of the personnel and at decreasing the waste generated by the plant operation.

The simultaneous increase of the quantity of the plutonium to be processed and of the neutron emission can lead to an increase radiation exposure of the personal : from 1995 to 1999, a four year, modernization program has been undertaken in the mondernization and/or mechanization

of the production equipment and infrastructure, including extensive shielding and remote control This modernization was performed without loss of capacity and has helped to reduce the collective and maximum individual doses by a factor 3 since 1987. Since 1997, the dosimetry at BN complies fully with the European directive translating the 1990 recommendations of the I.C.R.P. publication n° 60.

Waste originating from plant operation influences fabrication costs and personnel exposure. By identifying the waste generating operations, by optimizing the concerned fabrication process step and by educating the personnel with regard to those fabrication process steps and with regard to more adequate sorting of the operation waste, the volume of the fabrication waste over the last years has been progressively reduced 1.3 m^3 per ton HM in 1996, 1.0 m^3 per ton HM in 1998 while the Pu contained in those waste has been reduced to less than 0.1% of the Pu contained in the delivered MOX fuel.

6. Conclusion

The BN MOX plant started industrial production in 1986 and has reached its nominal capacity of 35 tHM per year in 1989, which is maintained consistently since then, at least at that level, approaching even the license limit of 40 tHM per year.

A little less than 400 tHM has been produced. The production is now very flexible, for various countries and designs, for PWRs as well as BWRs.

By the end of 1998, BN had fabricated about 40% of the worlds MOX industrial production.

The MIMAS process, developed by BN, has been transferred to COGEMA in France, where it is applied since 1995 with success in CFCa Cadarache and Melox. Taking into account COGEMA's fabrication, the MIMAS process represents 74% of the world's cumulated MOX production.

The excellent fabrication performance as well as the outstanding in-pile behaviour of the MIMAS fuel makes it the reference for any new MOX plant in the world. It has been recently selected for use in the USA in the frame of the weapon grade material disposition programme. It is a strong candidate for the J-MOX plant in Japan.

THE UNATTENDED NDA MEASUREMENT SYSTEMS AT MOX FUEL FABRICATION FACILITIES IN BELGIUM

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Abstract

In the framework of the NPA arrangements, unattended NDA measurement systems have been designed, developed and installed in Belgium at Belgonucleaire (WBNP) and Franco-Belge de Fabrication de Combustibles (WBFP) by IAEA and Euratom with the co-operation of Belgonucleaire and FBFC.

The simultaneous neutron and gamma unattended measurement system for the PuO_2 powder cans in the canister (KOKER) and the unattended PNCL measurement system for MOX pins in the box (BAR) have been tested and implemented for routine use at Belgonucleaire.

At WBFP, the installation of an unattended PNCL measurement system for MOX assemblies has been completed.

The implementation of these unattended systems has resulted in significant reduction of the inspection effort for the IAEA.

1. INTRODUCTION

Since the 1970s the inspection regime at Belgonucleaire for Safeguards control purposes was the Joint-Team. This consisted of simultaneous continuous inspections by both the **IAEA** and Euratom. However, in 1992, a New Partnership Approach (NPA) was formulated between both inspectorates. One of the aims of the NPA was to achieve manpower savings for the IAEA in Euratom countries.

In the Joint-Team, all fuel measurements were performed manually in the presence of inspectors from the IAEA and Euratom. The transition from Joint-Team to NPA, where the IAEA is present less frequently, involved the introduction of unattended, continuously operating measurement systems, with appropriate containment and surveillance devices (C/S) independently verifiable optical-electronic sealing systems and video surveillance.

'At Belgonucleaire, following the conception of an NPA system, equipment was installed from 1994 to 1996. Both equipment and procedures were extensively tested from 1996 to 1997, before coming into routine operation in October 1997.

At WBFP, equipment has been installed and tested since 1997, while software for data evaluation was being developed. It is expected that routine operation will commence in 1999.

2. RELEVANT SAFEGUARDS ASPECTS

Belgonucléaire is a large MOX fuel fabrication plant. The input materials are cans of PuO_2 powder within canisters (KOKER) received from COGEMA, La Hague, and UO2 powder in drums. The output products are MOX pins, which are stored and transported in metal boxes (BAK).

FBFC, Dessel is the fuel fabrication plant for manufacturing the MOX assemblies from fuel pins.

At WBNP systems were installed for the simultaneous measurement of gamma and neutron of PuO_2 powder cans in canisters (KOKER) on their receipt. The KOKERs are identified and measured before entering the store location. After the measurement, the KOKER is moved and stored in the storage pit and a metal shielding plug is placed on the top. A VACOSS seal is applied to each storage location by a EURATOM inspector.

The PNCL (Plutonium Neutron Collar) measurement system is installed around the box (BAK) containing the MOX pins. The BAKs are identified and measured before entering the store location. VACOSS seals are attached at each end of the BAK in party line mode by EURATOM inspector. The BAK is then transferred into the store and is ready for transfer to WBFP.

Both KOKER and BAK stores are under joint C/S, being VACOSS seals and video surveillance.

At WBFP, the unattended measurements are performed on the assembly by PNCL.

The IAEA inspect WBNP once a month for a short inventory verification (SIV), in order to evaluate the data collected from the unattended measurement stations, to verify the electronic seals (VACOSS) and to review the surveillance system. Euratom is present on a continuous basis.

At WBFP, the IAEA inspector is at the plant for a timeliness verification once a month, to evaluate the data collected from the unattended measurement stations, to verify the VACOSS seals and to review the surveillance system. An IAEA inspector is at the plant for all shipments of MOX assemblies to the nuclear power plants. Euratom performs all these inspections together with the IAEA and, in addition, further inspections without the IAEA to ensure that all C/S and unattended measurement systems are functioning correctly, and to evaluate data more frequently.

3. INSTALLED EQUIPMENT

The philosophy common to the measurement systems is the provision of duplicate data acquisition systems, in two cabinets accessible only in the presence of both inspectorates. Branching to a third cabinet enables access by Euratom at all times without IAEA presence. For the surveillance systems, duplicated recording is provided within one cabinet, with VACOSS seals completing dual C/S.

3.1 At WBNP

PuO_2 Receipt Measurement Station

This unattended station consists of both neutron and gamma measurement systems which operate simultaneously. The data from the neutron measurements are stored in a data logger. An adjustable support for the Ge detector allows measurements at five different positions on the Koker corresponding to the five cans inside.

The NDA electronic equipment is installed in three sealable cabinets as follows

Cabinet# 1 contains

Gamma chain: high voltage power supply for the HPGe detector, main amplifier and ADC, a PC with a S 100 card running software to capture and store the gamma spectra.

Neutron chain: A signal splitter, a JSR12, providing high voltage and Amptek power supply and receiving an output from the n-signal splitter. The JSR12 is logged by a Rustrak ranger data logger. Above a certain threshold level the neutron loggers switch to a higher recording rate corresponding to an event (KOKER measurement). The logger is interfaced with the PC to allow downloading.

Cabinet#2 contains

Gamma chain: a main amplifier, an ADC and a PC with a S 100 card .

Neutron chain: a JSR12 receiving a second output from the splitter in Cabinet#l and a logger for the JSR12 output. The logger is interfaced with the PC.

Cabinet#3 contains

the same equipment as Cabinet#2 and is used by Euratom only.

BAK Measurement Station

The PNCL is installed on a movable cart which allows the detector to be placed around the BAK for measurement. The operator positions the instrument on the BAK.

A JSR-12, HV unit and power supply ,computer system with Shift Register Collect software is installed in each of two cabinets. Cabinet#l and Cabinet#2 are sealed with common seals.

The MOSS (Multiple Optical Surveillance System) and Uniplex back-up system with 8 cameras is installed for the KOKER store and BAK store. There are two cameras in he KOKER store; one is used for reading the KOKER ID number and the other is used to cover all of the storage positions and to check the integrity of the measurement station.

There are six cameras in the BAK store. One is mounted on the BAK loading machine to read the BAK identification number. One to cover the whole measurement station and the way into the BAK store position. Two cameras are placed on opposite sides of the store to cover the whole roof area of the room. The other two cameras cover the incoming door for the BAK and the outgoing door for the BAK.

Sealing System

VACOSS seals are applied in the KOKER store and to each BAK. Every BAK is sealed with two VACOSS seals, one at each end. Every KOKER storage location is sealed by one VACOSS seal on the top of the shielding plug.

3.2 At WBFP

Fuel Assembly measurement station
LWR-MOX fuel assemblies are measured using an unattended PNCL station. The surveillance system is UNIPLEX with duplicate recording. VACOSS seals are used to seal the fuel assemblies into their storage locations. The assembly is lowered into the PNCL by crane and remains at a fixed position during the measurement. The PNCL detector head is attached to an adjustable platform that allows it's position on the assembly to be changed for a measurement to be made at any position along the assembly. The ID no. of the assembly is identified using a camera above the measurement position. The neutron monitor is used to detect the movement of the fuel assemblies in and out of the storage vault (known as the bunker). The MOX fuel assemblies are stored under multiple C/S system (VACOSS/Surveillance). The transfer route of the fuel assemblies is covered by cameras from their storage locations in the bunker until they are placed into the shipment container.

4. SOFTWARE PACKAGES

The data acquisition and evaluation software systems that are used for the simultaneous neutron and gamma unattended measurement of the KOKERs were developed by Euratom. For evaluation the combination of neutron and gamma data (recorded on duplicate computers) is made by time of event. Full multiplication correction, decay to the date of measurement, calibration and MGAPu analyses are incorporated into the software to enable comparison with operators Pu mass and Pu isotopic composition declarations.

The Shift Register Collect and Shift Register Review are used for the BAK measurement station and were developed by LOS ALAMOS National Laboratory under the US support program.

For the FBFC MOX fuel assembly measurements, the data acquisition and evaluation modules were developed by Euratom (Ref. 1). Neutron data (5 second counts) is stored as one file per day from each of primary and secondary chains on duplicated computers. Visualisation of the measurement count rate combined with date/time and calibration enables association and comparison of a measurement with its declared mass-isotopic data.

5. CONCLUSIONS

Both unattended measurement systems for the measurement of KOKER and BAK installed at Belgonucleaire have been thoroughly tested and routine use started in 1997. Performance has been reliable. Both systems were designed and developed to have redundancy, so that when a failure of the system occurred the problem would be solved without causing any additional burden to the facility operator. Any failure of any cabinet which is under control by both organisations is informed to the Agency in a timely manner by Euratom and the problem is solved with out any delay for the activities of the plant operation.

At WBNP , the Agency spent 150 mandays in 1997 and 1998 as opposed to the 250 mandays in 1996. At WBFP, the unattended measurement system was tested and has been in regular use since March 1999. An Agency inspector will be present at the plant once a month for timeliness verification and performs all the activities for LWR-MOX shipment. It is expected to spend 60 mandays per year instead of the 115 mandays in 1998 for this facility.

1. "EURATOMs Remote Acquisition of Data and Review (RADAR)" P. Schwalbach et al., ESARDA Sevilla 4-6 May 1999.

CONTINUOUS MONITORING OF PLUTONIUM SOLUTION IN A CONVERSION PLANT

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Abstract

This paper describes the implementation of a safeguards Tank Monitoring System (TAMS) in a Plutonium Conversion Plant (PCP). TAMS main objective is to provide the International Atomic Energy Agency (IAEA) (the Agency) with continuous data for safeguards evaluation and review of inventories and flows of plutonium solutions. It has been designed to monitor, in unattended mode, the inventory of each tank and transactions of solutions between tanks, as well as to confirm the absence of borrowing plutonium solutions from and to a neighboring reprocessing plant. The instrumentation consists of one electronic scanner that collects pressure data from electromanometers connected to the tank dip tubes, one uninterruptable power supply and one personal computer operating in a Windows-NT environment. The pressure data transmitted to the acquisition system is saved and converted to volume and density values, coupled with a graph capability to display events in each tank at intervals of 15 seconds. The system operation has not only strengthened the safeguards measures in PCP but also reduced inspection effort while minimizing intrusion to normal plant activities and radiation exposure to personnel. TAMS is a powerful, reliable tool that has significantly improved the effectiveness of safeguards implementation at PCP. The future combined use of TAMS with remote monitoring (RM) will further enhance efficiency of the safeguards measures at PCP.

1. INTRODUCTION

PCP is a mixed oxide (MOX) co-conversion facility that receives plutonium and uranium in the form of nitrate solution produced from spent fuel of the domestic Light Water Reactors (LWR) at a reprocessing plant. The facility uses a microwave heating direct denitration process to convert Pu-U nitrate mixed solutions into MOX powder. The newly produced MOX is stored in canisters until it is shipped to the MOX fuel fabrication plants for production of fast breeder reactor (FBR) fuel.

During normal plant operations, several shipments of plutonium nitrate solution are received every year. The typical batch size of one shipment amounts to a few tens of kilograms of plutonium.

The IAEA applies to PCP a safeguards approach based on a 2 MBA structure. MBA1 includes the solution tanks and the process line where there are 2 parallel microwave heating denitration apparatus followed by a calcination and reduction furnace, milling and sieving machine [4]. The process produces MOX powder that is temporarily stored in intermediate canisters before being blended on batch basis and filled into PCP cans. Up to 4 cans are put in a canister and

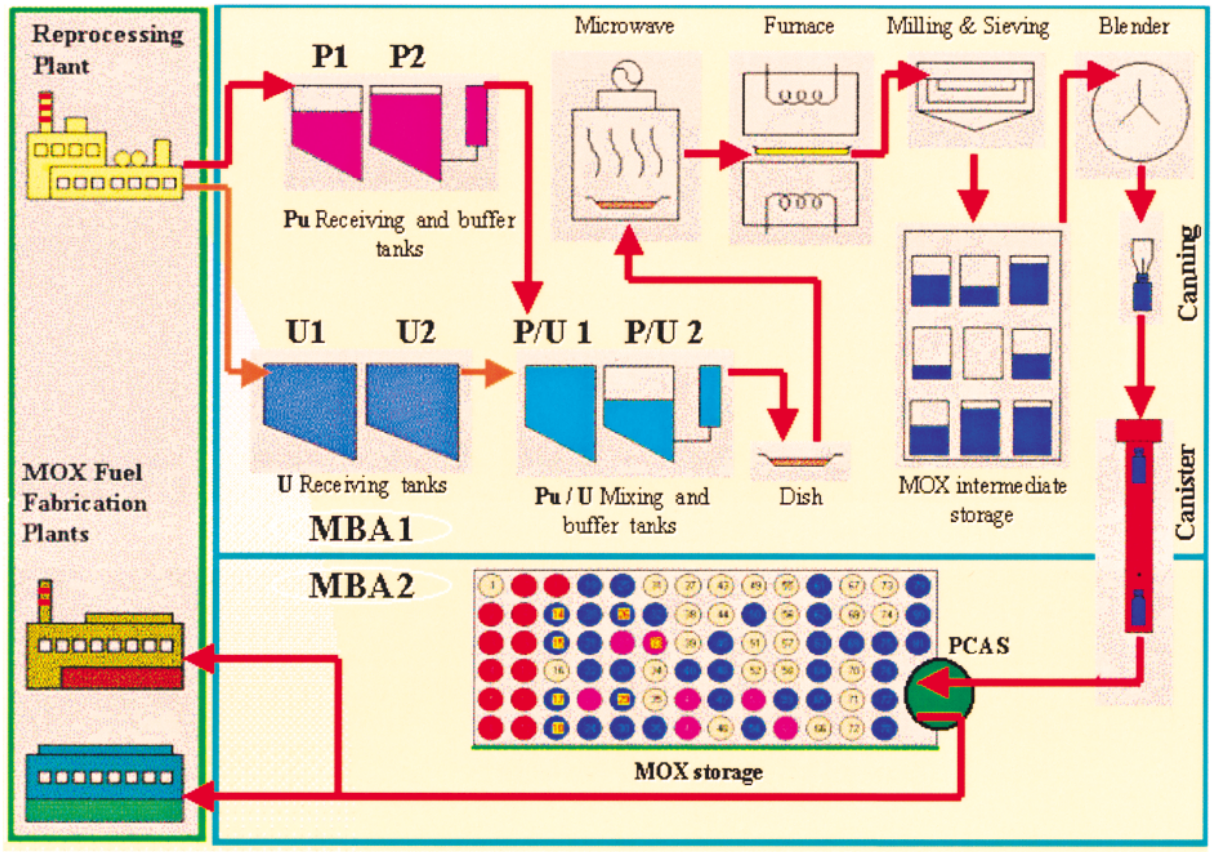

FIG.1. Flow of nuclear material at PCP

transferred to the MBA2, the MOX product storage. This storage area accommodates up to 81 canisters. Figure 1 above shows part of the fuel cycle and internal flow of nuclear material through PCP.

In MBA1, at the front end of the process line, there are four equally shaped annular tanks for plutonium nitrate and plutonium-uranyl nitrate solutions connected to each other by over-flow pots. Each tank has a nominal liquid volume of 300 liter and a nominal liquid level of 1200 mm. Because the plutonium nitrate is highly corrosive and radioactive, these tanks are located in two concrete shielded cells. One cell encloses the plutonium nitrate accountability input tank (P1) and the plutonium nitrate buffer tank (P2). The other cell contains the plutonium nitrate-uranyl-mixing tank (P/U 1) and the plutonium nitrate-uranyl storage tank (P/U 2) which feeds the microwaves of the co-conversion process line.

Until recently the Agency used a conventional accountancy method to verify the tank's inventories. This method consisted of reading of the pressures, measured by the operator's electromanometers, to determine the volume and the density of the liquid content within each tank. Inspectors validated the tank data by the calibration of the electromanometer with Agency's standard weights and took solution samples to determine the concentration and isotopic composition of the nuclear material by partial or bias defect tests.

Although this verification was independent, its accuracy still rested on the operator's system. In addition, the method required periodical, intrusive validation practices, which increased the workload of the operator and inspectors [1].

With a view to increase efficiency and effectiveneTs of the safeguards approach for PCP, the Agency and the State System of Accountancy and Control (SSAC) opted to install and operate a solution monitoring system. The use of process monitoring techniques most suited the goals to improve safeguards capabilities.

2. TAMS CONCEPTUAL DESIGN

2.1 Objectives

The Agency defined the design objectives of TAMS taking into account the economic impact of such system and, most importantly, its safety aspects. The design of the tank monitoring system was based on the results obtained and on the experience gained in previous work on monitoring solutions in accountancy tanks [2], and [3]. The objectives were as follows:

- To maintain continuity of knowledge of the nuclear material involved in transfers, flow and inventory of solutions in tanks.
- To minimize inspector's and operator's workloads and efforts.
- To reduce intrusion into normal plant operation and
- To guarantee that the plant is operated as declared by the operator and no changes were made on the design and operation procedures of the front end of PCP.

2.2 Requirements

To be effective, and in order to meet the above objectives, TAMS had to satisfy the following requirements:

- **overall system:**
  To complement the existing Agency safeguards measures.
- **adequate coverage:**
  To monitor any volume change and be capable to independently measure the solution volume and density at any time.
- **sensitivity:**
  To be sufficiently sensitive to detect pressure changes beyond a preset trigger level
- **reliability:**
  To be designed for high reliability so as to provide an overall high confidence level even in case of power failure.
- **tamper indication:**
  To be capable to indicate any attempts to alter the measurement parameters on hardware or software.
- **non-interference:**
  The system or its components should not interfere with normal operations of the facility.
- **output data** :
  The output data obtained from the system, in conjunction with data from other sources should enable the inspectors to determine at any time the volume and the density of the plutonium solution in any tank of PCP.
- **adaptability** :
  The system to be designed initially to collect data "in situ". And should also be suitable to be adapted for remote monitoring operation at a later stage.
- **availability:**
  All equipment and associated software should be of modular design and commercially available.

2.3 Project Milestones

The Agency invited engineering firms for competitive international bidding to provide detailed specifications after defining the basic requirements of TAMS in June 1996. Award of contract, based on best price and delivery time, was granted to Tsukasa Sokken Co.Ltd. in September 1996. The Agency approved engineering documents and specifications in November of that year followed by system procurement in December. By April 1997, TAMS was installed and after commissioning, the Agency tested TAMS in May 1997. The system was authorized for routine safeguards use in July 1997 and since then has been operated successfully.

2.4 Description and components

Each of the four tanks containing plutonium solution at PCP is equipped with a set of three dip-tubes of 1.2 cm in diameter. The shortest one called reference dip-tube or **R,** measures the pressure above the solution surface. The deepest tube measures the pressure at the lowest possible position in the tank and is referred to as the level dip-tube (major) or **L.** And the third dip-tube located in an intermediate well determined position, normally 20 cm above the end of the level dip-tube, is called the density dip-tube (minor) or **D**. The dip-tubes are continuously fed by a constant airflow rate of 6~7 Nl/h for differential pressure measurement, and the bubbling frequency is about two per three seconds.

The instrumentation piping for TAMS is connected with a Swagelok type T-piece to the operator's transmitters piping followed by a valve system to enable isolation when needed. Isolation of TAMS from the operator's system is required to perform maintenance without interfering normal plant operations. The entire system, as illustrated in figure 2, is contained in four identical sensing

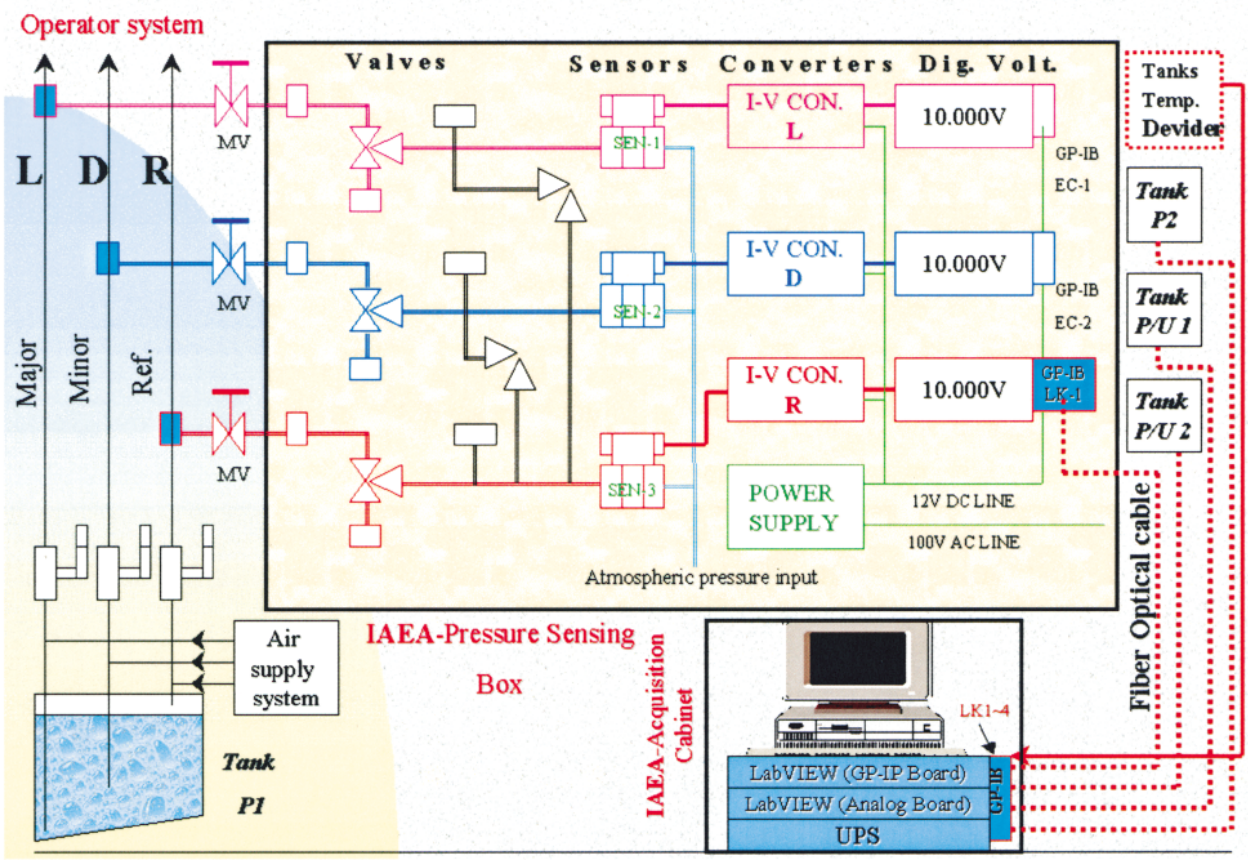

FIG.2. Schematic diagram of TAMS and its connection to the operator's system

boxes, one acquisition cabinet and one temperature splitterbox. The Agency seals all these tamper-proof boxes so as to keep continuity of knowledge on the hardware and setting parameters. Inspectors can easily confirm the integrity of the system by visual checks on the components, piping and cables.

The basic elements of a sensing box are:

- Three high accuracy (better than 0.05% full-scale), capacitance type, pressure sensors connected to the three dip-tubes of each tank. The differential capacitance between the sensing diaphragm and the capacitor plates is a current, which is directly proportional to the process pressure. [5j
- Three current to voltage converters and
- Three digital voltmeters

A fiber optic cable connects each sensing box to the acquisition system through GP-IB (General-Purpose Instrumentation Bus) boards.

The components of the acquisition cabinet are:

- One electronic scanner to collect pressure data in sequence for each measurement point.
- One personal computer operating in Windows NT environment coupled with a color monitor.
- One uninterruptable power supply.
- One interface; GP-IB board of 16 inputs and
- A software to run the acquisition system using Lab-VIEW language.

The temperature splitter box connects TAMS to the thermocouple emerged in each of the four plutonium solution tanks to supply the system with the actual solution temperature **(T).**

2.5 Installation and Acceptance test

The Agency and the SSAC planned and executed the installation of TAMS so as to prevent interference with operator's instruments. Therefore the Agency carried out acceptance and authentication tests that included the major following activities:

- **Documentation:** Reviewed at the Agency and at site to meet IAEA requirements.
- **Authentication and integrity:** All components were visually checked including piping, connections, cables, valves, sensors, converters, digital voltmeters and the whole acquisition system in order to meet the specified tolerances and as built drawings dimensions. Vulnerability of the hardware, software and cabinets were checked to include tamper indication provisions.
- **Functional tests:** All input ports were checked by feeding constant air flow and pressure values were read through digital voltmeters and acquisition system to satisfy the following points:
  - Independent confirmation of **L, D,** and **R** pressures and their sensitivities.
  - Confirmation of minimum pressure change.
  - Confirmation of system response.
  - Confirmation of circulation effect on pressure value.
  - Confirmation of periodical recording.
- **Maintenance:** Periodic preventive work to check performance of individual components and to identify replaceable modules.

Figures 3,4 and 5 show TAMS major components as built and installed at PCP.

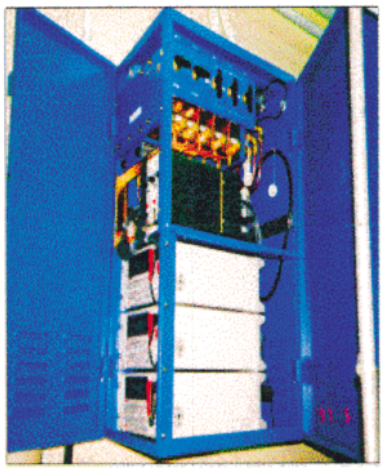

FIG.3. Inside of a sensor box cabinet

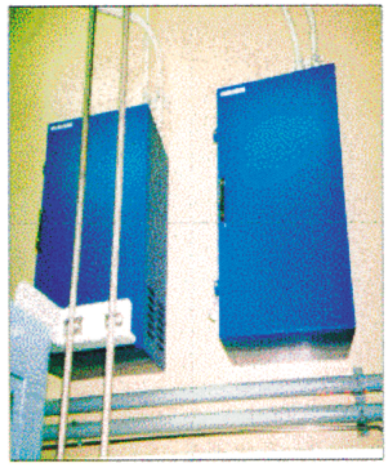

FIG.4. Sensor Boxes

FIG.5. Inside of acquisition

3. TAMS PERFORMANCE

3.1. Data collect

The system periodically scans the pressure signals from **R** (reference), **L** (major) and **D** (minor) dip-tubes of each tank in intervals of 15 seconds. The reading frequency, as shown in figure 6, is set to be 3 times for **R** (1, 6, 11), 4 times for **L** (2, 4, 7, 9), and 4 times for **D** (3, 5, 8, 10) followed by one time reading of the temperature **T** (12). The mean pressure values, **R**, **L**, **D**, and their standard deviations are calculated and saved before starting to read from the next tank. Therefore, the data acquisition frequency for each tank is once per minute.

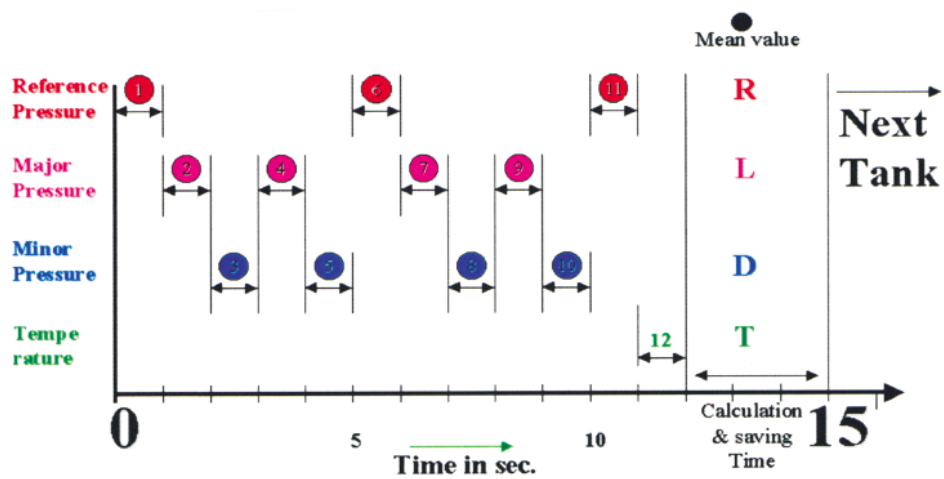

FIG.6. Collecting measurement data in one tank

3.2. Data save

The calculated mean value is compared with the previous value used as reference. If it is within the preset trigger level it is not saved. If it falls outside the preset trigger level then it is recorded in ASCII format file and starts to trigger a new reference value as shown in figure 7. When there is no change in the reading values, the system filters the data and periodically records one data

set every 10 minutes. The size of each data set which contains information on tank number, date, time, **R**, **L**, **D**, STDs, **T**, density, level and volume, as tabulated in figure 8 below, is 90 bytes/15 s saved in two different media; hard disk and magneto optical (MO) disk.

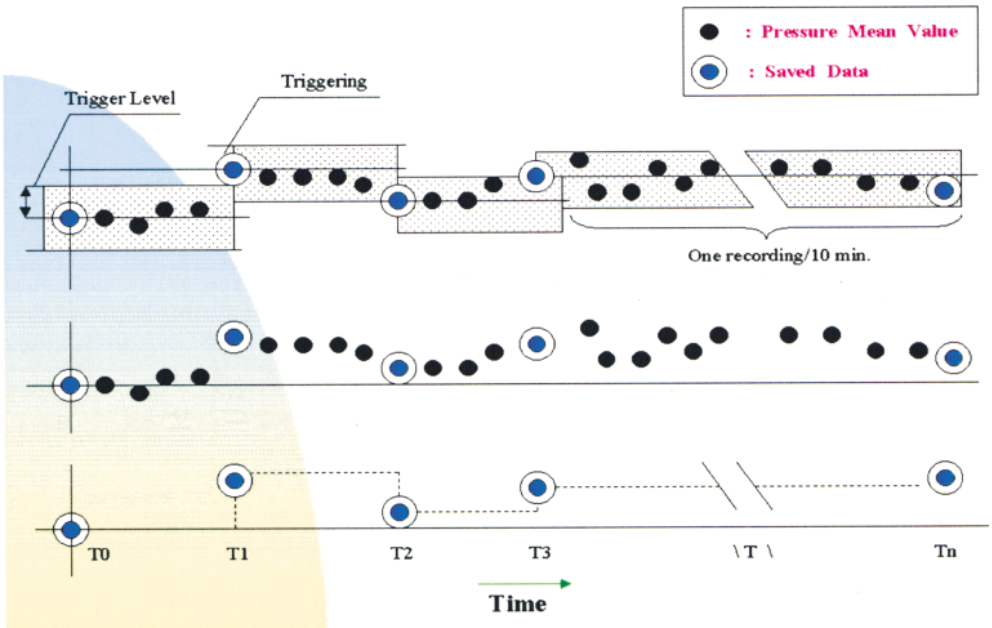

FIG.7. Filtering and saving measurement data

Whenever the system is serviced, normally during interim inspections, the MO disk is replaced and just after the first set of pressure data is collected, a new ASCII file is created in both media to save newly acquired data until the next service.

Tank No.	Date MM/DD/YYYY	Time HH:MM:SS	Ref.(Ave) kPa	Major(Ave) kPa	Minor(Ave) kPa	Ref.(STD) kPa	Major(STD) kPa	Minor(STD) kPa	Temp. deg.C	Dens. g/cm3	Level mm
1	07/01/199	00:05:31	-0.6496	1.0075	-0.1216	0.0002	0.0008	0.0001	25.2	0.6	289.4
2	07/01/199	00:04:46	-0.6507	15.3171	12.5599	0.0005	0.0006	0.0009	39.0	1.4	1142.6
3	07/01/199	00:05:01	-0.6539	11.7354	8.9038	0.0005	0.0018	0.0010	33.6	1.5	861.6
4	07/01/199	00:05:16	-0.6481	12.1294	9.1842	0.0010	0.0013	0.0018	34.1	1.5	870.0
1	07/01/199	00:15:31	-0.6515	1.0066	-0.1216	0.0004	0.0005	0.0001	25.1	0.6	289.8
2	07/01/199	00:14:46	-0.6488	15.3189	12.5623	0.0002	0.0007	0.0012	38.8	1.4	1142.9
3	07/01/199	00:15:01	-0.6532	11.7368	8.9039	0.0003	0.0006	0.0006	33.6	1.5	861.3
4	07/01/199	00:15:16	-0.6480	12.1302	9.1857	0.0005	0.0015	0.0011	34.2	1.5	870.2
1	07/01/199	00:25:31	-0.6491	1.0077	-0.1217	0.0003	0.0009	0.0001	25.2	0.6	289.2
2	07/01/199	00:24:46	-0.6490	15.3179	12.5625	0.0004	0.0011	0.0019	38.9	1.4	1143.3

FIG.8. Tabulated data as saved in ASCII file

3.3. Data review

TAMS represents a significant advance in the capability of inspectors to quickly and consistently make accurate volume determination for the control and accountability of plutonium solutions at PCP. The MO disk is reviewed at the facility so as to evaluate all events that took place since the previous inspection. The review package is a Lab-VIEW application, configured with constants and calibration parameters to simultaneously display graphs of two tanks with a printing option. Inspectors are capable to display the variation of any of the data set collected on mean pressure values (kPa) of **R**, **L**, **D** and their standard deviations, on temperature (degree C) density (g/cm3), level (mmH2O) or volume (l) versus time. The system sensitivity is such so as to detect change in pressure of one Pa while the overall system uncertainty is better than 0.5 %.

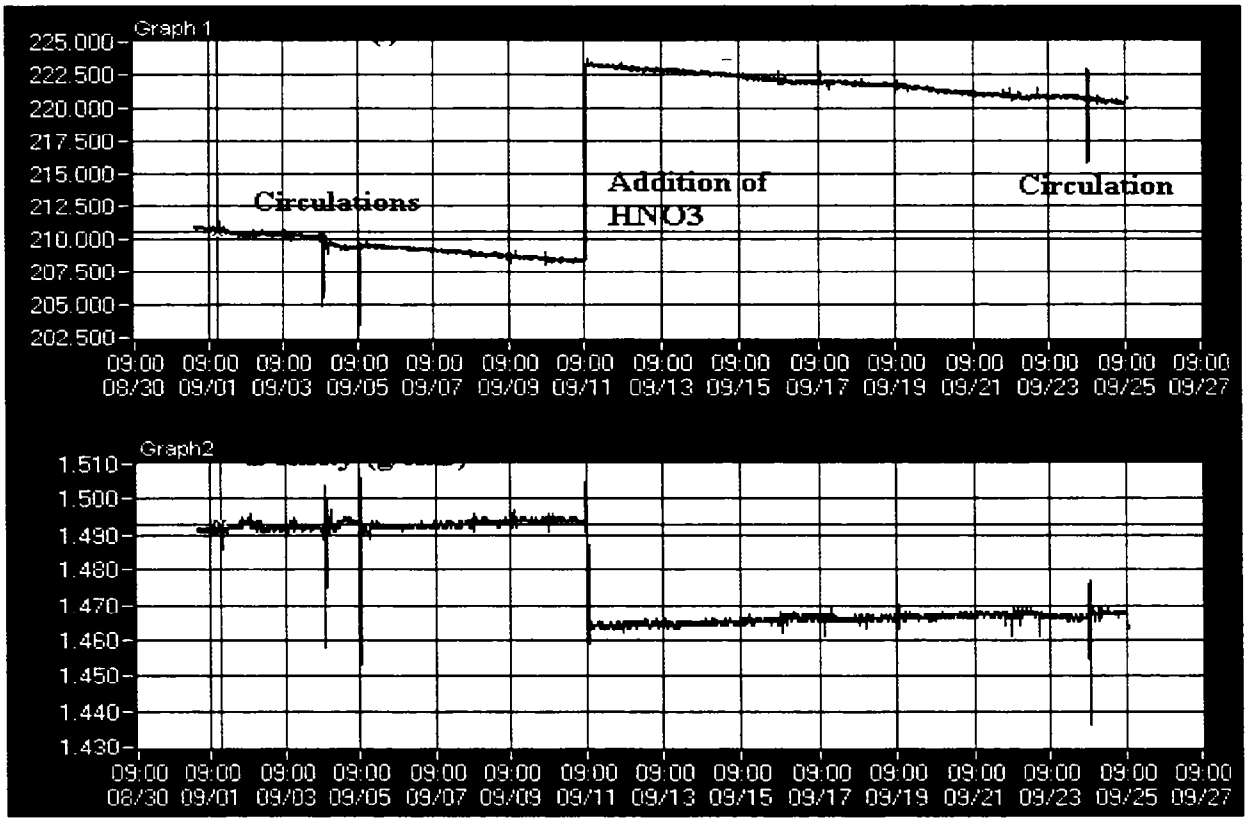

FIG.9. Plotted data showing 3 circulations and 1 addition of nitric acid

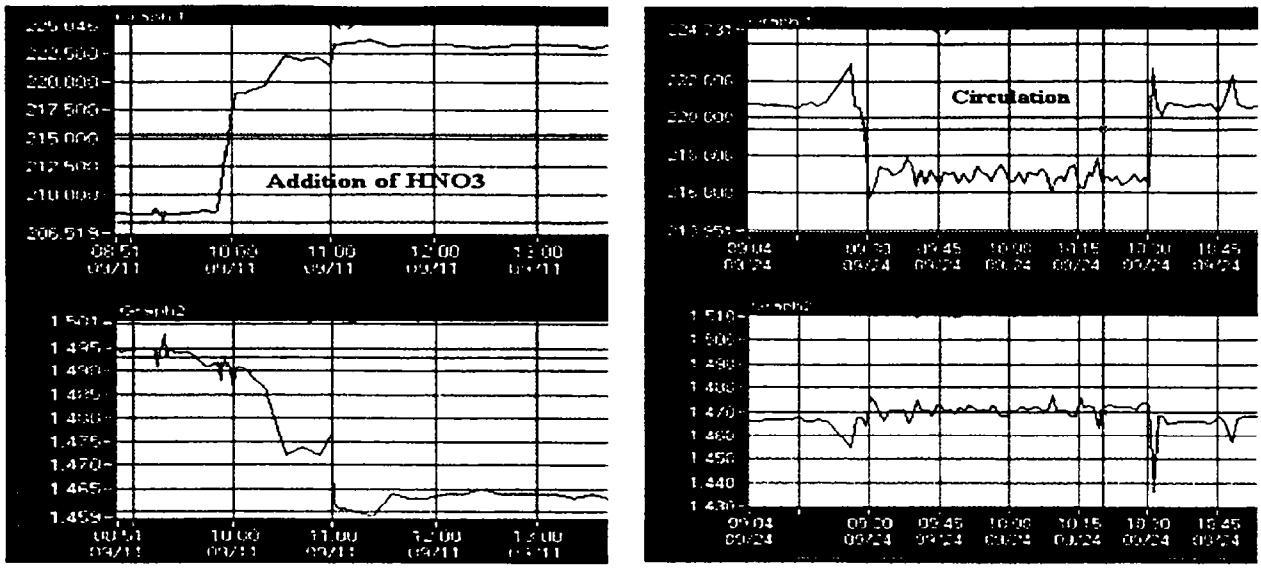

FIG.10. Addition of nitric acid reflected in volume and density (left) and circulation of 1 hour reflected in volume and density (right).

Until now, TAMS has performed as expected in maintaining continuity of knowledge on the PCP plutonium solutions. Equally important, it has shown a high sensitivity in detecting all changes in pressure values that confirm the operator's declaration, i.e. circulation and transactions, or events caused by natural phenomena such as evaporation. TAMS has provided the Agency a means to

verify declared operations of plutonium solution and, concurrently has helped to assure the absence of undeclared operations. Figure 9 shows the addition of nitric acid in one of the four tanks to compensate for evaporation. The graph clearly shows when about 15 liters of acid were added on September 11, at 10:00 hour. Figure 10 illustrates the detailed changes when that addition took place and the circulation of the solution before sampling for verification.

4. CONCLUSIONS

The system provides, in unattended mode, a continuous record of all transactions of nuclear material in the tanks. It has yielded substantial savings in inspection efforts while enhancing overall safeguards implementation. TAMS is an easy to use, valuable instrument that assures the Agency that plutonium solutions are adequately accounted for in a timely manner.

TAMS has been designed to be adapted for remote monitoring in the future. Data collected and remotely transmitted will provide the Agency with real time knowledge of inventories and transactions of plutonium solutions.

The determination of the Agency and SSAC and the cooperation from the operator brought the TAMS project to a successful completion.

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MOX FUEL FOR INDIAN NUCLEAR POWER PROGRAMME

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Abstract

A sound energy policy and a sound environmental policy calls for utilisation of plutonium (Pu) in nuclear power reactors. The paper discusses the use of Pu in the form of mixed oxide (MOX) fuel in two Indian boiling water reactors (BWRs) at Tarapur. An industrial scale MOX fuel fabrication plant is presently operational at Tarapur which is capable of manufacturing MOX fuels for BWRs and in future for PHWRs. The plant can also manufacture mixed oxide fuel for prototype fast breeder reactor (PFBR) and development work in this regard has already started. The paper describes the MOX fuel manufacturing technology and quality control techniques presently in use at the plant. The irradiation experience of the lead MOX assemblies in BWRs is also briefly discussed. The key areas of interest for future developments in MOX fuel fabrication technology and Pu utilisation are identified.

1. INTRODUCTION

From the very beginning of the Indian nuclear programme, Dr. Homi Bhabha, our founder, had laid great emphasis on (i) self reliance and (ii) efficient use of nuclear energy resources. The three stage Indian nuclear power programme is mainly based on these twin objectives. The first stage envisages use of natural uranium in Pressurised Heavy Water Reactors (PHWRs). The second stage envisages use of plutonium (Pu) generated in PHWRs in Fast Breeder Reactors (FBRs) and third stage envisages exploitation of our large reserves of Thorium using $Th\text{-}U^{233}$ fuel cycles.

The implementation of the second stage calls for development of U-Pu based fuel cycle for FBRs and one important part of this is the development of (U-Pu) mixed oxide (MOX) fuels. The third stage requires development of ($Th\text{-}U^{233}$) MOX fuels. In order to make the transition from second stage to third stage somewhat faster we now envisage the use of Advanced Heavy Water Reactors (AHWRs) which need fuels based on (Th-Pu) MOX [11. It is also felt Pu recycling in the existing nuclear power reactors (BWRs and PHWRs) will provide much needed technology base for manufacture of (U-Pu) MOX and compliment the needs of the second stage programme.

Thus it can be seen that MOX fuel in Indian context has its presence in all the three stages of the nuclear power programme. It also extends to all the three major actinides viz. U, Pu and Th as well as all the main nuclear power reactor systems viz. LWRs, PHWRs, ATRs and FBRs.

The paper outlines the fabrication, quality control and irradiation experience of MOX fuel in BWRs at Tarapur. It also makes reference to development work being carried out on MOX fuel fabrication for the Prototype Fast Breeder Reactor (PFBR) envisaged in the next decade. The key areas and technologies required for the manufacture of MOX fuels for higher burn-up in PHWRs, Th based MOX fuels for AHWRs are also briefly discussed.

2. MOX FUEL DESIGN FOR BWRs

The MOX fuel assembly (MOXFA) design currently used at BWRs Tarapur (TAPS) is “All Pu” type consisting of 6x6 square array. The hardware used and the assembly structure is similar to that used with standard low enriched uranium oxide (LEU) fuel except that segmented rod forms the water rod in case of MOXFA. The current MOX fuel design does not use any burnable poison. The

MOXFA incorporates fuel rods of 3 enrichments of (U-PurMOX and the reactor employs a 3 batch cycling of 18 months with average MOX fuel burn-up of 16,000 MWd/T [2]. Detailed safety studies (including design basis accidents and operational transients) carried out indicate that with this type of design, core loading upto 40% (112 out of 284 assemblies) is feasible at the present cycle length. However for further increases in core loading 7x7 MOXFA design with four Gd rods and four water rods may be required and is under study. This advanced MOXFA is also expected to permit higher flexibility in fuel management as well as cycle length.

3. IRRADIATION EXPERIENCE AT TAPS

A number of short length MOX experimental fuel clusters were irradiated in our research reactor CIRUS, at BARC, Trombay in the 80s which helped in finalising the fabrication flow sheet and the fuel material specifications to be followed for BWR MOX fuel [3].

Two "All Pu" full length 6x6 lead MOX assemblies were first loaded in TAPS-l commercial BWR in June '94. Subsequently four more MOX assemblies were loaded in TAPS-2 in May '96. As of December '98 the two lead MOX assemblies are in the third cycle in TAPS-l and six more MOX assemblies at various burn-up and power levels in TAPS-2. Table I gives details of irradiation levels of these MOX assemblies. The MOXFAs have always been loaded near the Travelling Incore Probe (TIP) locations to closely monitor the assembly power. Sipping tests have been carried out at the end of each cycle and these tests indicate satisfactory behaviour of the MOX fuel.

TABLE I. MOX IRRADIATION EXPERIENCE IN TAPS (BWRs) AS ON DECEMBER 1998

Sr. No.	Assembly No.	Reactor No.	Loading Date	Present Core Location	Burn-up MWd/T
1.	MB-726	TAPS-l	June '94	33-10	11880
2.	MB-727	TAPS- 1	June '94	19-40	12246
3.	MB-870	TAPS-2	May '96	17-36	7974
4.	MB-87 1	TAPS-2	May '96	33-12	8668
5.	MB-872	TAPS-2	May '96	23-38	923 1
6.	MX-006	TAPS-2	May '96	09-28	9304
7.	MX-007	TAPS-2	October '98	33-22	951
8.	MX-008	TAPS-2	October '98	09-22	847

4. MOX FUEL FABRICATION EXPERIENCE

Advanced Fuel Fabrication Facility (AFFF), BARC is presently manufacturing MOX fuel for commercial BWRs at Tarapur. The flow sheet of fabrication is based on mechanical mixing and milling of UO_2 and PuO_2 powders first developed at Radiometallurgy Division (RMD) at BARC, Trombay but several modifications and upgradations were carried out to improve the quality and scale up of operations. After detailed safety evaluations and several progressive partial clearances, the Atomic Energy Regulatory Board (AERB) gave the formal authorisation for production of MOX fuel in January, 1994 and the first two MOX assemblies were delivered to TAPS in May, 1994. The plant is one among the only two plants (the other being MDF of BNFL, Sellafield, U.K.) in the world using Attritor technology that produces micro-homogeneous fuel which is expected to be superior in performance and also easy to reprocess [4]. The conventional ball milling as used by some of the European MOX manufacturers creates a potential for dust dispersion within the glove box leading to higher operator exposure. The high temperature sintering of MOX pellets is carried out in ceramic tibre lined furnaces specially designed for easy maintenance in glove box conditions. The fully automated wet centreless grinding machine using composite diamond grinding wheel gives the required pellet surface finish and cut in one pass and is fitted with on-line coolant clarification and circulation system. The hot vacuum degassing of MOX pellets just before encapsulation ensures low

hydrogen content in MOX pellets. The fuel end plugs are closed using automated TIG welding technique.

As well known the radiotoxicity of Pu demands high integrity containment systems during MOX fuel fabrication which are not required for UO_2 fuel fabrication. For handling large quantities of Pu through industrial machinery many industrial and radiological safety issues have to be addressed. The multilevel passive and active containment systems at AFFF have been so designed that there is high degree of reliability in industrial and radiological safety. The glove box technology which is of fundamental importance has been improved with passive controls, digital instrumentation and superior materials of construction. The nuclear ventilation system is very carefully designed with multiple HEPA barriers, 100% standby with regards to equipment, power supply, interlocks and operated by programmable logic controllers (PLCs) with Y2K compliance.

The process machinery selected is not only rugged and easy to maintain but meets the high precision engineering product requirements. The process and the equipment undergo thorough testing and safety evaluation prior to adoption in the fabrication line. The equipment and the plant layout ensures faster material movement, minimum waste generation, radiological and criticality safety. The excellent radiological safety standards of the plant are clearly evident from Table II. This is the result of elaborate preventive, protective, detective and monitoring systems adopted for the radiological safety in the plant.

TABLE II. DATA ON RADIOLOGICAL SAFETY DURING MOX FIUEL FABRICATION

Parameter	Annual Exposure		
	1996	1997	1998
Collective dose Man Sievert per Ton of MOX	0.10	0.07	0.06
Maximum individual dose - mSv/yr.	2.15	1.90	2.10

5. SCRAP RECYCLE

The scrap generated during MOX fuel fabrication is broadly divided into clean rejected oxide (CRO) and chemically impure dirty rejected oxide (DRO). The sintered MOX pellets rejected due to physical integrity defects constitute the major fraction of CR0 and the centreless grinder sludge constitutes the major fraction of DRO. In UO_2 fuel fabrication the scrap generated is wet recycled using dissolution and precipitation methods. MOX scrap is recycled by dry processes to enable quick recycle and also to avoid any generation of liquid wastes. Quick recycle is essential in MOX fuel because Pu isotopic is likely to change from batch to batch and also due to radioactive decay.

Additionally the flow sheet was closely scrutinised and many new process control techniques/points introduced to minimise the waste/scrap generation. These measures have resulted in reduction of scrap generated from around 20% to less than 10%. At these levels the entire CR0 scrap can be dry recycled into the feed powders after appropriate thermo mechanical treatments [5]. The diameter and density variation of the as sintered MOX pellets was found to be much smaller when about 20 wt. of oxidised MOX scrap was added prior to Attritor milling of the feed materials [61.

Our preliminary investigations indicate the sludge from the diamond centreless grinder is virtually free from metallic impurities and hence can be dry recycled. The main chemical impurities seen are non-metallic viz. Carbon, nitrogen and chlorine. These could be easily removed by oxidation and pyrohydrolysis. Sinterability studies carried out indicate that upto 15 wt.% DRO addition can give acceptable quality pellets [7]. It is now hoped that with these advances MOX fuel fabrication can boast of no alpha liquid waste generation and achieve near zero solid scrap on net plant output.

6. QUALITY CONTROL

Although the scale of operation with MOX fuel is order of magnitude lower than that of UO_2 and data base on fuel performance very limited, nuclear power utilities expect MOX fuel to perform equally well if not better than UO_2. This challenge has been very well accepted by the MOX fuel designer and the fabricator by incorporating an elaborate quality control plan on a total quality control (TQC) principle. The quality control and the manufacturing flow sheets are so integrated that product is checked not only at the end of the flow sheet but at various intermediate stages so that any deficiency is corrected at the earliest opportunity. Figure 1 gives the simplified MOX manufacturing flow sheet with process/quality control points. In order to realise the dream of MOX fuel technology free from liquid waste it is necessary to use innovative quality control techniques. Our experience indicates that if the isotopic content of Pu is held within close limits by appropriate blending measures then use of coincident neutron counting or gamma assaying can be adopted for determination of Pu composition in MOX pellets and chemical determination be restricted to only border cases/random checks [8]. Similarly gamma auto radiography (GAR) which is simple and nondestructive method gives most of the information on Pu homogeneity - both macro and micro [9] and hence frequency of dissolution tests can be reduced. Also GAR is cost effective for detecting the presence of Pu rich clusters on pellet surface which may affect the transient behaviour of the fuel. At AFFF all the MOX rods go through 100% GAR test prior to assembly.

7. FUTURE CHALLENGES

The future challenges in MOX fuel fabrication are classified into medium term (200 1 - 2005) and long term (after 2005) challenges. The medium term challenges are with regards to further improvement of MOX fuel performance and processing technology in respect of PHWR and BWR MOX fuels. The long term challenges (after 2005) are with respect to fabrication of MOX fuel for PFBR and AHWR but the development work and feasibility studies have to be started immediately. Some of these issues are discussed in the following para.

7.1. Medium term challenges (2001-2005)

Indian nuclear power programme is at present mainly based on PHWRs and use of MOX here can not only improve fuel utilisation but also reduce spent fuel volumes. A preliminary study carried out earlier had indicated that an island type of MOX bundle (with 7 inner elements of MOX and 12 outer natural UO_2 elements) the present PHWR average bum-up can be increased from 6,700 MWd/t to 10,700 MWd/t. Further studies to design PHWR MOX bundles capable of reaching burn-up of 20,000 MWd/t are presently taken up. The increased fission gas release (FGR) is likely to be an issue to be addressed in high bum-up PHWR MOX bundles. Among various approaches to tackle FGR issue, studies on fabrication of annular pellets and higher grain sized MOX fuels for PHWR are taken up at AFFF. The plant is currently working on development of higher grain sized plastic (soft and ramp resistant) MOX (Table III) in line with such development being done abroad for UO_2 fuels [10].

Our work on TiO_2 doped MOX fuel, its characterisation and irradiation experience has been reported earlier [l l] but the present work is likely to improve steady state as well as transient FGR performance [123. Figure 2(a) and Fig. 2(b) shows the microstructure of standard and higher grain size silicate doped Plastic MOX.

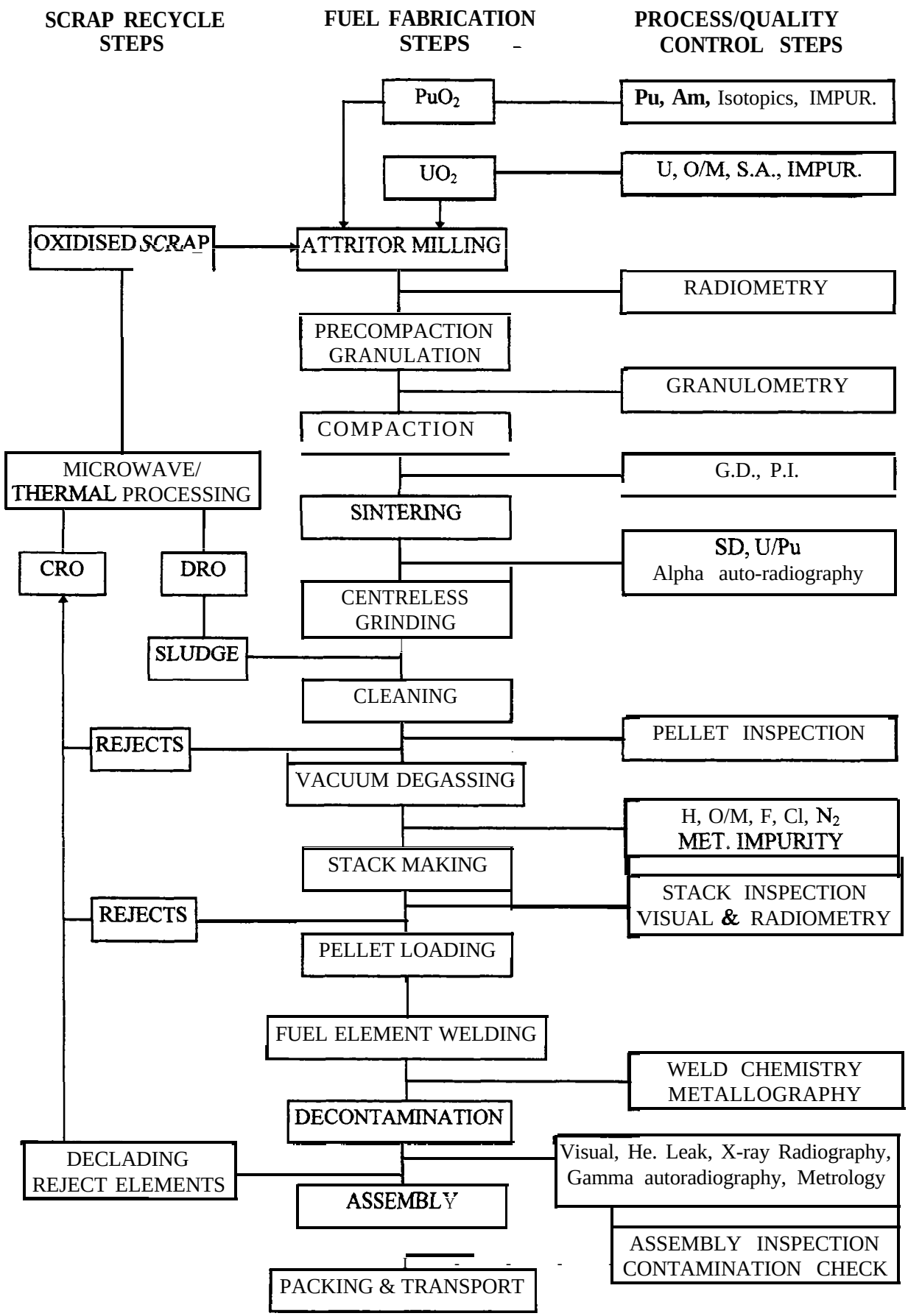

FIG. 1. FLOW SHEET FOR TOTAL QUALITY CONTROL OF MOX FABRICATION

MICROSTRUCTURE OF MOX

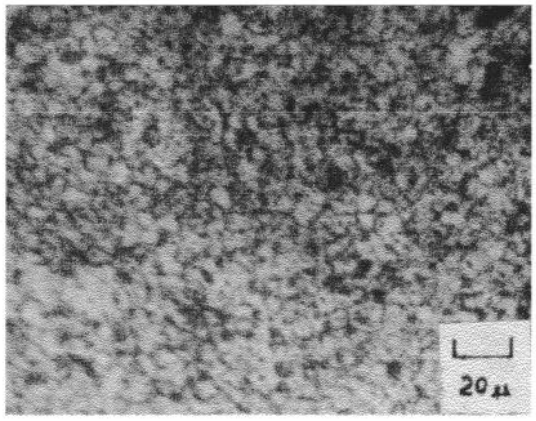

Fig. 2(a) Standard MOX

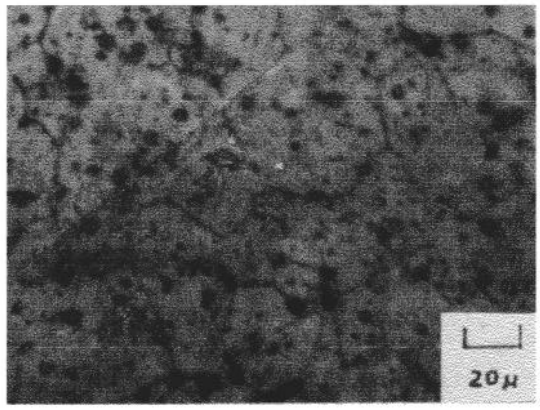

Fig.2(b) Ramp-Resistant Soft MOX

PFBR FUEL DEVELOPMENT

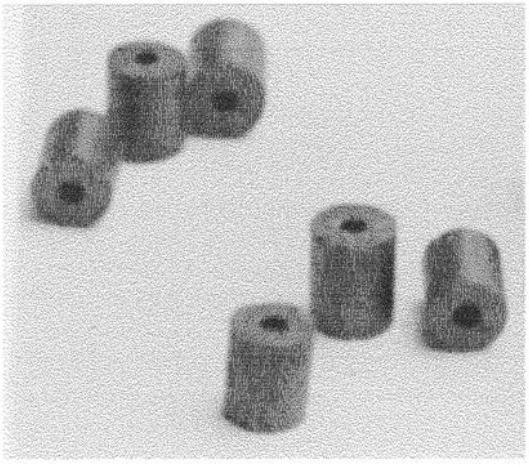

Fig.3(a) Annular Pellets

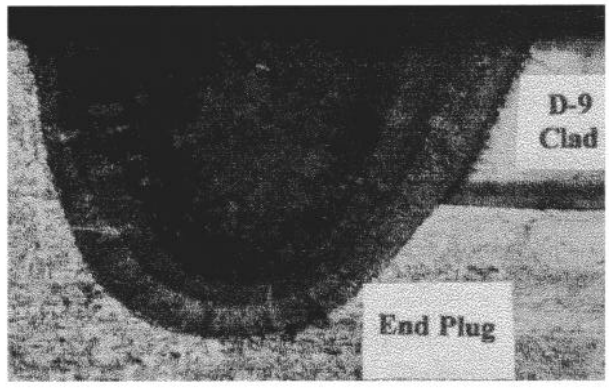

Fig.3(b) D-9 End Plug Weld Profile

TABLE III. CHARACTERISTICS OF SILICATE DOPED RAMP RESISTANT MOX PELLETS

Sr. No.	Dopant Wt.%	Normal density % T.D.	Average grain size
1.	Std. MOX (No dopant)	94.1	7
2.	0.1	94.3	28
3.	0.2	94.2	35

Microwave energy is gaining increasing acceptance as thermal source for speciality ceramic processing though it is material specific. Fortunately oxides of uranium are good absorbers of microwave making them amenable to be processed by this technique. At AFFF microwave furnaces developed with appropriate modifications for use in glove box environment under corrosive conditions are being used for thermal pulverisation of rejected MOX pellets and oxidation of DRO sludge. Studies on dissolution of UO_2, MOX and ThO_2 as well as denitration are currently carried out on small scale and show a definite advantage with respect to dissolution kinetics and product characteristics [13, 14]. The main advantage of microwave co-denitration is that it is the only wet process available for production of actinide oxides that practically generate no liquid waste.

7.2. Long term challenges (after 2005)

India envisages building of the PFBR of 500 MWe in the early part of the new millennium. The salient features of the PFBR and its fuel is given in Table IV [16].

The proposed fuel is an advanced version fast reactor fuel with D-9 cladding, annular pellet geometry and low O/M features. Identification of technology required for PFBR fuel has already been started. Development work for production of annular pellets using conventional and custom built mechanical, hydraulic and rotary presses as well as weldability studies on D-9 clad material (full austenetic solidification mode) using pulse TIG and laser welding (in collaboration with centre for advanced technology [CAT], Indore, India) are in progress. Figure 3(a) shows typical annular pellets being developed for PFBR and Fig. 3(b) gives metallography of trial end plug weld on PFBR fuel pin. Advanced agglomeration techniques using powder rolling & extrusion, near-net shaping, dry centreless grinding, real time radiography, precision radiometry for Pu assay are some of the other areas where development work is being initiated to meet the challenge of PFBR fuel fabrication. Microwave co-denitration is also being considered for mixed UO_2-PuO_2 fuel powder as alternate to mechanical milling.

The other important challenge to be tackled during the next decade is the technology development for fabrication of MOX fuel for AHWR. The AHWR fuel is a 52 pin cluster arranged in square lattice [1]. Some of the rods are (Th-Pu) MOX and the rest are (Th-U^{233}) MOX in the fuel assembly. Fabrication of Th MOX is easy in some respects and difficult in some other respects. The difficulty comes with respect of handling U^{233} containing U^{232} and its daughters which are hard gamma emitters. Hence Th MOX has to be fabricated in shielded cells using remote handling techniques. The advantage in respect of Th MOX fuel fabrication (even under remote operation) is due to single oxidation state of Th which permits sintering (or any other thermal treatments) to be carried out without protective atmosphere - even in air. The difficulty of remote operation and maintenance of high temperature furnaces is very much reduced and economics improved due to this characteristics.

VIPAC technique looks attractive for Th-U^{233} remote fabrication and detailed studies on smear density, stability and defect performance are initiated. The high density microspheres required for VIPAC can be fabricated by sol-gel technique or advanced agglomeration techniques. The sol-gel method of MOX feed production completely eliminates powder handling and reduces radio-toxicity hazards at the MOX plant. The process is based on internal gelation principle and the microspheres can be used either for pellet production or for VIPAC fuels [15]. Based on the extensive laboratory scale investigations carried out at Fuel Chemistry Division, BARC, Trombay a larger scale plant is under erection at AFFF, Tarapur. Sol-gel offers definite advantage for handling multicycled Pu and Th-U^{233} MOX as the microspheres are free flowing and hence fabrication operations can be more easily automated.

Impregnation of U^{233} on ThO_2 pellets or on sol-gel microspheres can be other approach possible [17]. A suitable permutation and combination of different techniques such as VIPAC, Sol-gel, co-precipitation, Impregnation, advanced agglomeration, air sintering can be considered. Table-V gives some of the combinations on which development work will soon be started.

TABLE IV. SALIENT FEATURES OF PFBR-500 AND ITS FUEL

Thermal power MWe	1250
Electric output MWe	500
Fuel material	$(U\text{-}Pu)O_2$
Inpile inventory	9.2 Tons
Fuel composition	UO_2 - 2 1% PuO_2 & UO_2 - 28.4% PuO_2
Number of assemblies	181
Pins per assembly	217
Fuel clad material	20% CW D9
Fuel rod length mm	2580
Fuel pin dia mm	6.6
Pellet outer dia mm	5.52 ± 0.04
Dia of central hole mm	1.6
Linear mass gm/cm	2.33 ± 0.08
O/M	1.95

TABLE V. R&D AREAS FOR REMOTE FUEL FABRICATION OF $(Th\text{-}U^{233})O_2$ FUEL

Sr. No.	Type of fuel	FABRICATION STEPS	Remarks
1.	VIPAC	(a) $Th\text{-}U^{233}$ Sol-gel microsphere (b) Air sintering (c) Loading & VIPAC (d) Laser welding & assembly	(1) All steps in shielded cells (2) Remote Fab. Feasible (3) Stability and defect behaviour to be studied
2.	VIPAC + IMPREG	(a) ThO_2 sol-gel/agglomeration & pre sintering (b) U^{233} nitrate impregnation (c) Heat treat (d) Load, VIPAC & weld	(1) Step (a) carried out in normal facility (2) Steps (b) and (c) in shielded cells (3) Behaviour likely to be similar to Sr. No. 1
3.	Pellet (SGMP)	(a) Co-ppt/Microwave denitration/Sol-gel (b) Consolidation (c) Air sintering (d) Encap & Assembly	(1) All steps in shielded cells (2) Remotisation difficult (3) Good stability
4.	Pellet + IMPREG	(a) ThO_2 comp. & pre sintering (b) U^{233} nitrate impregnation (c) Heat treat (d) Encap & Assembly	(1) Step (a) carried out in normal facility (2) Remotisation easier than Sr. No. 3 (3) Good stability expected

Thorium fuel cycle meets the three basic requirements of sustainable nuclear power generation viz. (i) Lower long living TRU wastes (ii) High-abundance of fuel in self-sustaining mode leading to resource assurance for future generations (iii) High chemical stability leading to excellent environmental safety. The single oxidation and strict stoichiometric state with open fluorite lattice as well as better thermal properties (Higher MPt, thermal conductivity and lower expansion co-efficient) of ThO_2 are pointers to improved chemical, mechanical, thermal, irradiation and defect behaviour of Th based MOX fuels through actual in-pile experience at present is very limited. The developed countries are realising that Th based MOX is not only excellent fuel material but also the best waste matrix to immobilise the actinide (weapon Pu disposition) [18] as well as high level activity with leach rates far better than vitrified glass [19]. The concept of ROX (Rock like MOX) in which a fuel matrix slowly transforms to a unleachable mineral rock incorporating the residual actinides and fission products as it completes its target burn-up in a nuclear reactor (so that it is ready for direct disposal) is possible only with Th based MOX [20,21]. India with its large data base and resources of Th as well as expertise in materials technology can play an important role in developing such cer-cer composites though we value ThO_2 as fuel rather than waste fixing matrix.

8. CONCLUSION

A sound energy policy and a sound environmental policy calls for recycling of fissile materials in power reactors. Closing the fuel cycle and developing MOX fuel cycle technologies are essential for sustainable development of nuclear power. India is one of the handful countries today which have demonstrated the capability to both fabricate and utilise MOX fuel in its commercial power reactors. In the years to come this capacity will grow further to encompass the area of FBRs and AHWRs. The process development and identification of technologies based on the feed back from the tonnage scale MOX fuel fabrication at AFFF, BARC is already in progress. We may certainly say at this stage that India is getting ready to fuel the future - the MOX way!

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FUEL DESIGN, PERFORMANCE AND TESTING UNDER NORMAL AND OFF-NORMAL CONDITIONS

(Session III)

Invited Paper

OVERVIEW ON MOX FUEL FOR LWRS: DESIGN, PERFORMANCE AND TESTING

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Abstract

This overview looks at the historical background to the design, performance and testing of LWR MOX fuel over the last 30 to 40 years. It briefly examines the scenarios which encouraged the development of MOX fuel for utilisation in LWRs and looks at the design changes required on moving from UO_2 to MOX fuel. The paper summarises the national irradiation testing programmes, the commercial developments and performance data obtained throughout this period, highlighting those aspects which have had an impact on manufacturing and design choices. The paper thus provides the historical background information for the contributed papers in Session 3 (Fuel Design, Performance and Testing) of the symposium.

1. INTRODUCTION

It has always been recognised that recycled plutonium is most effectively and efficiently utilised in Fast Breeder Reactors (FBRs). However, in the 1950's it was generally accepted that available reprocessing capacity would exceed the requirements of the FBR prototypes pianned at that time and a number of countries developed strategies which involved utilisation of the excess available plutonium in Light Water Reactors (LWRs) over an intermediate period of 10 to 20 years prior to deployment of FBRs. This led to the instigation of a programme of R&D work conducted within the framework of the US Atomic Energy Commission (AEC) -Euratom Joint Programme to promote nuclear energy.

The USAEC began plutonium recycling studies in 1956. The work was concentrated in two programmes, 'Plutonium Utilisation Programme' (PUP), carried out at Hanford National Laboratory using test reactor irradiations and the 'Saxton Programme' which was managed by Westinghouse and demonstrated recycle in a small PWR with MOX loadings starting in 1965. US MOX development had, by 1975, progressed to the commercial demonstration stage but commercial utilisation was delayed by the preparation of the "Generic Environmental Statement on the Use of Mixed Oxides" (GESMO) which US AEC organised to facilitate the industrial application of plutonium recycle [l] and by the availability of Pu from commercial reprocessing plants. With President Carter's Executive Order on non-proliferation in 1977 which indefinitely postponed commercial reprocessing

in the US, all research on MOX fuel in the US was abandoned by 1980 and the data gathered in earlier years has now been largely overtaken by experience in Europe where commercial MOX use is a reality.

In Europe, the Euratom sponsored work was carried out by Belgonucleaire (BN) and the Belgian National Laboratory (CEN/SCK) at Mol and the world's first LWR MOX assembly was loaded into BR-3 in 1963 followed by a MOX loading in the German BWR at VAK (Kahl) in 1966.

In the 1960's, as interest in plutonium recycle grew, an increasing number of development programmes were initiated in various European countries to investigate MOX utilisation in LWRs. Germany, Switzerland, France, UK, Italy, the Netherlands and Sweden all, to a greater or lesser extent, began work in this area. In the early 1970's, in parallel with the US AEC's 'GESMO' preparation, the Commission of the European Communities sponsored a "Research Programme for Plutonium Recycling in LWRs" to complement and co-ordinate the development programmes being pursued at national level.

Germany led the way in the 1970's in terms of introducing MOX fuel into commercial LWR power stations with Switzerland following closely behind. Today 12 of the 19 operating plants in Germany have been licensed for MOX loadings and Switzerland and Belgium have between them 5 PWRs licensed for MOX usage.

In France a full Reprocessing-Conditioning-Recycling strategy was chosen in the 1970's and studies performed between 1980 and 1985 confirmed the feasibility of recycling plutonium in PWRs. In 1985 EdF decided to recycle MOX in their PWRs and the Safety Authorities, having studied the Safety Report submitted in early 1987, gave permission in October 1987 for the first MOX reload which was then introduced to the St Laurent Bl core. Today 20 of the 28 available 900 MWe PWRs in France are licensed for MOX and 17 have MOX fuel loadings. Moreover, since 1991 two reactors containing MOX fuel assemblies have been operated under load follow and frequency control conditions for several cycles and as a result all reactors licensed for MOX in France have, since 1995, been authorised to operate in the hybrid mode and under load following conditions.

In the UK, thermal reactor development concentrated on MAGNOX reactors, which are based on metal fuel, and Advanced Gas-cooled Reactors (AGRs) which utilise UO_2 fuel. The UK's strategy on plutonium recycle concentrated on the FBR and only a very small programme of MOX fuel development for use in thermal reactors was carried out in the 1960's and 70's with irradiations in Windscale AGR and SGHWR. However, with the government decision in 1987 to abandon FBR development, the UK, through BNFL, decided to build on its experience in manufacturing MOX fuel for FBRs and enter the thermal MOX fabrication business. Consequently, this has led to a major involvement in LWR MOX design, performance and testing over the last 10 years as the company demonstrates the viability of its MOX fuel to the industry.

In Japan MOX use in LWRs has been actively pursued since the late 1980's with the irradiation of a number of lead test assemblies in Mihama 1 reactor in the period 1988 to 1991 [2]. Prior to this, plutonium fuel research in Japan concentrated on FBR fuel and fuel for the Advanced Thermal Reactor (ATR). The ATR is a heavy water moderated, light water cooled, pressure tube reactor designed by PNC (now JNC) and data on fuel for this reactor is applicable to LWR MOX utilisation. Various irradiation tests of MOX fuel have been carried out in ATR as well as transient tests in the Halden Boiling Water Reactor (HBWR) to study performance under reactivity initiated accident conditions [3,4]. Japan continues with its policy of implementing thermal MOX usage and plans to construct new advanced reactors which will be fuelled with full-MOX cores.

2. FUEL DESIGN

MOX fuel assembly design has been universally based on the UO_2 design with only two minor modifications relating to the neutronic and thermal mechanical properties of the MOX fuel itself.

The neutronic design of the MOX assemblies is based on the simple principle that the assemblies must be equivalent (or as near as possible) to assemblies containing UO_2 fuel in terms of dissipated power, total reactivity and accumulated burn-up. This equivalence is not completely satisfied because of the differences in neutronic properties of U^{235} and Pu^{239}/Pu^{241} isotopes. The design of the MOX assemblies has therefore been optimised to reduce the power peaking at the UO_2 and MOX interface and thus obtain the lowest possible shape factor - this is achieved by zoning the assembly using three different plutonium enrichments in a concentric distribution in the case of PWRs and up to seven in the case of BWRs.

The second design change, viz. increasing plenum volume in the rods, is sometimes applied to accommodate the higher fission gas release expected of MOX fuel. Because of the neutronic properties of the Pu isotopes in MOX fuel, the reactivity decreases less rapidly with burn-up than in UO_2 fuel and thus MOX fuel dissipates more power later in life releasing more fission gas; in addition, the thermal conductivity of MOX is known to be lower by a few percent than that of UO_2 which may give rise to higher fuel temperatures and thus slightly higher fission gas release.

With these exceptions, the fuel rod and assembly design are essentially unchanged from that of the equivalent UO_2 assembly. It is generally the case that if a modification is applied to UO_2 then the same change will be incorporated in MOX some time later.

3. IRRADIATION TEST PROGRAMMES

The world's first LWR MOX fuel irradiation began in 1963 in the Belgian BR3 PWR; the fuel was manufactured by BN. From 1963 to 1986 the reactor was used extensively to test and qualify successive MOX fuel types. Variations in feed material characteristics, fabrication processes, U^{235} and Pu contents, cladding materials, rod diameters and assembly geometries were among the parameters tested; unpressurised and pressurised rods were also tested. BR3, which was shutdown in 1987, offered a broad range of operating conditions and together with the test reactors at Petten and Halden, provided an important data base in support of LWR MOX utilisation.

For more than 18 years BN and CEN/SCK have jointly managed a set of international programmes designed to provide fuel validation and licensing data in support of MOX utilisation in LWRs [5,6]. Organisations involved have included fuel designers and manufacturers, research laboratories, utilities and other nuclear service providers. Such international prograrnmes are mutually beneficial in terms of information and cost sharing. Table 1 lists the most recent of the MOX fuel programmes which are designed to give information on fuel behaviour.

TABLE 1. RECENT INTERNATIONAL TEST IRRADIATION PROGRAMMES

Experiment	Fuel Type
PRIM0	PWR
CALLISTO	PWR
DOMO	BWR
FIGARO	PWR
NOK - M109	PWR
NOK - M308	PWR
GERONIMO	BWR

The PRIM0 and DOMO programmes were completed in 1994 and 1996 respectively. Both focused on fission gas release and fuel microstructure at high burn-ups in PWR and BWR environments. From these experiments the MOX rod failure threshold has been shown to be similar to that of UO_2 rods [7,8]. The CALLISTO programme was in effect, an extension of the PRIM0

programme in that selected rods from that experiment werZ subsequently ramp tested in the Callisto loop of BR2 reactor. The results of this experiment indicated that the behaviour of MOX fuel under transient conditions is equivalent to or even better than that of UO_2 fuel [9].

The FIGARO programme, which is now nearing completion, was designed to evaluate the thermal behaviour of MOX fuel at a bum-up of around 50 GWd/t HM and to compare the fission gas release threshold of MOX and UO_2. Two rods were extracted from assembly NOK - Ml09 which was irradiated in the Beznau-1 reactor to high bum-up and the irradiation continued in the Halden test reactor (with on line instrumentation) under PWR conditions. On-line pressure measurements indicated that the temperature threshold for fission gas release in MOX is close to that of UO_2 and the PIE results showed that fuel conductivity degradation with bum-up follows the same trend in both MOX and UO_2.[10].

The NOK - M 109 programme took advantage of the rod extraction required for the FIGARO programme and an additional 8 rods were extracted at the same time. A PIE programme was then proposed which would extend the available irradiation performance data of MOX fuel. The fuel was irradiated for 5 cycles (approx 50 GWd/t HM) and the PIE carried out focused on fission gas release; this work has recently been completed.

The NOK -M308 (PWR) and GERONIMO (BWR) programmes are also designed to extend the MOX fuel performance database. The M308 assembly was irradiated in Beznau - 1 to a peak pellet bum-up of 58 GWd/t HM while the fuel in the GERONIMO programme which is still being irradiated in Gundremmingen BWR, will reach 65 GWd/t HM when finally discharged. These international programmes have only recently been established and will involve PIE and ramp testing.

In France the LWR MOX feasibility studies relied mainly on results from international test irradiation programmes - see Table 2. Of particular value was the programme supported by the Commission of European Communities (CEC) during 1974 to 1986 which concentrated mainly on PIE and isotopic analysis of MOX fuels. The fuel assemblies tested covered a range of designs and fabrication routes and were irradiated in several PWRs throughout Europe - BR3 in Belgium, Dodewaard in Netherlands, Garigliano in Italy, Lingen in Germany and CNA in France. In addition, some MOX fuel rods were irradiated in the CEA experimental reactor, CAP IV between October 1985 and early 1987. These programmes together with the international PRIM0 programme which ran between 1987 and 1994 provided physics data to verify core design codes for MOX fuel and to develop specific MOX fuel performance models.

TABLE 2. MAIN IRRADIATION TEST PROGRAMMES USED BY FRANCE TO SUPPORT LWR MOX UTILISATION

Time Period	Scope of Work	Description	Purpose
1974 - 1986	Irradiation + PIE	CEE Programme: investigation of MOX fuel performance (10 contracts) Rod Bum-up - 48 GWd/t	Demonstration, Fuel Performance, Modelling
1987 - 1991	Irradiation + PIE	Irradiation of MOX fuel rods in the small CAP PWR under load following conditions Rod Bum-up - 20 GWd/t	Fuel Performance, Modelling
1987 - 1994	International PRIM0 programme	Examination of 15 rods irradiated in BR3 + ramp test Rod Bum-up - 55 GWd/t	Core physics data, Fuel Performance, Modelling

In Germany two sets of test irradiation programmes in support of thermal MOX were carried out. The first, in the 1970's utilised MOX fuel fabricated using a process which resulted in poor homogeneity giving rise to solubility problems. The second set of test irradiations, carried out during the 1980's and early 1990's, concentrated on the irradiation verification of modern MOX fabricated using the OCOM process [11]. Table 3 lists the main irradiations carried out under the first set of test programmes and Table 4 summarises those carried out during the second phase.

TABLE 3. SIEMENS IRRADIATION TEST PROGRAMME FOR GERMAN 'FORMER STANDARD FUEL' IN SUPPORT OF LWR MOX UTILISATION

Reactor	Scope of Work	Description
KWO	Power Transients	14 short test rods Max powers between 260 and 417 W/cm Rod Burn-ups - 9 to 27 GWd/t HM
HFR Petten	Power Transients	10 short rods pre-irradiated in KWO Ramp terminal powers - 480 to 560 W/cm Rods Burn-ups - 9 to 32 GWd/t HM
Halden BWR	Instrumented Irradiation to determine fuel temperature and fuel densification	IFA 4271428

TABLE 4. SIEMENS IRRADIATION TEST PROGRAMME FOR GERMAN 'MODERN' MOX FUEL IN SUPPORT OF LWR UTILISATION

Year	Rod/FA Number	Type of Fuel	Rod Burn-up GWd/t HM	Transient Testing
1980	Segmented rods	AUPuC	23 - 39	HFR Petten
1981	Reactor A/FA 1	OCOM/AUPuC	6 - 42	
1984	Reactor A/FA 2	OCOM	9 - 34	HFR Petten
1986	Reactor A/FA 3	OCOM-30 & -15	8-41	

For the first test of this second phase programme, 15 segmented long rods with 7 short rods in each case were irradiated for up to 4 cycles. The short rods were axially reduced rods modified in length to match the thermal flux field of the HFR pool facility at Petten and thus allow simultaneous power increase of the whole rod. In total 12 short rods with modem MOX fuel have been transient tested to date. MOX fuel manufactured using both the OCOM and AUPuC processes have also been included in this programme and thus a comparison between PuO_2 powder and liquid Pu nitrate from different reprocessing plants has been carried out. This has shown that despite different powder properties, the fact that both manufacturing processes were optimised with respect to Pu homogeneity, resulted in comparable behaviour. In addition to the segmented rods, demonstration fuel assemblies (FA 1, 2, 3) were manufactured and extensively characterised before irradiation. Included in one of these assemblies was some experimental fuel which had a reduced Pu content of 15% in the agglomerates; this was irradiated to a local burn-up of 45 GWd/t and was designed to study the influence of Pu homogeneity on irradiation behaviour [12].

Such test irradiations provided the data necessary to compare MOX fuel behaviour with that of UO_2, to develop specific MOX fuel performance models and to verify core design codes. Work continues in this area today with a number of international collaborative programmes as well as national studies being carried out in the Halden test reactor [13]. Such programmes are of particular importance to the UK (i.e. BNFL) who, as a late entrant to the thermal MOX arena, has to rapidly demonstrate the viability of its fuel under all operating conditions and as such is involved in several major national and international test programmes.

4. COMMERCIAL IRRADIATIONS

Commercial irradiations of MOX fuel began in earnest in the mid 1980's and today 12 out of 19 plants in Germany are licensed for MOX usage, while in France, 20 reactors are authorised to load MOX; Switzerland and Belgium have between them 5 PWRs licensed for MOX usage.

Table 5 summarises the commercial irradiations of MOX fuel which have been carried out in European reactors since the 1960's.

TABLE 5 . EUROPEAN NATIONAL EXPERIENCE WITH MOX FUEL IN COMMERCIAL LWRs SINCE 1960's

Reactor (Type)	Initial Year of Loading	Total Number of Assemblies*	Total Number of Rods	Max Assembly Burn-up Achieved (GWd/t HM) **
GERMANY				
BWR reactors	1966	177 + (1)	3389	21 (34)
PWR reactors	1972	502 (0)	91248	45 (14)
(French manufactured MOX)		124 (0)	28768	
ITALY				
Garigliano (BWR)	1968	24 (46)	2072	approx 25
Trino (PWR)	1975	8 (0)		(approx 3 5)
FRANCE				
17 x900MWe (PWR)	1987	1032	272448	51
NETHERLANDS				
Dodewaard	1971	7 (0)		(47)
(BWR)	1988	5 (0)	25 seg	(58)
SWEDEN				
OKG- 1, Oskarshamm (BWR)	1974	3	51	(19)
SWITZERLAND				
Beznau -1,2 and Goesgen (PWR)	1978	168 (0)	>21500	51
BELGIUM				
Doe1 3 & Tihange (PWR)	1995	72	19008	45

* Figure in parenthesis denotes number of MOX island assemblies loaded
** Figure in parenthesis denotes peak pellet burn-up
\+ This figure includes some MOX island assemblies

In Germany a total of 768 MOX fuel assemblies, manufactured by Siemens, have been irradiated in LWRs at burn-ups up to 45 GWd/t HM in the years since 1966. In France, 1032 assemblies have been loaded into EdF stations since 1987 with similar maximum burn-ups; 476 assemblies have successfully completed 3 irradiation cycles. In Belgium 64 assemblies have been loaded into 2 PWRs and 16 of these have been irradiated for 3 cycles. In Switzerland MOX fuel was first loaded into Beznau-1 in 1978 and since then a total of 168 assemblies have been irradiated in the Beznau-1 and 2 and Goesgen reactors. The assembly burn-ups are again around 45 GWd/t HM but, as in the case of Germany and France, individual assemblies have reached levels of around 51 GWd/t HM.

5. PIE PROGRAMMES AND FUEL PERFORMANCE

Post Irradiation Examination programmes including poolside inspection, non-destructive and destructive hot cell examinations, have been incorporated into the MOX utilisation studies from the very beginning and the data base is now fairly extensive, although still well behind that of UO_2 in quantitative terms.

In Germany approximately 200 fuel assemblies with modern MOX fuel have undergone under-water poolside inspection as part of the normal fuel assembly and control rod inspection programmes which are carried out during reactor reloads. In addition 170 MOX fuel rods have been withdrawn from various assemblies and oxide thickness, length and diameter measurements carried out prior to re-insertion and continued irradiation.

Table 6 summarises the major hot cell campaigns which were carried out to verify the irradiation behaviour of MOX fuel in Germany. The first phase concentrated on the irradiation behaviour of the original inhomogeneous fuel. The second phase delivered base irradiation data on the improved fuel; this included data on fuel which had experienced different power histories and fuel which had undergone transient tests. The third phase has recently started with MOX fuel irradiated at high power ratings to burn-ups in excess of 60 GWd/t HM.

TABLE 6. PIE PROGRAMMES ON GERMAN MOX FUEL MANUFACTURED BY SIEMENS

Time Period	Reactor/No of Rods	Objective	Rod Burn-up MWd/t HM	Result/Comment
1976 - 1978	A / 12	Basic irradiation data for rod design & neutron physics (former standard fuel)	8 - 37	Rod behaviour comparable to UO_2 in spite of 100% Pu agglomerates
1982 - 1993	A / 12 + 15 segmented	Basic irradiation data for modern fuel	6 - 42	Rod behaviour comparable to UO_2, no significant influence of Pu inhomogeneity
1987 - 1989	B / 4	Data base extension for different power histories at increased burn-up	35 - 41	Fission gas release will be sensitive to the power history of the later cycles
1999 - 2003	C	Verification of high rated MOX fuel to high burn-up	1 to 4 cycles > 60	Rods under irradiation

The results from these PIE programmes indicate that heterogeneous Pu distribution (and hence burn-up and fission product distributions) in MOX fuel does not adversely affect the

irradiation performance compared with UOZ fuel. This is demonstrated for fission gas release, fuel density and transient behaviour in that:

- Most of the MOX fuel fission gas release data match that of UO_2; the higher values can be attributed to variations in cladding/fuel gap or differences in irradiation history.
- MOX fuel density measurements lie in the middle of the UO_2 data range.
- Under transient conditions all MOX rods tested have remained intact even under conditions above the UO_2 defect threshold.

In France a rigorous MOX fuel qualification programme has been underway since 1974. Poolside examinations performed on irradiated assemblies after 1, 2, 3 and 4 cycles have not revealed any abnormal mechanical behaviour and have confirmed the overall good condition of the MOX assemblies. No differences in operational characteristics (bow, growth etc.) have been observed compared with UO_2 fuel.

45 fuel rods at burn-ups up to 52 GWd/t HM and covering a range of Pu contents have been examined in hot cells. The PIE results indicate that MOX rods behave similarly to UO_2 rods in terms of waterside corrosion and rod dimensional effects; in addition, MOX assemblies irradiated under load following conditions behave similarly to reference rods operated under base load conditions.

Experience in France with respect to fission gas release and performance under transient conditions is very similar to that observed in Germany

- Under the operating conditions applied, fission gas release in MOX fuel is slightly higher than that in UO_2 fuel. This is due mainly to higher heat ratings experienced in the MOX rods, particularly at high burn-up.
- The behaviour of MOX fuel under transient conditions has been shown to be equivalent to or even better than that of UO_2 fuel. End of Life ramp levels up to 480 W/cm have been reached without clad failure, indicating that MOX fuel has an inherent resistance to Pellet/Clad Interaction (PCI) which allows greater flexibility in terms of core management.

The R&D programmes organised by BELGONUCLEAIRE have included PIE of fuel irradiated under normal and transient conditions and at burn-ups close to 60 GWd/t HM. The main findings of these programmes are supported by those carried out independently in France and Germany and can be summarised as follows:

- Thermal conductivity decreases with burn-up in the same manner as in UO_2 fuel.
- Any Pellet/Clad Mechanical Interaction observed under steady state conditions is similar to that observed in UO_2 fuel. The power failure threshold in transient conditions is higher than in UO_2. Overpower transients up to 500 W/cm have been achieved and sustained without failure in fuel at burn-up close to 60 GWd/t HM.
- In-reactor fuel central temperature and segment inner pressure measurements confirm that temperature threshold for fission gas release is close to 1200°C when approaching 50 GWd/t HM, i.e. a value similar to UO_2.
- Helium production is significantly higher than in UO_2 fuel, due mainly to production and a-decay of Cm^{242}. This leads to an increase in rod internal pressure, which is more easily detected in unpressurised, highly rated rods.
- Radial distributions of actinides and fission products determined by EPMA and SIMS techniques have provided numerous data on thermal flux depression, and local Pu content versus burn-up, thus allowing accurate calculation of radial power distributions during life.

- The microstructure of fuel has been extensively investigated. The fragmentation pattern is similar to that observed in UO_2 fuel. MIMAS fuel, with a pre-irradiation structure consisting of a fine dispersion of Pu, is characterised after irradiation at low temperature (<1000°C) by agglomerates of fine porosity resulting from fission gas associated with high burn-up in Pu rich-zones. These zones represent a small fraction only of burn-up

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- Any increase in FGR (Fission Gas Releas) observed in MOX fuel can be attributed to differences in power histories as well as differences in thermal performance as indicated above.

6. CONCLUSIONS

The fuel rod and assembly design for MOX fuel has been universally based on UO_2 design with only two minor modifications relating to the neutronic and thermal mechanical properties of the MOX fuel itself. It is generally the case that if a modification is applied to UO_2 then the same change will be incorporated in MOX some time later.

National and collaborative test irradiations on LWR MOX fuel have been carried out in Europe over a period of 35 years. These programmes, which are still ongoing, are providing the data necessary to compare MOX fuel behaviour with that of UO_2, and to develop specific MOX fuel performance models and verify core design codes. A wide range of variables has been investigated in these test irradiations e.g. fabrication processes, cladding materials, rod geometries, operating conditions.

In the same period, over 2000 MOX fuel assemblies have been irradiated in European commercial PWRs and BWRs with failure statistics indicating that the reliability of MOX fuel is at least as good as that of UO_2.

The PIE prog-rammes accompanying both the test programmes and commercial irradiations all indicate that the overall performance of the MOX fuel has proved to be as good as that of equivalent UO_2 fuel but with the added benefit of improved resistance to PCI.

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MOX FUEL PERFORMANCE IN' FRENCH PWR REACTORS: RECENT RESULTS AND IMPROVEMENT PROGRAMME

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Abstract

By the end of 1998, 992 assemblies had been irradiated since the first loading of MOX fuel assemblies in French PWRs in 1987. 20 reactors are now authorized to use MOX fuel. Operating MOX fuel in PWRs up to an average discharge burnup of 37 GWd/t did not cause any particular problem and MOX fuel reliability, based on increasing experience feedback is as good as the UO_2 one.

In parallel, a considerable experience on MOX fuel performance has been accumulated by post-irradiation examination of rods irradiated up to more than 50 GWd/t (4 cycles) in PWRs (surveillance programme) and in experimental irradiation programmes.
Ongoing R&D at the CEA, in collaboration with the EDF and FRAMATOME, is directed towards gaining a better understanding of MOX fuel behaviour in normal and off-normal conditions in order to improve and validate the models used to justify fuel behaviour prediction calculations and to support the licensing of extended burnup.
Recent results in PWR experience feedback as well as R&D programmes will be discussed.

As MOX fuel has now reached a sufficient maturity, the next step is to achieve parity between UO_2 and MOX fuel. This involves a single management strategy for UO_2 and MOX assemblies with an annual quarter core reload type.

The goal is to reach a maximum assembly discharge burnup of 50 GWd/t for MOX and UO_2 fuel assemblies. The new FRAGEMA's AFA 3G structure will increase margins in fuel rod design with regard to the end-of-life internal pressure criteria.

The objective of obtaining a parity between UO_2 and MOX must be achieved even beyond the initial goal of 50 GWd/t for maximum discharge burnup. This is why another programme was launched with the purpose of substantially improving MOX fuel assembly discharge burnup, i. e. up to 70 GWd/t, as it has been the case for advanced UO_2 fuel previously.

The R&D programme mainly focuses on increasin@ission gas retention, which is the main obstacle to reaching very high burnup. The improvement of fuel assembly design will also be integrated to increase margins.
The main steps of the programme are the following :

- optimization of pellet microstructures and validation in experimental reactors,
- build-up of experience feedback at elevated burnup in commercial reactors, both for current and experimental products,
- adaptation and qualification of the design models and tools.

The product resulting from this new development could be qualified around 20 10

I. INTRODUCTION

Since the first loading of MOX fuel in a French 900 MWe PWR in 1987, the number of EDF units recycling plutonium has increased progressively (9 in 1996, 13 in 1997, 17 in 1998 out of a total of 20 authorized). In agreement with the EDF's recycling policy, this number will increase up to 28 in order to maintain an equal flow between reprocessing and recycling plutonium. At the present time the production capacity of the MELOX plant, which has reached 100 tHM/year is adequate for MOX assembly production of the 20 units concerned.

992 MOX assemblies were irradiated in French PWRs between 1987 and the end of 1998, 476 having achieved three irradiation cycles, and a few assemblies were authorized to be loaded for a fourth and even a fifth cycle for an experimental purpose.

The « hybrid » fuel managemant scheme with 28 UO_2 fuel assemblies (3.7% enriched) irradiated for four annual cycles and 16 MOX assemblies for three annual cycles has been used for all « moxified » reactors since 1994, with load follow conditions authorized for all the plants since 1995. Recently the maximum average plutonium content of MOX fuel assemblies was increased up to 7.08 %.

The product AFA 2G MOX, designed for an assembly burnup of 43-45 GWd/tM, integrated the improvement in the current UO_2 product with a modified fuel rod design (mainly lower initial helium pressure and a slightly increased free volume).

Since the use of MOX fuel in French PARs has now reached its industrial maturity, the main challenge remains to follow UO_2 performances in terms of discharge burnup and maneuverability (the authorization was given recently to operate UO_2 fuel up to 52 GWd/tM). The parity between UO_2 and MOX fuel in terms of fuel management and burnup is an objective to be achieved in 2004-2005.

For this reason the understanding of MOX fuel behaviour at high burnup must be continously improved by :

- using a surveillance programme of standard fuel rods irradiated in PWR plants and associated hot-cell examination,
- using ongoing R& D programmes based on analytical out-of-pile or in experimental reactor programmes, under nominal and transient conditions.

II. RECENT RESULTS IN MOX FUEL PERFORMANCE

The highlights of the important experience feedback of nearly 1000 MOX assemblies irradiated in French 900 MWe reactors are :

- the average discharge burnup of MOX assemblies having achieved three irradiation cycles is 37 GWd/tM (Figure l),

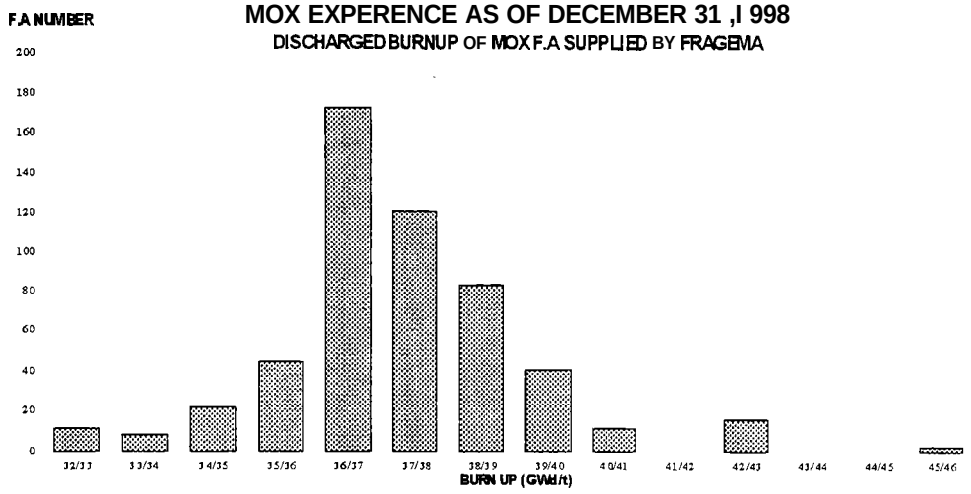

FIG 1 MOX fuel assembly irradiation experience in France up to the end of 1998

- the average assembly burnup obtained after a fourth irradiation cycle operated on 6 assemblies for experimental purposes is about 46 GWd/t,
- one MOX assembly was reloaded in Gravelines 4 in mid 98 to achieve a fifth cycle ; the expected maximal burnup is 54 GWd/tM.

The reliability of MOX assemblies remains as good as for UO_2 fuel (evaluated now at 9 10^{-6} leaking rod per cycle) since only two clad failures, due to debris, occurred on MOX rods without any consequence on the primary circuit activity level. The release rates of gaseous fission products were similar to those observed with defective UO_2 fuel /1/.

II. 1 Surveillance programmes and hot-cell examinations

Up to now, more than 50 fuel rods with different burnups, different plutonium contents and different fabrication processes have been examined in hot cells.

The examination of three-cycle rods irradiated in St Laurent Bl and St Laurent B2 was reported several years ago /2/ showing a higher fractionnal fission gas release as the main difference with UO_2 fuel. This behaviour was mainly explained by the linear heat rates of the MOX rods which are higher than in UO_2 rods at the same burnup and to a lesser extent by specific physical properties of MOX fuel (thermal conductivity, microstructure). Afterwards, a later paper /3/ reported the first hot-cell examination results on the Gravelines 4 four-cycle rods (52 GWd/tM average rod burnup). The results did not show any enhancement of the fractionnal gas release between the third and the fourth cycle. This was attributed to the low heat rate the rods were subjected to during the last irradiation cycle. Comparison with UO_2 rods in terms of fuel rod growth, cladding waterside corrosion, fuel density and fission gas release were also drawn as a function of burnup, without showing any particularity in four-cycle MOX fuel behaviour.

More recently, additional examinations were performed on fuel rods also irradiated for a fourth cycle in Gravelines 4 but at a higher heat rate (170W/cm as opposed to 130W/cm). The measured fractional gas releases were similar to those observed on fuel rods with a lower heat rate at the end of irradiation. All the results avaible up to now on UO_2 and MOX fuel fission gas release are gathered on Figure 2. The conclusion is that no fission gas release enhancement occurs due solely to the burnup effect, the two sets of 4-cycle rods have been irradiated under the fission gas release threshold.

Other destructive examinations did not reveal any other differences with the previous rods.

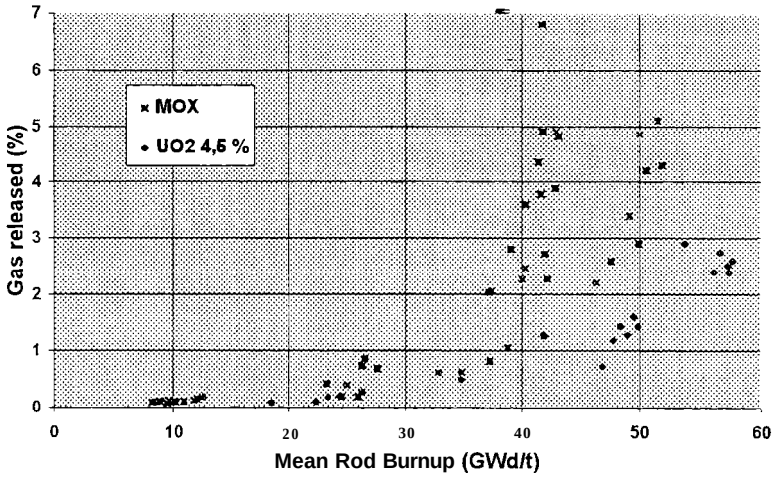

FIG 2 MOX and UO_2 fission gas release versus burnup

By the beginning of 2000 the first fuel rods having experienced a fifth irradiation cycle will be shipped to hot-cells in order to be examined (maximum rod burnup of about 60 GWd/tM).

II.2 Analytical programmes in nominal and transient conditions

Within the framework of the R&D conducted in common programmes by the CEA, FRAMATOME and EDF, the need for experimental tests devoted to an improved understanding of MOX fuel behaviour, in both normal and off-normal conditions has been stressed, mainly concerning thermal properties, fission gas release and mechanical properties.

- Thermal behaviour of MOX fuel at beginning of life has been studied through out-of-pile thermal conductivity measurements and in-pile experiments involving the measurement of centre-line temperature of the fuel rod. The recent out-of-pile data /4/ are consistent with the GRIMOX 2 irradiation results /5/ and confirm a slightly lower thermal conductivity for MOX fuel,

- Experience feedback on fission gas release in nominal conditions is constantly enlarged from surveillance programmes results and, in addition to this, analytical experiments are performed in order to characterize FGR in transient conditions /5/. The existence of a thermal threshold beyond which release is accelerated is confirmed for MOX fuel as it has been for UO_2 but with a higher level of release. Studies are now underway to examine thoroughly the links between the heterogeneity of the MOX microstructure (size and Pu content of $(U,Pu)O_2$ particles, microstructure of the UO_2 matrix) and the mechanisms and kinetics of fission gas release in normal and transient conditions. An important test programme is performed consisting in local and global thermal annealings of several fuel samples with different manufacturing processes, different burnups and plutonium contents in order to improve and validate the understanding and modelling of fission gas release mechanisms,

- Though it has been proved by ramp tests that MOX fuel behaves particularly well from the pellet-cladding interaction, and therefore from the power plant maneuverability standpoint /3/, studies are still in progress to understand and characterize this effect attributed to a higher MOX pellet creep in comparison to the UO_2 pellet during power transients.
  For that purpose two kinds of tests are performed : ongoing mechanical property measurements on fresh fuel and the in-pile programme DEFORMOX which is devoted to intrinsic UO_2 and MOX thermal-mechanical behaviour studies on fresh and irradiated fuels /5/. Recent results on irradiated fuel confirm the results of the first DEFORMOX tests on fresh fuel (Figure 3) i. e. a more homogeneous deformation of the MOX pellet under PCI conditions than for UO_2. Post irradiation examinations are still in progress.

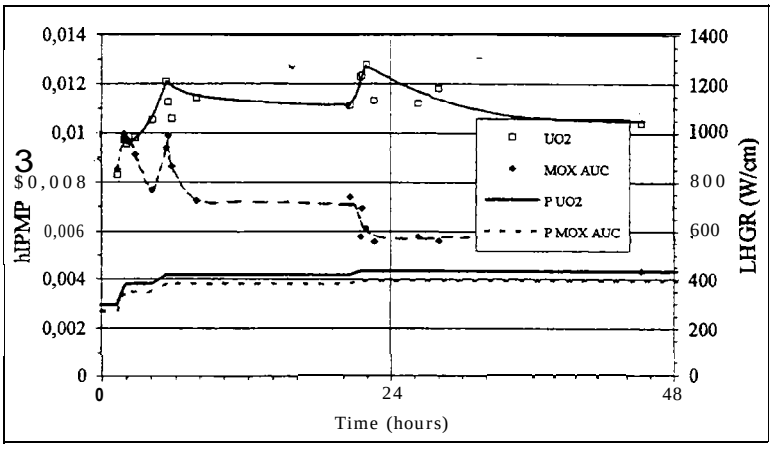

FIG 3 Time and power dependence between inter-pellet and mid-pellet diameters, for MOX and UO_2

III. MOX DEVELOPMENT PROGRAMMES

III. 1 Parity MOX

The current hybrid management with quarter core UO_2 and third core MOX reloads leads to an average discharge burnup of 37 GWd/tM for the MOX fuel and 43 GWd/tM for the UO_2 fuel, compared with an average 45 GWd/tM for UO_2 quarter core reload management,

Since the next goal is to achieve parity between MOX and UO_2 fuel performance by 2004-2005, the « MOX parity » project has been launched /6/. The reference fuel management is of an annual quarter reload type, in which each reload is made up of 28 UO_2 assemblies (3.7%) and 12 MOX assemblies (equivalent 3.7%) with a maximum assembly discharge burnup of 50 GWd/tM for UO_2 and for MOX.

The preparatory phase, which started in 1997, is now completed. This phase has made it possible to analyse the effects of this new management on :

- the operating conditions,
- the NSSS systems,
- the worst-case accident events,
- the design of the MOX assembly,
- reactivity injection accident (RIA)

The main accident events under analysis are :

- reactivity control in the various cold and hot shutdown states during the spurious dilution and RCCA ejection accident,
- cooldown accidents (large and small secondary breaks),
- loss of coolant accident.

The locating of four additional shutdown RCCAs and the increase in boron concentration, particularly in the refuelling water storage tank, allow the safety criteria to be met.

The reference fuel product for this management strategy is AFA 3G MOX. The new AFA 3G structure allows the design of a fuel rod with increased margin to end-of-life internal pressure. The design models are validated for the envisaged discharge burnup and are based on the results of the MOX surveillance programmes (4-cycle irradiation of a MOX assembly) and on analytical R&D

programmes. These include the HALDEN and FIGARO programme results for heat transfer and elevated-bumup fission gas release data and the three-partner CEA-EDF-FRAMATOME programmes DENSIMOX and GONCORMOX dealing with the behaviour of the MOX pellet (densification and gaseous swelling).

The detailed design phase for a gradual introduction of this new UO_2-MOX fuel management in PWR 900 MWe nuclear power plants began early in 1999.

III2 High burnup MOX fuel assembly development

Beyond the objective of achieving parity between MOX and UO_2 fuel at a burnup ratio of 50 GWd/tM in 2004-2005, the equality must be established on a long-ten-n basis and the management of MOX must be developed in the same way as that of UO_2.

The main objective of the high burnup MOX fuel assembly development programme being run jointly in the three-partner CEA-EDF-FRA framework and in the common CEA-COGEMA R&D framework is to prove, over the next 10 years, that MOX fuel is capable of achieving burnup ratios of 70 GWd/tM with an appropriate fuel and assembly design /7/.

This ambitious objective is based on a large-scale R&D programme involving :

- An in-depth knowledge of how MOX behaves at high burnup joint to a good understanding of how it differs from UO_2. The thermal behaviour of oxide and its influence on fission gas release, among the most significant differences, has been successfully modelled up to about 50 GWd/tM and must be extrapolated up to 70 GWd/tM from an experimental irradiation programme. High burnup MOX fuel behaviour in accident conditions must also be investigated thoroughly.

- Improvements of the present product, in order to meet the required objective ; modifications are under consideration on both oxide $(U,Pu)O_2$ and fuel rod and assembly design.
  Concerning oxide performance improvement, the main goal is the reduction of fission gas release, without any unfavourable change with regard to overall behaviour, which is in principle satisfactory for MOX at high burnup (pellet cladding interaction, physical/chemical and thermal properties).
  Design assembly improvement will be based on the new Alliance™ product already aimed at a burnup of 70 GWd/tM for UO_2. The use of the M5 alloy as cladding material and the increase in the volume of plenum will have a favourable effect on allowable fission gas release.

- Development of manufacturing processes adapted to the new product and the study of their industrial feasibility consistently with the objective of 70 GWd/tM in 20 10.

Two main steps have been identified :

- Improving the current product by 2005 and achieving a significant fission gas release improvement. A specific irradiation, involving an optimized MlMAS fuel, will be performed in a power reactor in order to give an assessment of the improvement in 2005,

- Developing MOX 70 GWd/tM, with first a phase of optimization of pellet microstructures, then analytical experiments in experimental reactors, followed by a demonstration irradiation in a commercial reactor (2005-2010). At the same time, an analytical programme for modelling the behaviour of the new product will be carried out, in order to prepare its licensing.

IV. CONCLUSION

With an increasing experience feedback, the use of MOX fuel in French 900 MWe PWRs has now reached industrial maturity. Besides the constant accumulation of surveillance programme results, an extensive R&D is ongoing to understand, model and validate high burnup fuel behaviour in nominal and transient conditions, especially regarding fission gas release which constitutes the main obstacle in reaching high burnups.

Beyond the objective of achieving parity between UO_2 and MOX in 2004 at a burnup of 50 GWd/tM an important programme involving consequent R&D begins with the first objective of current product improvement in 2005 and a second goal of developing a new product qualified for 70 GWd/tM around 20 10.

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HIGHLIGHTS ON R&D WORK RELATED TO THE ACHIEVEMENT OF HIGH BURNUP WITH MOX FUEL IN COMMERCIAL REACTORS

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Abstract

Part of the R&D work made at BELGONUCLEAIRE in the field of high burnup achievement with MOX fuel in commercial LWRs is made through lnternational Programmes. Special attention is given to the evolution with burnup of fuel neutronic characteristics and of in-reactor rod thermal-mechanical behaviour

Pu burning in MOX is characterized essentially by a drop of Pu239 content. The other Pu isotopes have an almost unchanged concentration, due to internal breeding. The reactivity drop of MOX versus burnup is consequently much less pronounced than in UO_2 fuel. Concentration of minor actinides Am and Cm becomes significant with burnup increase. These nuclides start to play a role on total reactivity and in the helium production.

The thermal-mechanical behaviour of MOX fuel rod is very similar to that of UO_2. Some specificities are noticed. The better PCI resistance recognized to MOX fuel has recently been confirmed. Three PWR MOX segments pm-irradiated up to 58 GWd/tM were ramped at 100 W/cm.min respectively to 430-450-500 W/cm followed by a hold time of 24 hours. No segment failed.

MOX and UO_2 fuels have different reactivities and operate thus at different powers. Moreover, radial distribution of power in MOX pellet is less depressed at high burnup than in UO_2, leading to higher fuel central temperature for a same rating. The thermal conductivity of MOX fuel decreases with Pu content, typically 4 % for 10 % Pu. The combination of these three elements (power level, power profile, and conductivity) lead to larger FGR at high burnup compared to UO_2.

Helium production remains low compared to fission gas production (ratio < 0.2). As faster diffusing element, the helium fractional release is much higher than that of fission gas, leading to rod pressure increase comparable to the one resulting from fission gas.

1. INTRODUCTION

Plutonium recycling in LWRs has reached today its industrial maturity with MOX fuels. It is fabricated and loaded in reactor in large quantities [1]. It is recognized that the MOX fuel has an in-reactor behaviour almost identical to UO_2 [2]. This similarity is a consequence of the close properties of UO_2 and PuO_2 materials over a broad range of properties (crystallography, physics, neutronics,.), allowing easy replacement of U atoms of UO_2 lattice by Pu atoms, without deep modifications of UO_2 characteristics, and to develop a MOX industry based on UO_2 options regarding fuel fabrication and assembly/rod design, and to irradiate this MOX fuel in reactors mainly conceived to burn UO_2.

However, significant differences exist between the two fuel types. These differences have a neutronic character (mainly the larger fission and capture cross sections and the reduction of the number of delayed neutrons for Pu) or a physical character (lower thermal conductivity and lower creep strength of MOX).

As for UO_2 fuel, a demand for high burnup achievement with MOX fuel exists to reduce the electricity production costs. This demand is even greater for MOX due to a fuel cycle generally estimated to be more expensive than for UO_2. It is also economically meaningful to increase MOX burnup as a high initial Pu content is financially not very penalizing compared to UO_2 fuel for which the enrichment cost markedly increases with U235 content.

The present paper aims to highlight some topics on neutronic and in-reactor thermal-mechanical behaviour of MOX fuel needing appropriate consideration for high burnup achievement. These topics are illustrated by experimental results often obtained in the framework of R&D programmes organized by BELGONUCLEAJRE. The programmes are briefly reviewed.

2. BELGONUCLEAIRE R&D PROGRAMMES

As MOX fuel manufacturer, BELGONUCLEAIRE is traditionally concerned by the validation, the performance and the behaviour of MOX fuel in various irradiation conditions.

The major interest in such field is focused on a continuous updating and completeness of MOX fuel data bases through the promotion of R&D work mainly devoted to pending validation and licensing questions of plutonium recycling in commercial reactors.

Since more than 25 years, such work is proposed through a set of international programmes, the unal nf which being an attractive share of the budget and of the resulting data between several organizations directly concerned by MOX fuel and its various aspects within all phases of the fuel cycle.

Such organizations are mainly fuel Designers and Manufacturers, Research Laboratories, Utilities as well as Organizations in charge of wastes, fuel handling and storage activities.

These international programmes which are initiated, negotiated and managed by BELGONUCLEAIRE can be divided in two major classes (Table 1) :

MOX Fuel Core Physics Data	MOX Fuel Behaviour
VIP-PWR	PRIMO (PWR)
VIP-BWR	CALLISTO (PWR)
VIPO	DOMO (BWR)
ARIANE	FIGARO (PWR)
VIPEX	NOK-Ml09 (PWR)
REBUS	NOK-M308 (PWR)
VIPOX*	GERONIMO (BWR)
	HELISARA *

* under preparation

Table 1. MOX International Programmes

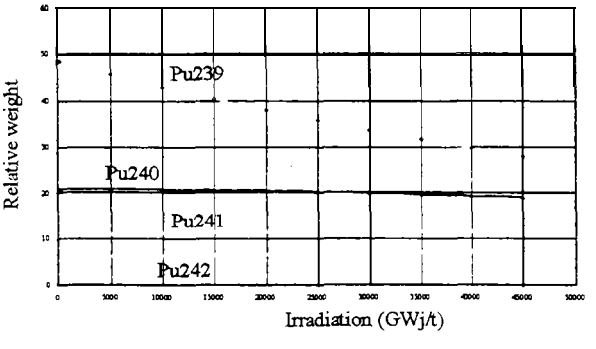

Fig. 1. Evolution of Pu content (8.9% initial) in MOX versus burnup

The first set of programmes focuses on neutronic **investigations** in direct relation to MOX fuel performance, *with two main topics.*

The ***first*** *topic,* generally organized jointly by BELGONUCLEAIRE and SCK(CEN is mainly devoted to the determination of reactor physics parameters using the VENUS critical facility located in Mol (Belgium), and simulating recent designs of PWR or BWR assemblies, and mock-ups containing more than 12 % Pu average.

- **The VIP Programme**

From 1990 to 1992, the Pu recycling in LWR's was investigated in the VENUS critical facility at SCK(CEN. The programmes called VIP (VENUS International Programme), used fuel with high Pu and Gd content. The aim of the VIP programmes was the validation of reactor codes with respect to MOX-fuel for both PWR's and BWR's. They were focused on the criticality and fission rate distribution calculations inside the MOX fuel assembly, especially at the UO_2 interface.

These programmes were divided in two parts :

VIP-PWR

mock-up 1 : All MOX 17X17 subassembly
mock-up 2 : MOX-Gd 17X17 subassembly.

VIP-BWR

mock-up 1 : All U02 : 8X8 subassembly
mock-up 2 : All MOX : 8X8 subassembly
mock-up 3 : Island MOX : 8X8 subassembly

- **The VIP0 Programme**

Since 1993, the void coefficient in LWR's has been investigated. Calculations have shown the possibility that at high plutonium contents the void coefficient could become positive. The VIPO Programme (Void Coefficient Measurement in Plutonium Mixed Oxide Lattice) was devoted to the measurements of the perturbation caused by void bubbles in a LWR reactor using high Pu enrichment (i.e. from 10 % to 15 %) and the validation of the related computer codes. A special experimental device has been developed and constructed in order to simulate a void in the reactor's core, the so-called void box.

- **The VIPEX Programme**

As a complement to the VIP PWR Programme, the VIPEX Programme provides basic MOX physics data derived from specific investigations based on critical MOX mock-up measurements in VENUS.

The parameters investigated are the effect of Am241, the Beta-effective, the flux tilt inside a MOX comer rod, the effect of simulated water density (by introduction and removal of Al microrods in the MOX lattice), the control rods worth in MOX fuel assembly and, as supplementary investigation, the detector (fission chamber) response in MOX fuel assemblies.

The second ***topic*** of these MOX neutronic evaluations includes fuel irradiation studies at high burnup with experimental determination of actinides and fission products inventory.

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The ARIANE Programme investigates the irradiated MOX fuel source term and will serve to validate licensing computer code of the ORIGEN type by means of an extensive programme of radiochemistry.

Attention is focused on the accuracy in determining actinides, minor actinides and fission products contents through analyzes in three laboratories (SCK(CEN, TUI and PSI) on fuels unloaded after extended irradiation in commercial reactors, such as BEZNAU-1 (MOX samples), GÖSGEN (UO_2 samples) and DODEWAARD (MOX and UO_2 samples).

The highly fissile minor actinides (Am242m, Cm243, Cm245) are investigated as well as strong alpha emitters (Pu238, Cm242). Fission products investigations focus on neutron absorbing (Sm149, Sm151, ..) or long-lived (1129, Tc99,...) nuclides. More that 20 elements and 50 nuclide contents are determined in most of the selected fuel samples.

The second set of programmes concerns complete investigations of MOX thermal-mechanical fuel behaviour under normal conditions obtained through large scale irradiations in various reactors like BR3, DODEWAARD, BEZNAU-1 and GUNDREMMINGEN and under off-normal conditions which are simulated in BR2, OSIRIS, HALDEN and HFR testing reactors.

Such prograrnmes focus on the determination of MOX fuel rods characteristics for what concerns fission gas release and inner pressure, fuel central temperature, fuel microstructure, rod integrity and pellet-cladding interaction, measured for a wide range of irradiation conditions.

. PRIM0 and DOMO

These programmes, completed in 1994 and 1996 respectively, contributed to provide base information on fission gas release and microstructure at extended burnups in PWR and BWR respectively, and to verify that rod power failure threshold is similar or better than for UO_2 fuel. Detailed results of both programmes were published elsewhere [3,4].

. **FIGARO**

The objective of that programme, now completed, was to evaluate the thermal behaviour of MOX fuel at burnup of about 50 GWd/tM and to determine whether fission gas release threshold was different or not from UO_2. The results show essentially that the fuel conductivity degradation with burnup follows the same rule as for UO_2. About fission gas release, on-line pressure measurements indicate that the temperature threshold for gas release is close to that of UO_2 [5]. These items are detailed in § 4.1 and 5 4.4.

. NOK-Ml09

Taking the opportunity of the extraction of the 2 FIGARO rods from a BEZNAU-1 MOX assembly Ml09 irradiated during five cycles, 8 additional MOX rods were extracted as well. The PIE programme, focusing mainly on fission gas release, is now completed.

Results allow to study MIMAS MOX behaviour at high burnup, considering various power histories and fuel fabrication parameters.

. NOK-M308 (PWR) and GERONIMO (BWR)

These two programmes aim at extending the MOX performance database at still increasing burnup (58 GWd/tM peak pellet for NOK-M308, 65 GWd/tM for GERONIMO). Post irradiation examinations and ramp testing are performed on MIMAS fuel segments.

Another objective for the NOK-M308 Programme is a comparison with an alternative MOX fabrication process. These alternative MOX segments have been fabricated by PSI from the gelation process.

The NOK-M308 Programme started in 1997 with the extraction of segmented rods from the mother assembly irradiated in BEZNAU-1. The PIE programme is underway. The ramp programme has been recently completed, with no cladding failure of pelletized MOX fuel rods tested up to 500 W/cm after a power ramp at 100 W/cm min.

The GERONIMO Programme, just officially started, focuses on irradiation of full-length rods and segmented rods in GUNDREMMINGEN BWR. Extractions are foreseen in 1999 and 200 1,

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Release of helium produced during irradiation significantly contributes to rod inner pressure increase. Conservative prediction of such increase includes uncertainty margins which may penalize rod operation. Uncertainties result from source term calculation, kinetics of release and helium solubility in fuel. The programme aims to perform experimental work focusing on these questions.

3. NEUTRONICS AT HIGH BURN-UP

3.1. Actinides inventory

0 Initial Pu content

Initial fissile Pu content in MOX fuel is progressively increased to achieve high burnup objective. The use of Pu from highly burned UO_2 leads also to an additional increase of the Pu content in MOX to compensate for the reduction of the initial Pu_{fiss}/Pu ratio. Typical initial Pufiss/Pu ratios range from 70 % when using Pu from low burned UO_2 (- 33 GWd/tM, 3.25 % U235) to 65 % for highly burned UO_2 (- 53 GWd/tM, 4.4 % U235). This ratio decreases very slowly for higher burnup.

Maximum achievable burnup is very depending on thermal-mechanical rod performance and maximum authorized Pu content. With a 8 % Pu content, an assembly average discharge burnup of about 45 GWd/tM can be achieved in PWR (this is the present situation of the Belgian reactors loaded with MOX). To reach 60 GWd/tM an initial 8.8 % Pu content (with 0.2 % U235) is needed. This burnup is reached also with UO_2 fuel having an initial U235 enrichment of 4.5 %.

The non linearity of initial Pu content versus burnup allows us to contemplate burnups as high as 70-80 GWd/tM with initial Pu content around – 10 % Pu. This content is well below the (conservative) 12 % upper limit beyond which the reactivity coefficient becomes positive under emptying conditions,

- Evolution with burnup

Pu burning in MOX fuel continues to reduce the fissile Pu content to a ratio of 50 % at 50 GWd/tM, starting from 70 %. At the same time, the Pu240 content increases in the isotopic composition from about 24 % to 35 %.

The evolution of the compositions with burnup is due to the marked drop of Pu239 (Figure 1) consumed by fission and capture. The concentration of isotopes of mass 240 and above is remarkably stable with burnup. This stability creates a drop of reactivity with burnup markedly less pronounced than in UO_2 fuel (Figure 2).

The fissile minor actinides Am242m, Cm243 and Cm245 originate from the evolution of actinides towards isotopes of higher mass. At high burnup they contribute significantly to the reactivity. Good experimental data and well calibrated neutronic codes on actinide chain are therefore imperative when achieving high burnup with MOX fuel.

A second consequence of the internal breeding is the rather slow drop of total Pu content with burnup. A typical relative decrease of about 30 % only is noticed, resulting essentially from the reduction of the Pu239 content.

3.2. Radial distribution

The presence of Pu in the pellet increases the thermal flux depression towards the centre, due to large absorption of thermal neutrons on fissile and fertile Pu isotopes. The radial power profile is therefore, at least at beginning of life (BOL), more depressed than in UO_2.

With burnup increase, the consumption of the initial Pu239 is therefore more pronounced at pellet periphery, leading to a higher residual Pu239 content at centre, as clearly demonstrated by SIMS measurements (Figure 3). The other isotopes show radial distributions in agreement with neutronic properties, e.g. Pu240 shows a strong depression at periphery, resulting in an increase of Pu241. The depression of Pu240 at centre results from the isotopic composition normalisation and the relative increasing abundance of Pu239. Absolute Pu240 content measurements in fact confirm that residual Pu240 content increase towards pellet centre as expected.

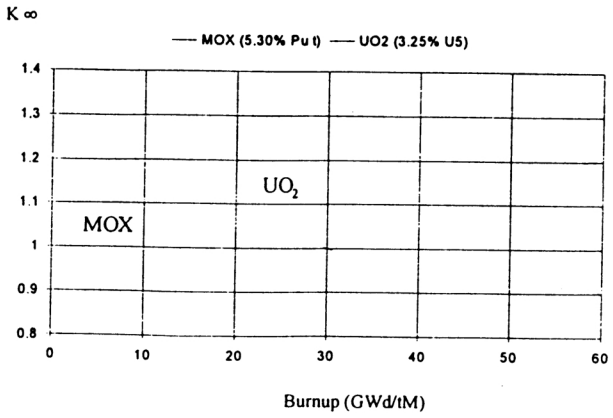

Fig. 2. Drop of reactivity as a Function of Burnup for MOX & UO_2 Fuel

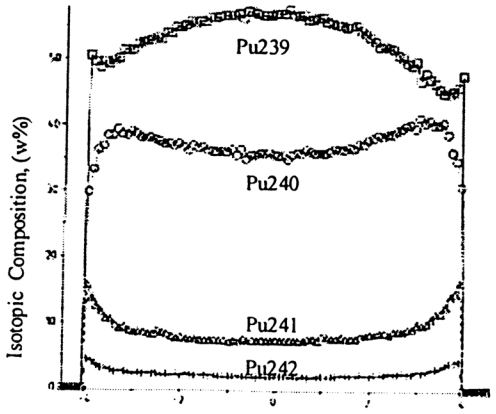

Fig. 3. Radial distribution of Pu isotopes across pellet measured by SIMS on irradiated MOX

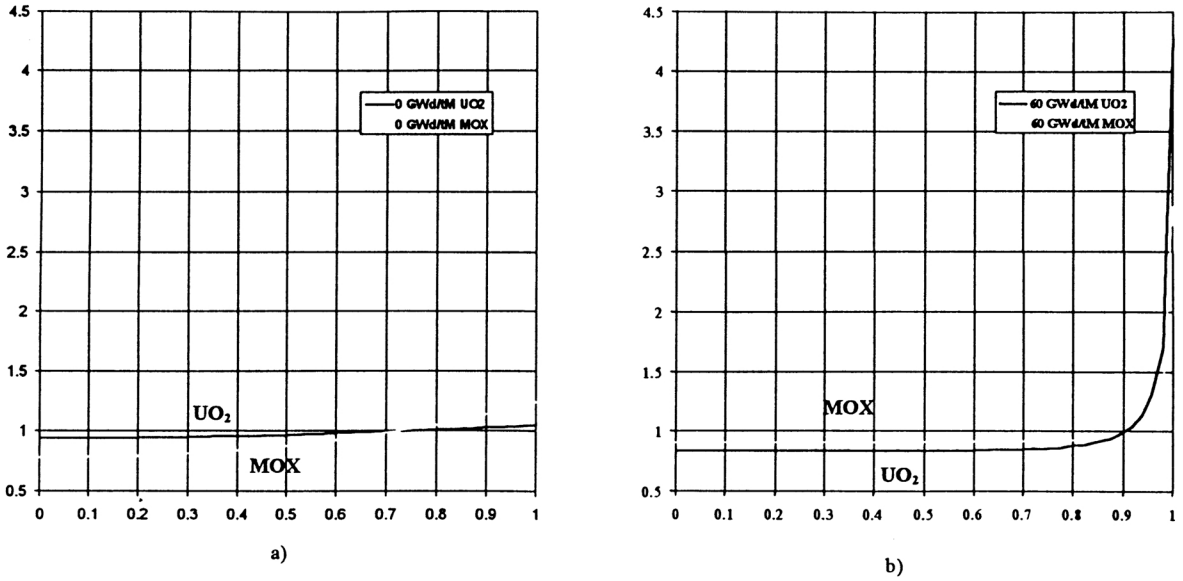

Fig. 4. Radial distribution of power in UO_2 (4.5% U5) and MOX (0.2% U5, 8.8% Pu) fuel pellet at a) 0 and b) 60 GWd/tM in PWR

At pellet rim, an increase in Pu239 content is observed. This build-up exists in MOX as well as in UO_2 fuel. Calculations indicate that the breeding of U238 in Pu239 is almost identical in MOX and UO_2. However, the net Pu239 production in MOX is influenced by the balance between the enhancement of the number of epithermal neutrons due to higher neutron production per fission, and the presence of Pu242 which absorbs neutrons on resonances overlapping with resonances of U238. Calculations suggested that breeding at rim is slightly less important in MOX than in UO_2.

Figure 4 illustrates the calculated evolution of radial power profiles versus burnup for UO_2 and MOX fuels. At BOL, the power is more depressed in MOX compared to UO_2, whereas at end of life (EOL), the higher residual Pu content at centre leads to higher local power in MOX fuel. The reduced breeding at MOX pellet periphery at high burnup contributes also to slightly increase the relative power at pellet centre.

3.3. **The fission products**

Fission yields for Pu239 and Pu241 are generally close to that of U235. Significant differences are however noticed for the light fission products, the peak of which is shifted towards higher masses. Practically, this means a decrease of Kr (leading to a Xe/Kr -15 compared to 7 for UO_2), the stability of Mo, and an increase of Ru. The total fuel solid swelling versus burnup is therefore expected to be almost identical to UO_2, with enhanced Ru content in metallic inclusions created during irradiation.

3.4. **Helium production**

Helium is produced in MOX fuel in greater quantity than in UO_2. The main contribution to helium production in MOX is Cm242 (half life 0.45 years). The quantity produced is very depending on initial Pu content and quality, and on burnup (Figure 5). The total amount of produced helium remains always small compared to Xe and Kr contents, with a He / (Xe + Kr) ratio of 0.07 at 40 GWd/tM and 0.18 at 62 GWd/tM (according to assumptions of Figure 5).

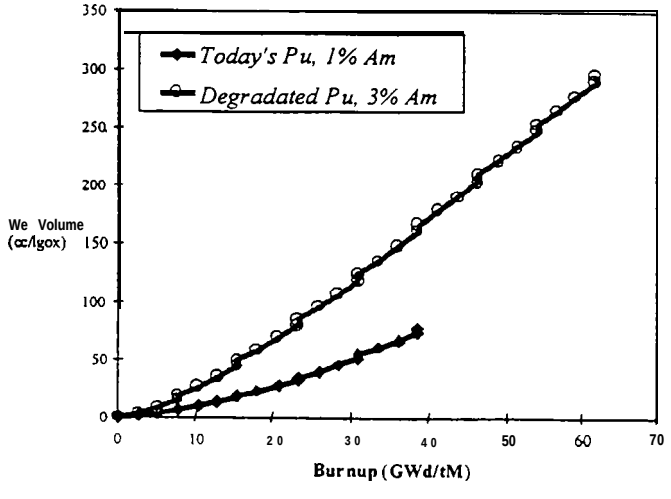

Fig. 5. Present & future production of helium in LWR MOX fuel

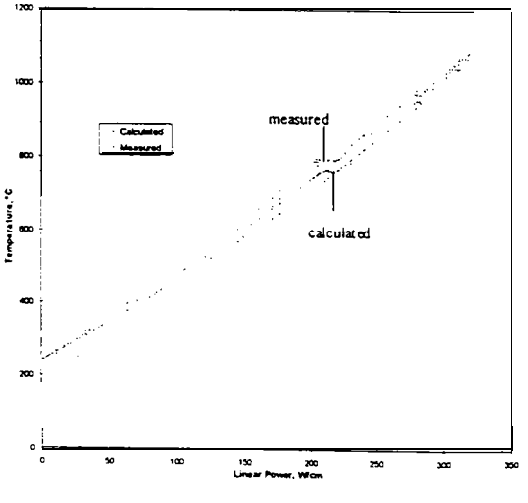

Fig. 6. FIGARO Experiment (IFA - 606) Temperature in the annular region during phase 2 of HALDEN irradiation

4. THERMAL MECHANICAL BEHAVIOUR

4.1. Thermal conductivity

A brief review of the thermal conductivity of MOX fuel has recently been published elsewhere [6]. This review concludes that the presence of small quantities of PuO_2 in UO_2 slightly decreases the thermal conductivity. Assuming that the thermal resistivity can be represented below 1500°C by the expression 1 / k = A + BT, the A and BT terms representing phonon scattering and phonon-phonon scattering respectively, then the presence of Pu increases the A and B terms due to reduction of atomic radius and of melting temperature respectively (Table 2). The overall conductivity decrease is small, typically 4 % for 10 % Pu/U+Pu.

With burnup, the conductivity of MOX fuel degrades due to irradiation damage and accumulation of fission products in stable or dynamic solution. The degradation is expected to follow quantitatively that of UO_2 as MOX is composed by UO_2 for more than 90 % and as the fission yields are close for Pu239, Pu241 and U235.

Measurements of fuel central temperature on MOX irradiated up to 50 GWd/tM were made in the framework of the FIGARO Programme [5]. Temperature calculations performed with the COMETHE-4D Code show that measured temperatures can be well reproduced assuming that the conductivity degradation with burnup in MOX is the same as in UO_2 (Figure 6).

4.2. Fuel central **temperature**

Calculations of fuel central temperature versus burnup are summarized in Table 3. At BOL, the MOX fuel operates with central temperature lower than in UO_2 (the difference reaches about 30°C at 250 W/cm), due to higher depression of radial power at centre. At 80 GWd/tM, the reverse situation is noticed, MOX operates about 85°C above UO_2 fuel at 250 W/cm, due to the increasing residual Pu239 towards centre.

Material	UO_2	PuO_2
crystallographic structure	Fluorite SC : O^{2-} ; FCC : M^{4+}	
lattice parameter (Å)	5.470	5.396
melting temperature (°C)	2840	2290
cation atomic radius (Å)	0.97	0.93
density (g/cm³)	10.96	11.45
molecular mass	~ 270	~ 271

Table 2. Comparison of UO, and PuO_2 properties having an influence on thermal conductivity

k type	k (UO_2, Bu = 0)	k* (UO_2, Bu)	k* (MOX, Bu)
$\delta(T_c - T_s)$(°C) (2)			
BOL			
150 W/cm	-25	-25	-16
250 W/cm	-51	-51	-33
EOL			
150 W/cm	11	17	27
250 W/cm	24	65	85
T_c(°C) (3) EOL			
150 W/cm	570	840	850
250 W/cm	770	1250	1260

(1) UO_2 (4.5 % U5) and MOX (0.2 % U5 ; 8.8% Pu) up to 60 GWd/tM
(2) $\delta(T_c - T_s) = (T_c - T_s)$ MOX - $(T_c - T_s)$ UO_2
T_s ~ 500° C BOL - open initial gap
350° C cbscd gap (400° C at EOL/250W/cm for k = k*)
(3) $T_c = (T_c(MOX) + T_c(UO_2))/2$

Table 3. Comparison between MOX and UO, fuel central temperatures for different types of MOX thermal conductivities (1)

4.3. **Pellet cladding mechanical interaction**

Post-irradiation examinations on several hundreds MOX and UO_2 rods indicate that both fuels have the same level of mechanical interaction with the cladding. This result can be explained once again by the fact that MOX is made for more than 90 % of UO_2 and generates nearly the same fission products.

In ramp conditions, the superiority of MOX over UO_2 has been demonstrated [7]. This superiority has recently been confirmed in the M308 International Programme with tests on MOX fuel fabricated following the MIMAS process. Three PWR segments pre-irradiated up to 58 GWd/tM were ramped at 100 W/cm min., followed by a hold of 24 hours respectively at 430, 450 and 500 W/cm. None of the segments failed.

4.4. Fission gas release (FGR)

Fission gas released from fuel is extensively investigated, as being a key parameter in high burnup achievement. A topic largely debated is the possible enhancement of FGR in MOX fuel, especially in case of Pu distributed in Pu-rich zones across pellet. Early results (accumulated in the years 1970-80) suggested an acceleration of FGR in MOX fuels compared to UO_2. These observations were made on unpressurized fuel rods loaded with unstable fuel. Moreover, the concerned fuel was prepared by mixing UO_2 granules with PuO_2 powder, giving a highly heterogeneous fuel, with large open and flat porosity and preferential path for gaseous fission products. Such fuel promoted FGR and thermal feedback.

Since these early observations, the fabrication process has been changed ; numerous experimental results concerning more recent fuel fabrications were obtained and detailed neutronic studies were performed.

The experimental results of the PRIM0 and DOMO international programmes in which various fuel types were irradiated and examined, conclude that FGR in MOX and UO_2 are similar and governed mainly by power history [3, 41.

French results indicate [8] that MOX fuel irradiated three cycles show an acceleration of FGR compared to UO_2 fuel. Analysis of that situation reveals that FGR increase can be attributed to a large extent to the higher linear power sustained by MOX fuel in these fuel assemblies. This assumption is confirmed in the same paper by examination results of a MOX assembly loaded at core periphery for a fourth cycle and showing no additional FGR.

Study of fuel operating conditions conclude that MOX and UO_2 fuels are indeed never strictly irradiated in the same power conditions, due to smaller drop of reactivity versus burnup for MOX fuel. This promotes – depending on core reshuffling policy – higher linear power in MOX late in life compared to UO_2. On the other hand, with the same rating, MOX fuel sustains larger central temperature (§ 4.1) with increasing burnup, favouring also FGR.

The conclusions drawn from these studies and our programmes are that FGR in MOX and UO_2 fuels are governed by the same parameters, i.e. rod linear power, fuel surface temperature, central temperature and burnup, and fuel microstructural parameters such as open porosity and grain size. The same conclusion was reached elsewhere too [9].

The COMETHE code includes model of FGR developed and calibrated for UO_2 fuels. It has been used to simulate the pressure increase in MOX fuel segments irradiated in Halden reactor in the framework of the FIGARO Programme [5] (Figure 8). Onset of FGR is observed around 1200°C, which is close to the Halden threshold for FGR at a burnup of 48 GWd/tM. Total FGR observed at end of test reached 12 %, which is also well reproduced by the code. The inner pressure however is overestimated, due to overprediction of helium release during test.

One major conclusion of our studies on FGR is that the MOX fuel releases more than UO_2 because it sustains generally larger central temperatures. For this reason, achievement of burnup with MOX (and UO_2) well above the present limits has required and will continue to require optimization and changes in

fuel fabrication, fuel design and design criteria, and operating conditions. A non exhaustive list of actions needing particular attention is :

increase of rod free volume at fabrication
diminish excessive free volume reduction during irradiation (cladding creep-down, fuel accommodation,)
optimize core reshuffling strategy (reduction of maximum power late in life,)
develop fuels more resistant to FGR (large gram,).
accept new design criteria (non lift-off, .).

4.5. **Microstructure**

Microstructure of MUX fuel is governed to a iarge extent by the UO_2 matrix in which PuO_2 is finely dispersed at fabrication. The MOX fuel will therefore sustain during irradiation the common phenomena of densification, swelling accommodation, and intergranular and intragranular porosity development associated to fission gas production and accumulation.

MOX fuels obtained by mechanical process show a fine dispersion of Pu across pellet, with the presence of some microscopic Pu-rich zones of Pu content larger than pellet average. The detailed study of such fuels provides useful information on evolution of fuel at high burnup in the Pu-rich zones and in the UO_2 matrix. These observations can to some extent be applied to the rim structure occurring in UO_2 when local burnup reaches 60-80 GWd/tM.

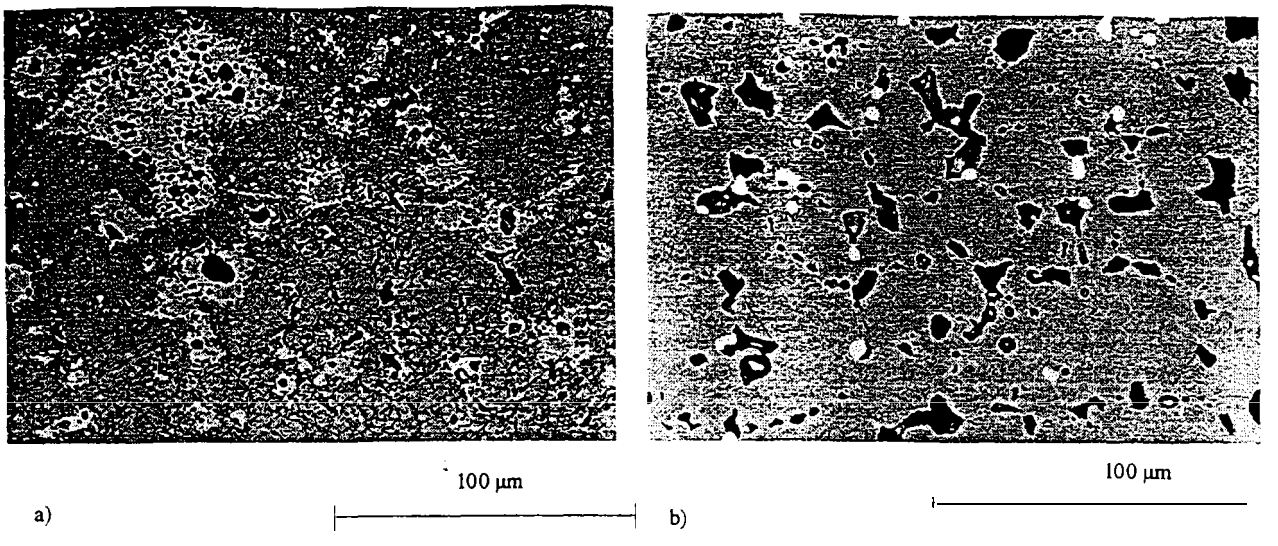

Fig. 7. Microstructure evolution of Pu-rich zone irradiated at (a) low and (b) high temperature

The negligible solubility of fission gas leads, in the Pu-rich zones irradiated at low temperature, to the formation of intragranular nanometer-sized bubbles. With burnup increase, they reach a micron size and become visible by optical microscopy (Figure 7.a). When irradiation proceeds, these bubbles continue to grow whereas their density number decreases. This process is pursuing as long as the temperature remains low (< 1000°C), and for burnup much beyond 200 GWd/tM.

Grains subdivide in Pu-rich zones reaching a submicron size, typical of the "rim structure" [10]. This structure is observed for temperature as high as 1000°C. Formation of rim structure at such a temperature is noticed also for UO_2 [11].

The extrapolation of this result to UO_2 fuel indicates that the existence in UO_2 of rim structure over a depth of about 300 µm and its rapid disappearance towards pellet centre must be attributed to local burnup drop and not to the temperature increase towards centre as long as thermally assisted diffusion processes remain negligible.

The structure of the Pu-rich zones changes when fuel operates above some temperature, with a restructuring similar to UO_2 fuel operating in the same conditions [10]. At sufficiently high temperature, grain growth and development of intergranular porosity typical of UO_2 are noticed for MOX as well (Figure 7.b).

About the rim structure development in MOX fuel, the neutronic studies conclude to accumulation at pellet periphery of burnup similar to UO_2. In the MIMAS fuel, the Pu is distributed at fabrication between the UO_2 matrix where it is finely dispersed, and few Pu-rich zones. The average burnup reached in UO_2 matrix is therefore lower than pellet average, which has for effect to postpone the development of a continuous rim structure at pellet periphery.

4.6. Helium release

Helium is produced in MOX fuel in significant quantity, but lower than that of noble fission gas (§ 3.4). Experimental inventory of helium content in free volume of irradiated MOX and UO_2 - pre-pressurized and unpressurized - rods has been made, with the following conclusions :

- excess of helium compared to as fabricated content is found in low-pressurized MOX rods, increasing with rating and burnup,
- increase in helium partial pressure can be of the same order as pressure increase due to noble fission gas,
- helium balance is often negative at low burnup,
- no excess of helium is found in rods pressurized at 20 bars,
- UO_2 rods show a negative balance of helium,
- dispersion in obtained results is large.

The absence of helium content increase in free volume in pre-pressurized rods over a broad range of burnup has been observed for MOX rods as well [12].

Study of helium behaviour made elsewhere shows that helium diffusion coefficient in UO_2 is several orders of magnitude larger than that of noble fission gas [13] and that helium is highly soluble in UO_2 [14]. In view of that, the above experimental observations were interpreted :

- large dispersion on helium balance comes from uncertainty on initial rod free volume and pressure,
- negative helium balance for UO_2 could be attributed to infusion of helium in fuel during irradiation,
- helium is not released from fuel as long as its content does not exceed the solubility limit, which - depends on helium partial pressure in rod,
- fraction of released helium is depending on operating conditions, and can be correlated to fraction of FGR.

Modelling of helium release has been made on base of experimental results and an empirical correlation set up between helium and FGR release [15]. Simulation of helium release during a BWR MOX irradiation is shown at Figure 9. Conservative results indicate an helium partial pressure increase comparable to fission gas partial pressure. The conservative curve corresponds to use of the envelop correlation inferred from experience. Best estimate result is unaccurate as many uncertainties exist in helium production term, solubility effects and extent of diffusion.

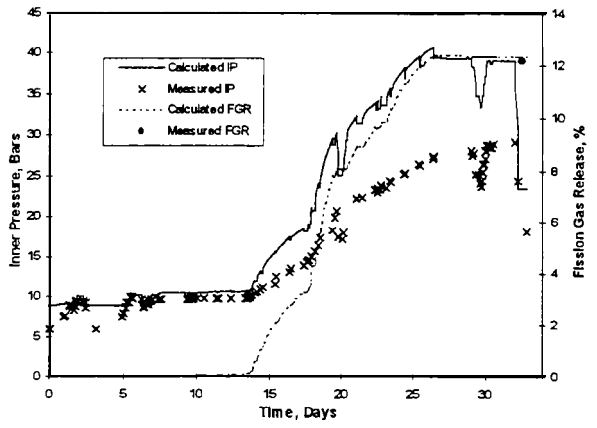

Fig. 8. Calculated and measured FGR and inner pressure in FIGARO experiment (IFA 606/phase 2)

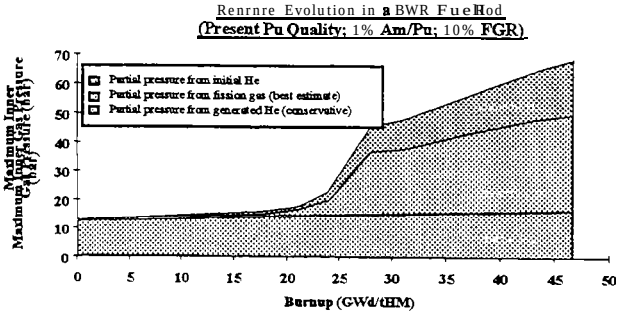

Fig. 9. Pressure Evolution in a BWR Fuel Rod (Present Pu BOL 1% Am/Pu; 10% FGR)

5. Conclusions

UO_2 and PuO_2 materials have properties relevant to in reactor performance sufficiently close to each other to fabricate (UO_2-PuO_2) fuel and to burn it according to scenarios largely conceived to burn UO_2. The R&D efforts made to achieve high burnup with MOX fuel are therefore largely similar to UO_2 activities, with special emphasis however on the evolution of the actinide chain and its consequences, and FGR. The conclusions of our observations on MOX behaviour at high burnup are :

1□ With burnup, the Pu content is modified due to a drop of Pu239. The other isotopes remain almost constant,
2□ The reactivity drop with burnup is much less pronounced than in UO_2,
3□ The minor actinides Am242m and Cm243/245 start to play a role in total reactivity with a contribution of several %,
4□ Difference in radial distribution of Pu burning across pellet results in high residual Pu at pellet centre, and thus to higher fuel central temperature with burnup increase,
5□ The presence of Pu slightly reduces the fuel thermal conductivity, about 4 % for 10 % Pu,
6□ The burnup degrades the fuel thermal conductivity as in UO_2,
7□ MOX fuel operates under different temperature conditions than UO_2 due to conclusions 2, 5 and 6. This increases FGR in MOX and must be taken into account for high burnup achievement,
8□ PCMI in steady state conditions is similar to UO_2. The behaviour in ramp condition is favourable to MOX fuel,
9□ The microstructure evolution during irradiation follows that of UO_2 which represents 90 % of the fuel. Investigations of microscopic Pu-rich zones included in pellet indicate a rim type structure. With burnup increase, such zones show pore size increase and density number decrease. Such a structure is observed up to about 1000°C,
10□ The decay of Cm242 leads to observable helium production and release, especially for lowly pressurized high rated fuel.

In view of these conclusions, the achievement with MOX fuel of burnups higher than achieved in routine today in LWRs is feasible. Recommendations for further R&D work are :

- accumulate experimentally burnups of at least 80 GWd/tM at low power
- obtain accurate data on actinide chain
- confirm fuel central temperature evaluation
- obtain data on helium production, solubility and release
- develop fuels having improved gas retention
- study rods in non lift-off conditions.

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PIE OF BNFL'S FIRST COMMERCIALLY IRRADIATED SBR MOX FUEL

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Abstract

Assemblies containing SBR MOX fuel. fabricated in the MOX Demonstration Facility (MDF) at Sellafield, were loaded into NOK's Beznau-1 reactor in 1994. The fuel was irradiated for 3 cycles to assembly average burnups of 33 MWd/kgHM and subsequently rods were extracted and sent for Post Irradiation Examination (PIE) at the European Institute for Transuranium Elements (ITU). This paper presents the detailed results of this PIE investigation so far. Comparisons are also made between the PIE results and predictions from the fuel performance code ENIGMA-B to place the PIE results in context. Overall it is shown that the performance of the SBR MOX fuel is good.

I. INTRODUCTION

BNFL started making LWR MOX fuel via the Short Binderless Route (SBR) [1] in 1990 using small scale laboratory facilities. This fuel has been loaded into a number of test irradiations including a Halden experiment [2] and the Belgonucleaire (BN) Callisto/PRIMO 2 programme [3]. BNFL started producing commercial. assembly scale fuel in the MOX Demonstration Facility (MDF) in 1993. In 1994. four assemblies manufactured in MDF containing SBR MOX fuel were loaded into region MS of NOK's Beznau-1 reactor, a 2 loop Westinghouse PWR. All the M5 assemblies are standard Westinghouse 14x14 design. As this was the first commercial reactor fuel produced in MDF some of the rods in the M50 assembly were specially characterised and measured prior to irradiation with a view to performing PIE. The fuel has now been irradiated for three. 12 month cycles to assembly average burnups of order 33 MWd/kgHM. In February 1998. 7 fuel rods were extracted from assembly M50 and transported to ITU for PIE.

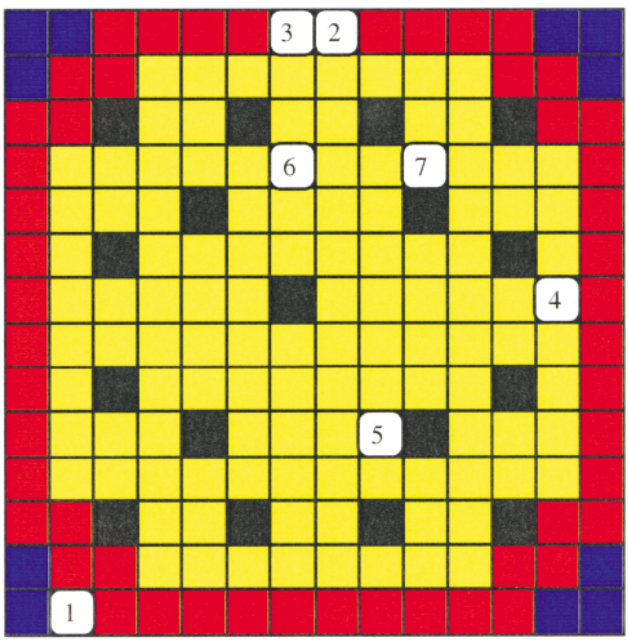
BNFL maintains a state of the art fuel rod performance modelling code ENIGMA-B [4,5]. The code has the capability to model UO_2, Cid doped UO_2 and MOX fuel and has been used to license fuel of all three types. The code itself and in particular the changes to the models for calculations on MOX fuel are presented in another paper at this conference [6]. ENIGMA has been extensively validated against irradiations of 414 UO_2 and 55 MOX rods in a range of designs. The code prcdictions help place the PIE results in context and give confidence that the M50I fuel performance is within the expected range

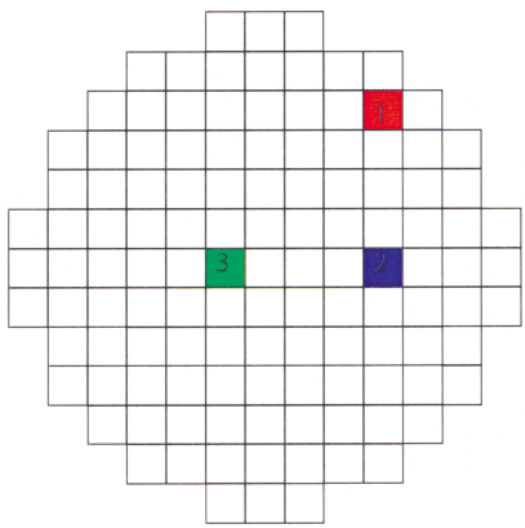
2. RODS EXAMINED

Table I shows the rods extracted for PIE. The burnups and power histories were calculated by BNFL using the CASMO-4 and SIMULATE-3 (CMS) nuclear design codes [7]. The position of each rod in the assembly is shown in Figure 1a below, whilst Figure 1b shows the placement of the assembly in the core during the 3 cycles of irradiation.

TABLE I : RODS EXTRACTED FOR PIE

Rod	1	2	3	4	5	6	7
Pu_{total} Enrichment (%)	2.92	3.72	3.72	5.54	5.54	5.54	5.54
Burnup (MWd/kgHM)	32.5	31.2	31.2	33.9	33.7	32.7	35.6





Water rod Low enrichment Med. enrichment High enrichment

Figure 1a : Assembly diagram

Figure 1b : Assembly placement in core

It is worth noting that the calculated burnup in the medium enrichment rods is significantly lower than that in the high enrichment rods whilst the burnup of the low enrichment rod is not suppressed to the same extent. This is due to the orientation of the assembly during cycle 1 when the low enrichment rod was closer to the core centre than the medium enrichment rods and the proximity of the low enrichment rod to UO_2 fuel. Chemical burnup determinations will be performed to confirm the calculated burnups.

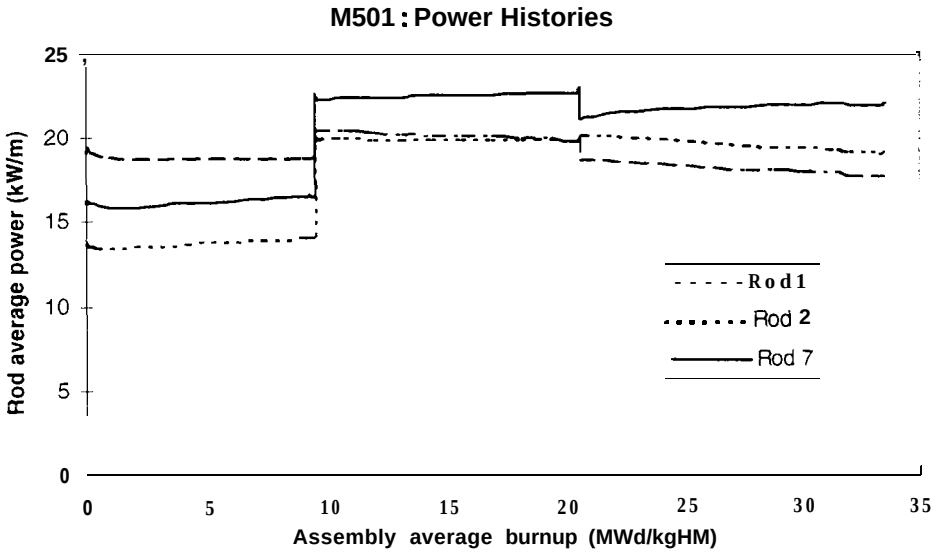


Figure 1c : Power history *for low, medium und high enrichment rod*

The power histories for Rods 1, 2 and 7 are shown in Figure lc. These power histories were calculated with the CMS codes and the same power histories were used in the ENIGMA predictions shown in this paper. The three reactor cycles are clearly visible; the relative power difference between the cycles is due to the location of the assembly in the core.

The calculated axial burnup profile for Rod 7 which had the highest burnup is shown in Figure Id.

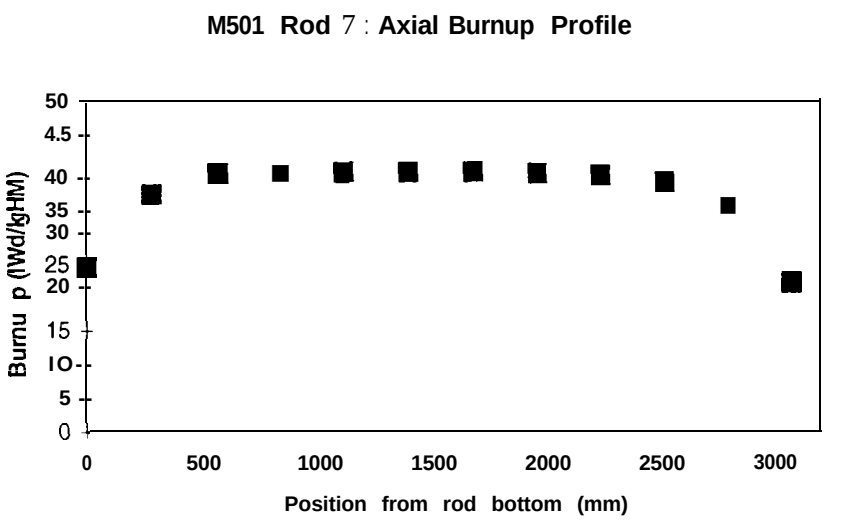


Figure 1d : Rod 7 axial burnup profile

The predicted burnup profile is flat with the expected local reductions at the top and bottom of the fuel stack.

3. NON-DESTRUCTIVE EXAMINATIONS

Each rod was examined using the following NDE (Non-Destructive Examination) techniques,

- Visual Examination
- Eddy Current oxide thickness measurement (ECT) and defect scans

- Rod diameter profiles and rod length measurements
- Gross γ - scanning and stack length measurement
- Isotopic γ - scanning measurement for 6 isotopes

3.1 Visual Examinations

Photographs of the outside of each rod were taken on two axes with 180 degrees separation. The rod identification number was used to define the 0 degree axis for the visual and further examinations. Approximately 8 images were taken on each axis to cover most of the rod length giving 16 pictures / rod. The overall impression from the complete set of photographs is of good fuel rod condition with the expected pattern of corrosion for the duty experienced and no indication of abnormalities in the cladding.

3.2 ECT Measurements

The waterside oxide thickness was measured along 4 axes at right angles using ECT. The average oxide thickness along Rod 6, which is typical of the M501 data, is shown in Figure 2 along with the predicted oxide thickness using ENIGMA. The clad is low tin Zircaloy-4 and the core inlet temperature is 283 °C.

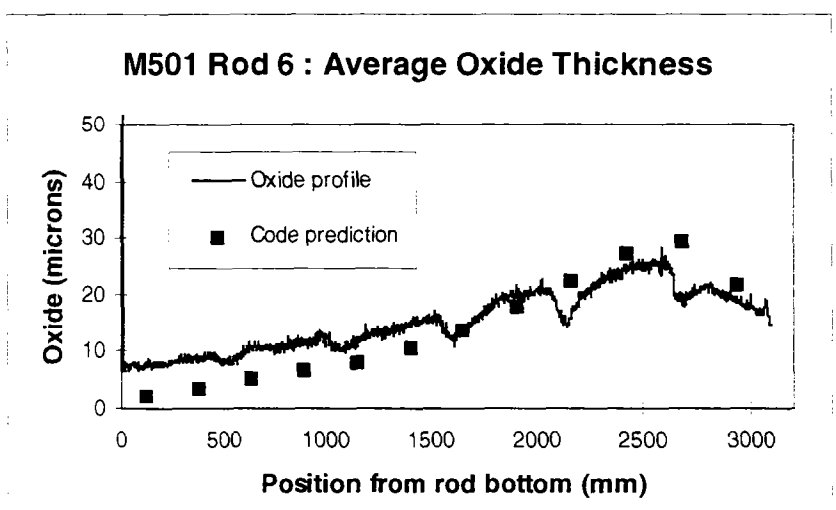


Figure 2 : ECT oxide profile for Rod 6

The oxide distribution is principally determined by the temperature profile on the clad exterior and this is reflected in the increase in oxide thickness up the rod. The reduction in oxide at the grid sites is also clearly seen in the figure. Comparison of the individual traces along separate axes revealed a degree of variation in the oxide thickness around the circumference of the rod with measurements along the different axes varying by up to 10 μm at the peak oxide position.

ENIGMA gives an underprediction of the oxide thickness at the lower end of the rod and a slight overprediction in peak oxide thickness but in general the oxide profile is well modelled confirming that the corrosion behaviour of the fuel is in line with experience from similar irradiations.

ECT defect scans were also performed on each rod. These scans confirmed the integrity of all the rods.

3.3 Rod Length Measurement

The length of each rod was measured before and after irradiation. The % length change v burnup is plotted in Figure 3 along with two prediction curves. The growth curve is derived from measurements on

empty, irradiated, Zircaloy tubes whilst the design curve is a correlation derived from PIE measurements on commercially irradiated UO_2 fuel. A fast fluence value of $1.73*10^{24}$ n m^{-2} / unit burnup (>1 MeV), which lies within the range of the fast fluence values of the M501 rods, was used to calculate these curves. The data tend to be slightly below the UO_2 design curve which suggests a slightly lower mechanical interaction between pellets and clad in MOX fuel. However they are generally well predicted, with the scatter of order x / ÷ 1.5. The M501 data is consistent with published German and French results which indicates that rod growth for MOX fuel lies within the scatter of the UO_2 data [8,9].

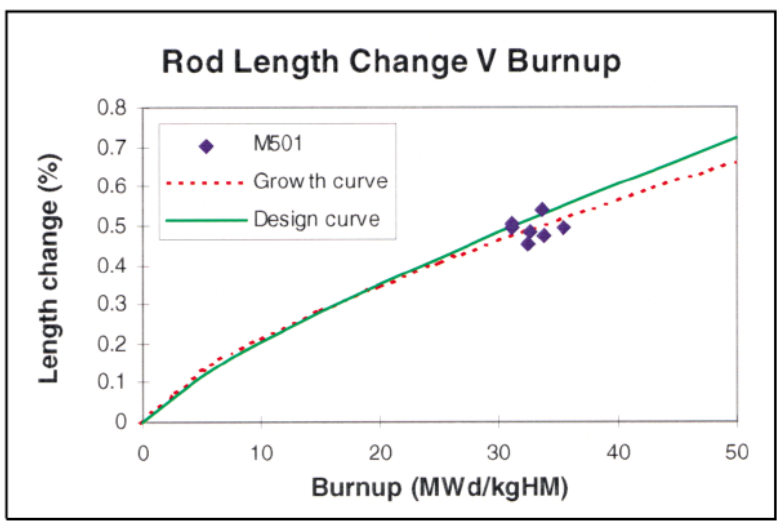


Figure 3 : Axial growth of M501 rods

3.4 Rod Diameter Profiles

Each rod diameter was measured using a contact probe method on two axes. The measurements are accurate to ± 5 µm.

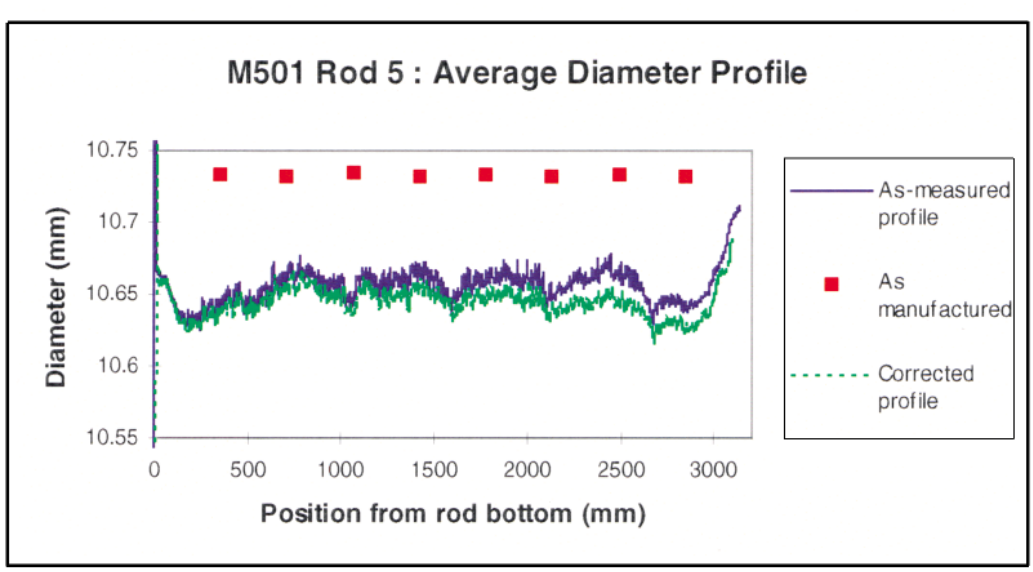


Figure 4 : Diameter profile for Rod 5

Figure 4 shows the rod diameter after fabrication, the average diameter profile and the oxide corrected profile using the Pilling-Bedworth ratio as recommended in [10]. Both profiles show clad creepdown along the length of the rod. At the top and bottom of the fuel stack where creepdown is most pronounced the maximum clad strain is of order 1 % which is in line with other published data on MOX fuel [8]. Both

profiles show fuel/clad contact and creep reversal due to fuel swelling which has occurred along the length of the rod reflecting the axial burnup distribution, (see Figure 5a). The M50 1 diameter profiles are consistent with other data available to BNFL from comparable MOX fuel irradiations.

3.5 Gross γ -Scans

Gross y-scans were taken on each rod and the scan for Rod 7 is shown in Figure 5a. On the rod length scale the scan gives an indication of the rod axial burnup distribution. This shape is generally in agreement with the calculated burnup distribution (Figure Id). The effect of the grids in lowering the local burnup can clearly be seen. On the finer scale, pellet-pellet interfaces can also be resolved as shown in Figure 5b. Detailed anaiysis of the y-scan data has revealed that there are no significant inter-peiiet gaps and that the fuel column is in a sound condition. The clearly resolved pellet-pellet interfaces are a sign that there has been very little migration of the volatile element Cs and limited dimple filling due to fuel creep.

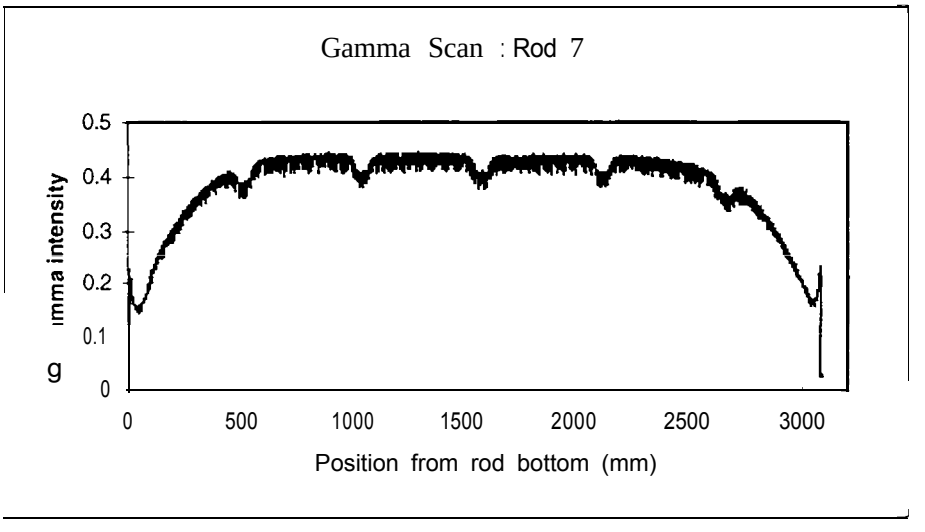


Figure 5a** : **Gross γ-scan for Rod 7

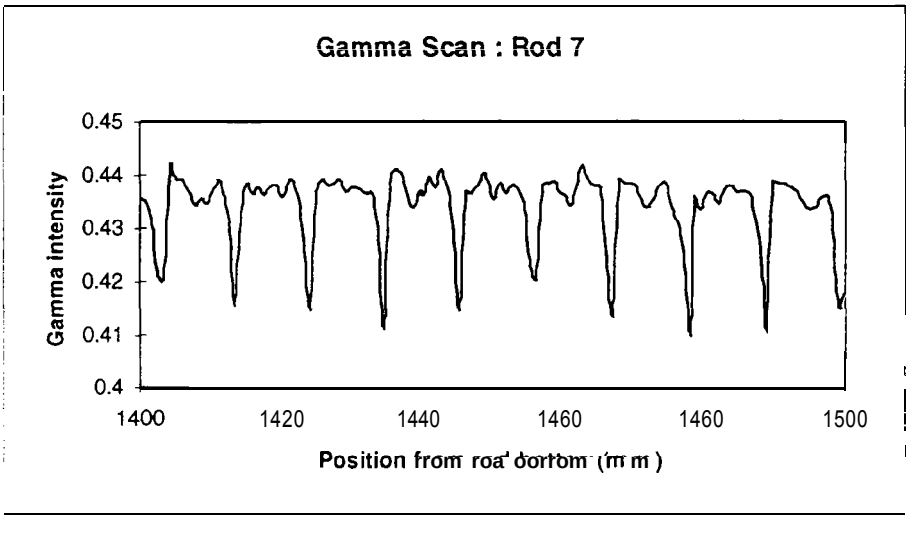


Figure 5b** : **Detailed view of gross γ-scan

The fuel stack length has been measured from the y scans and compared with the as-fabricated stack length measured from the full-length radiographs. The average length increase was 0.5 1 %. The results are shown in Figure 6.

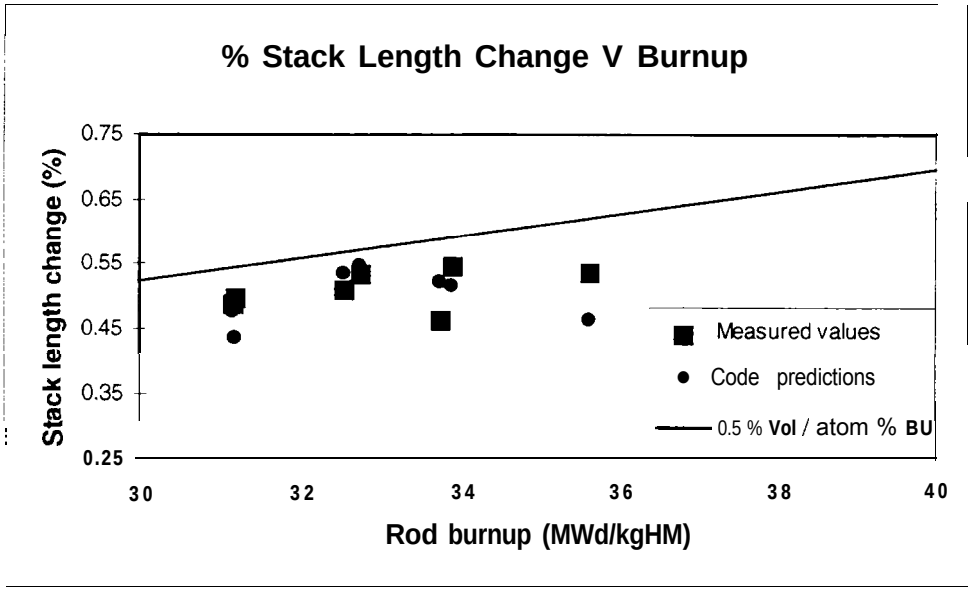


Figure 6 : Stack length change for M501 rods

The change in fuel stack length is primarily determined by fuel dimensional changes caused by densification and swelling. A line corresponding to 0.5 % vol / atom % burnup (9.55 MWd/kgHM) is plotted to give an indication of the total swelling rate with no densification. The stack length change data are well modelled by ENIGMA giving confidence that the dimensional behaviour of SBR MOX fuel is good.

3.6 **Isotopic γ Scans**

Six isotopes (^{144}Ce, ^{137}Cs, ^{134}Cs, ^{106}Ru, ^{95}Nb and ^{95}Zr) were selected for measurement and isotopic y-scans were performed along the length of each rod. The ^{137}Cs scan confirmed that the axial burnup distribution was in agreement with the calculated profiles.

4. GAS PUNCTURE RESULTS

All **7** rods were punctured to determine the rod internal pressure, free volume and isotopic content; the pressure and volume were measured at room temperature. Results are shown in Table II.

TABLE II : FISSION GAS PUNCTURE RESULTS

Rod	Pressure (bar)	Volume Reduction (%)	FGR (%)	Xe/Kr ratio
1	33.1	37.1	0.39	16.7
2	32.7	38.2	0.42	16.8
3	32.0	35.6	0.41	17.0
4	32.6	37.6	0.55	17.1
5	33.2	38.2	0.59	16.4
6	33.4	36.8	0.50	16.8
7	34.0	35.1	1.09	16.2

The pressure values, normalised to the coolant temperature are plotted in Figure 7.

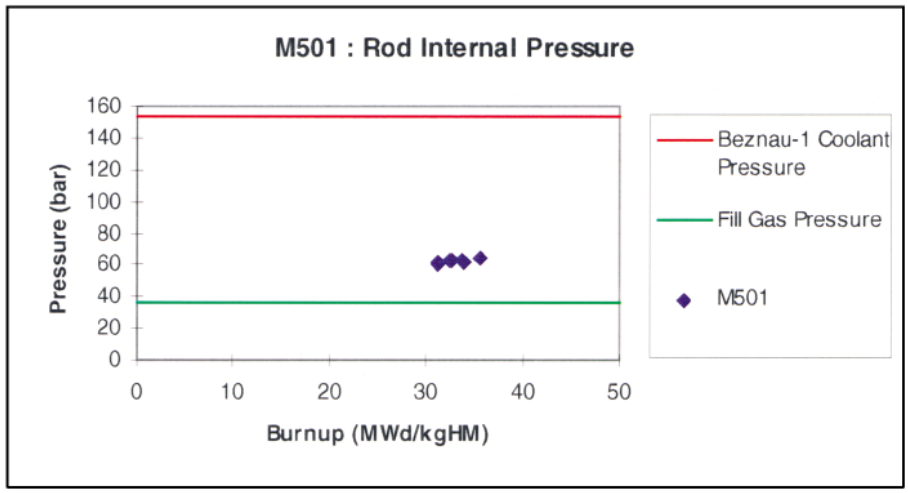


Figure 7 : Puncture results normalised to coolant temperature

The graph shows that there is a significant margin between rod internal and coolant pressure and also shows that there has been an increase compared to the fill gas pressure. Further analysis shows that with 0 % FGR the rod gas pressure would be ~ 28 bar at room temperature due to free volume reduction alone. Much of the volume reduction is associated with clad creepdown along the fuel stack and therefore much of the pressure increase during irradiation is not related to the behaviour of the fuel.

The fission gas release data are plotted in Figure 8 as a function of burnup. The fission gas generation rate is not so well defined in MOX fuel as it is in UO_2 and primarily depends on the relative concentrations of Pu_{239} and Pu_{241} in the fuel. In [11], White proposes a method for calculating the gas generation rate and using this method the gas generation rate for M501 is 27.43 cc/MWd, whilst in UO_2 the rate is 29.4 cc/MWd. Using this rate the FGR values have been calculated by dividing the total measured volumes of Xe and Kr (at STP) by the calculated STP volumes of Xe and Kr generated in each rod during irradiation. For M501 the values are well below 1 % with the exception of Rod 7 and the reason for this is discussed below.

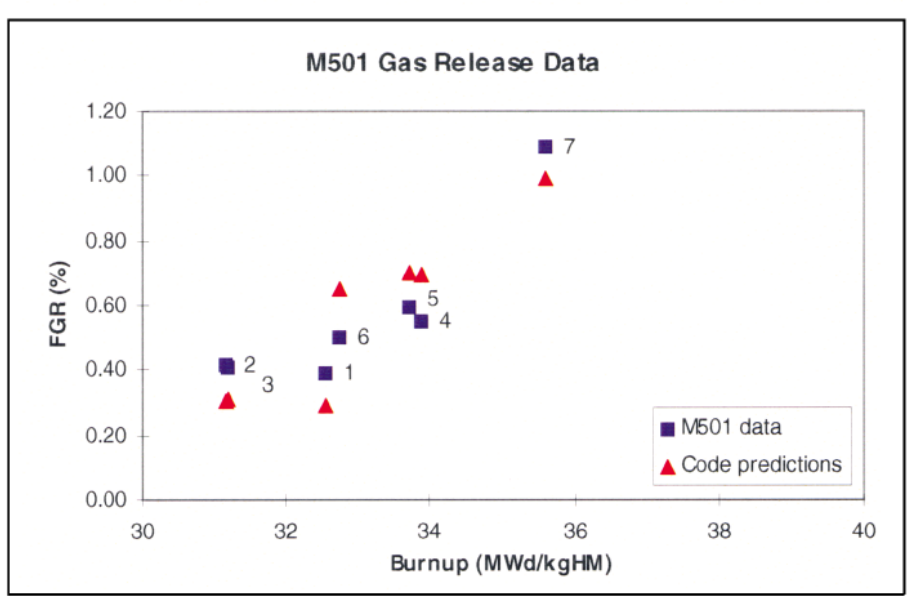


Figure 8 : FGR results for M501 rods

In general the FGR data is well modelled by ENIGMA, most of the predictions lie within 0.1 % FGR of the measured values and there is no systematic bias. Nitrogen release is also well modelled by the code. Rod 7, which had the highest burnup of the seven rods chosen, also displays the largest fission gas release. Figure 9 plots ENIGMA's prediction of the peak centreline temperature as a function of burnup. Also plotted on the graph is the Vitanza threshold [12] for 1 % fission gas release.

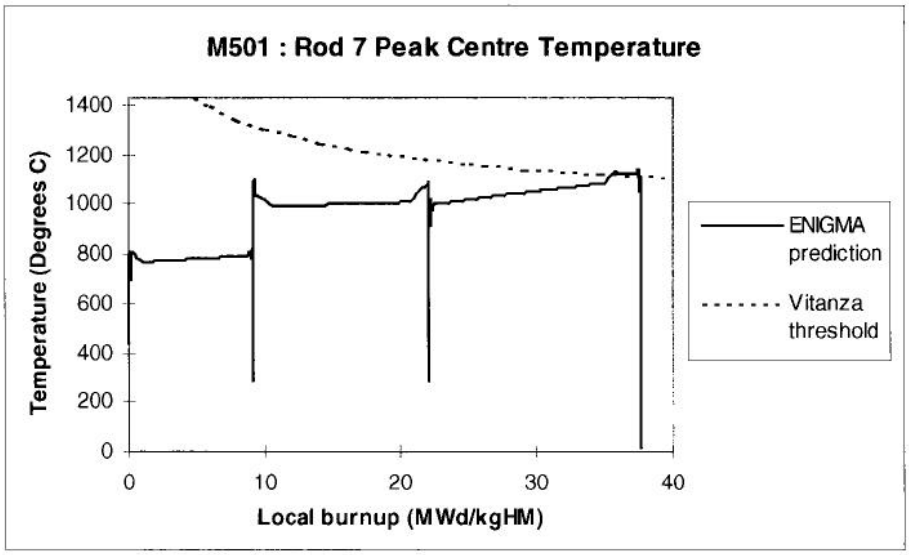


Figure 9 : Predicted centre temperature for Rod 7

The temperature curve is seen to intersect the threshold at the end of the third cycle of irradiation and this explains the increased fission gas release from Rod 7. Since the Vitanza threshold was originally derived from UO_2 fuel, the measured gas release of 1.09 % in Rod 7 adds weight to the argument that the onset point for thermal gas release is not significantly different in MOX fuel.

The magnitude of the Xe/Kr ratio reflects the fissile content of the fuel. UO_2 fuel would typically give values of around 8; the data in Table II illustrate the reduced yield of Kr isotopes from plutonium fissions.

5. DESTRUCTIVE PIE

Destructive PIE work is still in progress at ITU. Figure 10 shows an axial ceramography section from Rod 4. The section was cut between 928.5 and 948.5 mm from the bottom of the rod.

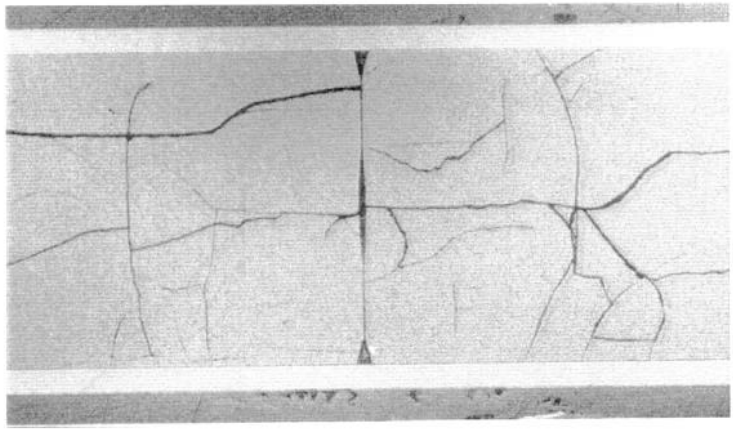


Figure 10 : Axial ceramography sample from Rod 4

The degree of cracking observed in the photograph is consistent with that in UO_2 fuel irradiated at similar power ratings. The chamfer and dimple are clearly visible in the centre of the picture confirming the results from they scans. This sample is typical of a series of axial sections taken from the M501 rods to examine pellet fragmentation and dimensional changes during irradiation.

6. SUMMARY AND CONCLUSIONS

Results have been presented from non-destructive testing, gas puncture and ceramography on SBR MOX fuel irradiated in a commercial PWR reactor to rod burnups in excess of 35 MWd/kgHM. The overall picture from the PIE of M501 fuel is that of good performance with significant margins to the design limits. The observations are within the envelope of MOX fuel experience with much of the data matching that from UO_2 fuel.

Destructive PIE is continuing at ITU and results from EPMA (Electron Probe Microanalysis), SEM (Scanning Electron Microscopy), density and other measurements will be published at future meetings.

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MOX FUEL IRRADIATION BEHAVIOUR IN A THERMAL REACTOR

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Abstract

JNC has developed the MOX fuel for thermal reactors over more than 30 years. As a part of this effort, various irradiation tests of the MOX fuel were conducted, and a lot of useful data on the MOX fuel behavior were accumulated. JNC has utilized these data to develop a MOX fuel rod performance evaluation code, which has been already applied to the design study of the ATR high burnup MOX fuel.

1. Introduction

The Japan Nuclear Cycle Development Institute (JNC) has developed the MOX fuel for the advanced thermal reactor (ATR), which is the heavy-water-moderated, boiling-light-water-cooled, pressure tube type reactor, and LWR more than 30 years. As a part of this development, JNC conducted various irradiation tests of the MOX fuels in thermal reactors such as ATR "Fugen" and Halden Boiling Water Reactor (HBWR). In this paper, we summarize the MOX fuel irradiation tests conducted in ATR "Fugen" and HBWR, and review the MOX fuel irradiation behavior in a thermal reactor.

2. Outline of MOX Fuel Irradiation Tests

JNC's major MOX fuel irradiation tests conducted in ATR "Fugen" and HBWR are summarized in Table 1. The irradiation condition and the specifications of ATR fuel are quite similar to those of BWR fuel.

2.1 Outline of ATR

The ATR is the heavy-water-moderated, boiling-light-water-cooled, pressure tube type reactor. Light-water coolant becomes in the core steam-water mixture to be carried to the steam drum for separation of steam before it is directed to the turbine. The coolant temperature and pressure are 284°C and 6.8MPa respectively, which are comparable to those of the BWR.

The ATR has the advantage of the flexibility in the fuel utilization because fast neutrons are slowed down in the heavy water moderator region. In the case of plutonium utilization in the ATR, plutonium isotopic composition slightly affects on the nuclear characteristics of the ATR.

2.2 Outline of ATR MOX Fuel

The ATR MOX fuel assemblies are shown in Fig. 1. To fit into the pressure tube design of the reactor, the fuel assembly is of a cylindrical configuration in which fuel pins are arranged in concentric rings. The fuel assembly is mainly composed of fuel pins, spacer supporting rods, the upper and lower tie-plates, and spacers. The upper and lower tie-plates, and spacers

Table I MOX Fuel irradiation Tests in HBWR and ATR "Fugen"

Irradiation Tests	Max.Pellet Burn-up (MWd/t)	MLHGR (kW/m)	Pu-fissile Content (wt%)	Pellet Diameter (mm)	Pellet Type	MOX Powder
HBWR						
IFA-5 I4	56000	46	4.6	10.5～ 10.57	Hollow&Solid	MB
IFA-529	34700	44	6.0	10.47～10.64	Solid	MB/MH
IFA-554/555	37400	56	3.4	12.4	Hollow &Solid	MB
IFA-565	65000	46	4.6	10.54～10.57	Hollow &Solid	MB
Fugen						
DATR-type (E04,E05)	40300	44.5	I .O-2.5	12.4	Solid	M H
Segment-type (EO6, E07)	32600	29	1.5～3.0	12.4	Hollow&Solid	MH
Gd203-type (E08, E09)	49200	45.7	I.5～3.9	12.4	Solid	MH
Standard-type (P06, P2R)	24400	49.8	0.55-I .56	14.4	Solid	MB/MH

MB : 100% PuO_2 powder was mechanically blended with UO_2 powder.

MH : 50% PuO_2-UO_2 powder was prepared by microwave heating, then mechanically blended with UO_2 powder.

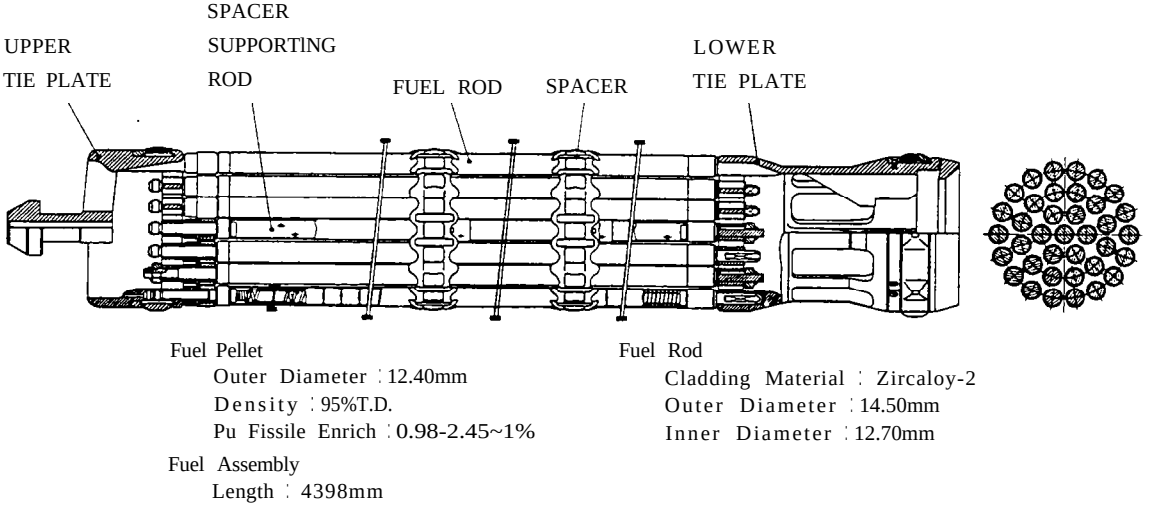

Fig. I Schematic drawing of an ATR36-rod fuel assembly

maintain the fuel pins in their desired positions. The fuel assembly is about 4.5m long and 110mm in diameter. Each pressure tube houses one assembly.

The Fugen standard fuel assembly is composed of 28 fuel pins of 16.26mm in diameter and is designed for the maximum assembly burn-up of 20GWd/t.

The 36 rods type fuel assembly was designed for the demonstration ATR (DATR). It is composed of 36 fuel pins of 14.5mm in diameter and is designed for the maximum assembly burn-up of 38GWd/t.

2.3 MH Process

Recent JNC MOX fuel pellets are fabricated from master blend MOX powder prepared by the microwave heating (MH) process[l]. In the MH process, the master blend is obtained through direct co-conversion of U-Pu mixed nitrate solution by microwave heating. Then, master blend is diluted and mixed with UO_2 powder by ball milling to obtain specific Pu content. The MH process flow diagram is shown in Fig. 2, compared with the mechanical blend (MB) method.

Due to direct co-conversion and subsequent -ball milling, MH pellets have excellent homogeneity of Pu distribution. Fig. 3 shows an example of the a-autoradiography of a MH MOX pellet.

3. MOX Fuel Irradiation Behavior

3.1 Fission Gas Release

Measured fission gas release rate of the MOX fuel rods are shown in Fig. 4 as a function of rod average burnup and of the experienced maximum linear heat rate, compared with those of UO_2 fuels irradiated in a Japanese BWR [2,3].

Fission gas release rates of the MOX fuels are within the data spread of the UO_2 fuels. That is, fission gas release behavior of the MOX fuel is quite similar to that of UO_2 fuel. It is well known that the homogeneity of MOX fuel affects its fission gas release rate at high burnup. However, distinguishable difference was not observed between JNC MOX fuel and the UO_2 fuels, and the excellent homogeneity of JNC MOX fuel was confirmed by the post irradiation examinations.

3.2 Helium Release

In MOX fuels, significant amount of helium is detected by puncture tests. In Fig. 5, measured amount of helium is plotted as a function of rod average burnup, compared with those of UO_2 fuels irradiated in a Japanese BWR [2, 3].

In MOX fuels, ^{242}Cm generation during irradiation is much greater than UO_2 due to its high Pu content, which results in larger generation of helium. Measured amount of released helium was apparently larger in MOX rods than in UO_2 rods.

Also Fig. 5 depicts relationship between amount of helium and fission gas released. It is observed that behavior of helium release was similar to that of fission gas release with respect to its dependence on burnup and linear heat rate.

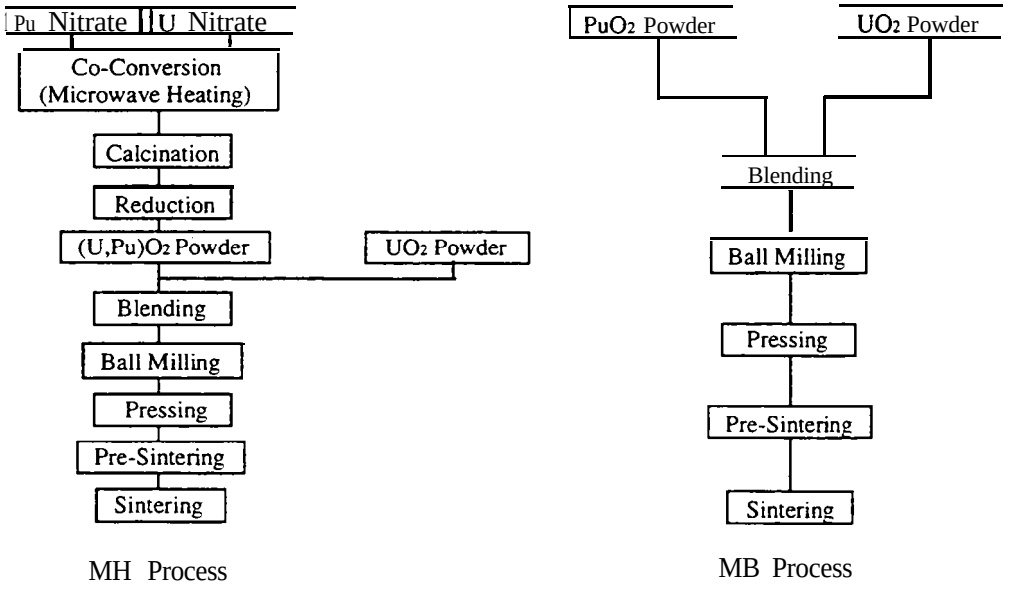

Fig.2 MOX Fuel Pellet Fabrication Process

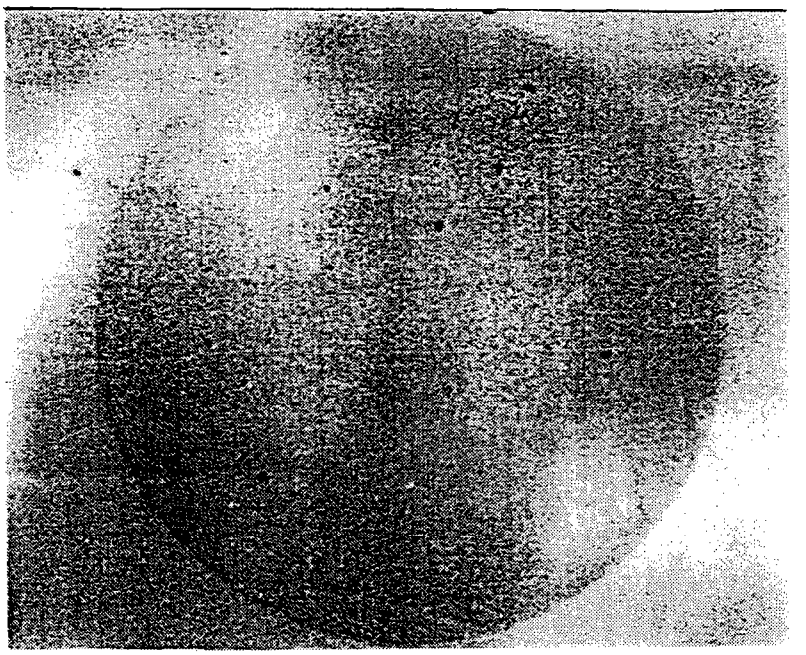

Fig.3 a -autoradiography(MH MOX Pellet)

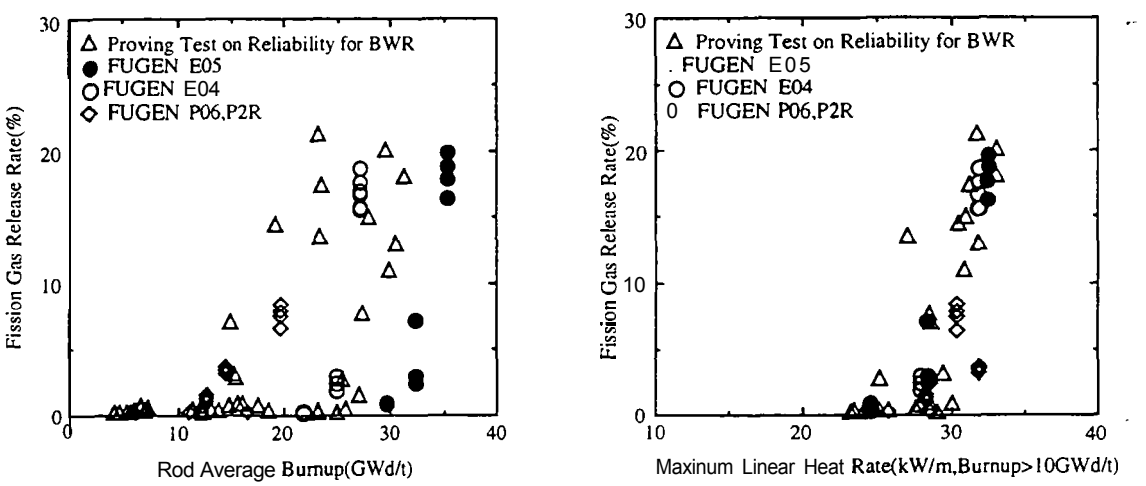

Fig.4 Fission Gas Release Rate As a Function of Burnup and Linear Heat Rate

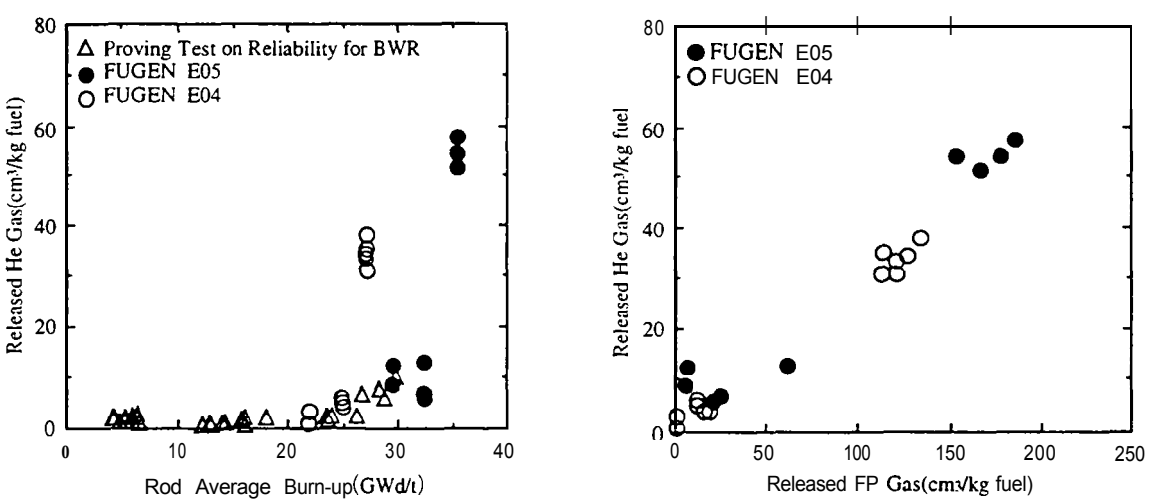

Fig.5 He Gas Release As a Function of Rod Avereage Burnup and Fission Gas Release

3.3 Pellet Microstructure

Fig. 6 shows typical microstructure of a JNC MOX pellet of "Fugen" E04 fuel [2]. Grain growth and fission gas bubble precipitation on grain boundary was observed at pellet center region.

In the pellet peripheral region of the outer ring fuel rod, occurrence of narrow porous band was observed discontinuously. However, no experimental result was obtained through the post irradiation examinations. Also, porous structure of plutonium agglomerates was observed in peripheral region, but no measurable effect of Pu agglomerates was detected on irradiation behavior of MOX pellets. It is partly because Pu concentration in the agglomerates was low in these MOX fuel pellets which were fabricated by MH process.

3.4 Pellet Stability

In Fig. 7, MOX fuel pellet density change during irradiation is plotted as a function of burnup, compared with those of UO_2 fuels irradiated in BWR.

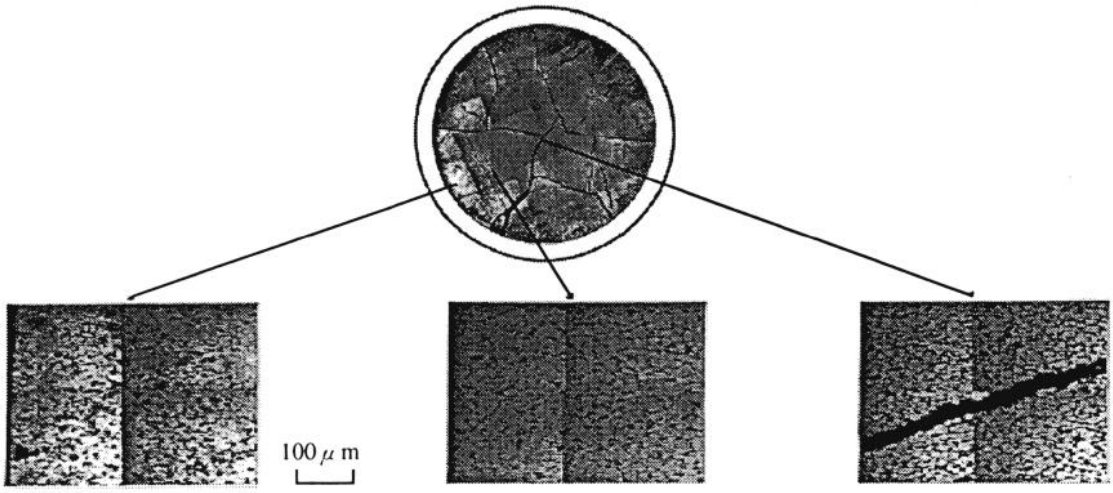

Fig.6 Typical microstructure of a JNC MOX fuel pellet irradiated in Fugen up to a pellet burnup of 30.8GWd/t

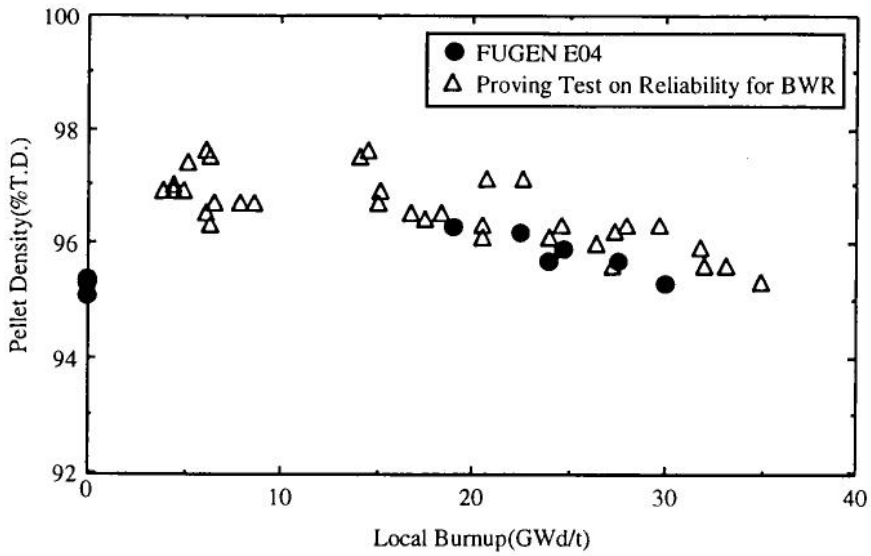

Fig.7 MOX Fuel Pellet Densification and Swelling

Fuel pellet density increases by densification at the beginning of the irradiation, and decreases gradually by swelling as the burnup increases. This behavior of the MOX fuels pellet had good coincidence with that of the UO_2 fuels. That is, pellet stability behavior of the MOX fuel is quite similar to that of UO_2 fuel.

3.5 Cladding Inner Surface Oxidation

It is anticipated that the thickness of oxide layer on cladding inner surface of a MOX rod become larger than in a UO_2 rod at high burnup. It is because Pu atoms generate more noble metal atoms by fission than U atoms and oxygen potential of MOX fuel is higher than that of UO_2. However, as shown in Fig. 6, oxidation of cladding inner surface in a MOX fuel rod of "Fugen" E04 fuel was benign. Relationship between location of corrosion and presence of Pu agglomerate was also vague. These results suggest that oxidation of cladding inner surface of a MOX fuel rod is small and may not be affected by presence of Pu agglomerates in the burnup range up to 35 GWd/t.

3.6 Mechanical Behavior during Power Transient

A series of power ramp test on ATR MOX fuel segments exposed up to 22GWd/t revealed that failure threshold of the MOX fuel was higher than 60kW/m, which was higher than that reported for UO_2 BWR fuel [4, 5] as shown in Fig. 8. ATR MOX fuel rods were also subjected to power cycling irradiation simulating a daily load follow operation. It was confirmed by diameter measurement and fuel instrumentation that cladding deformation by PCMI once occurred at the beginning of the cycling was immediately relaxed and that there was no mechanical effect by repetition of power change.

4. Development of MOX Fuel Rod Performance Code

Along with these irradiation tests, a MOX fuel rod performance evaluation code "FEMAXI-ATR"[6] has been developed to predict thermal and mechanical performances of the MOX fuel rod during irradiation with a reasonable safety margin.

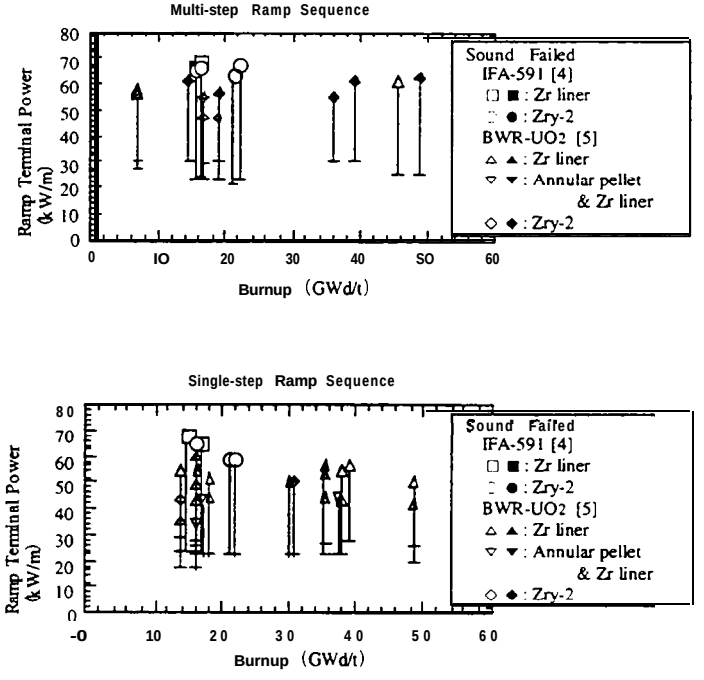

Fig.8 Ramp Tesr Results for ATR MOX (IFA-591) compared with BWR U02 fuel

"FEMAXI-ATR" is based on "FEMAXI-III" code [7] that had enough experience in evaluating UO_2 fuel rod performance. It has fuel material property models and irradiation performance models specific to the MOX fuel. Introduced MOX fuel material property models are melting point, thermal conductivity, thermal expansion factor, Young's modulus, creep rate, and so on. Introduced irradiation performance models are FP gas release rate, Xe to Kr ratio, heat generation distribution in a pellet, which are based on the results of the irradiation tests in Fugen, and so on.

A code verification was done by comparing calculated values of fuel center temperatures, fuel rod pressures, FP gas release rates, fuel cladding diameter changes, etc., with the results of the irradiation tests. Calculated values and measured ones had a good coincidence at a burn-up of up to about 60 GWd/t as shown in Fig.9~14.

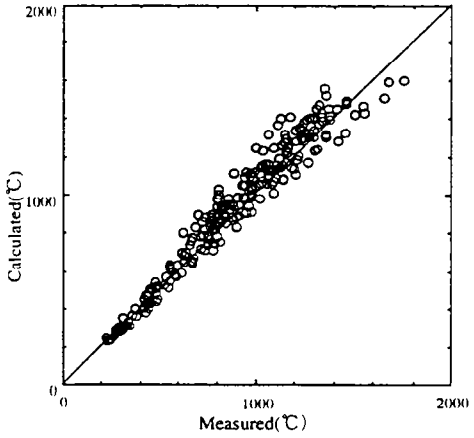

Fig.9 Verification Results of FEMAXI-ATR (Fuel Centerline Temperature)

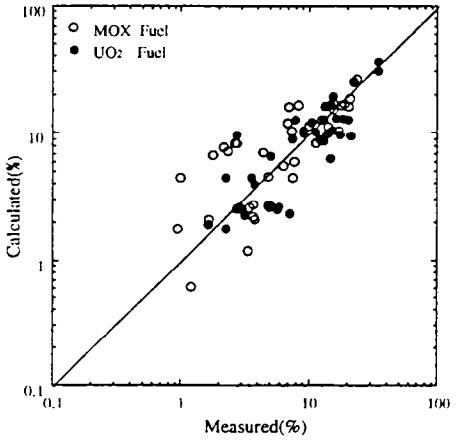

Fig.10 Verification Results of FEMAXI-ATR (Fission Gas Release)

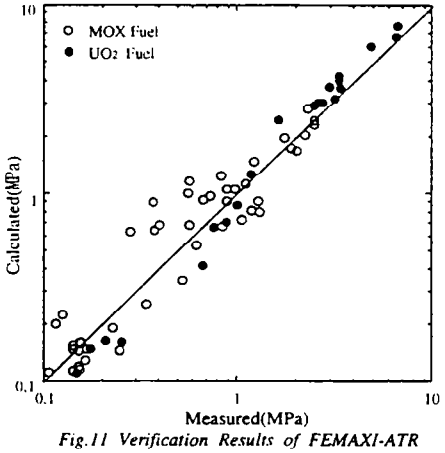

Fig.11 Verification Results of FEMAXI-ATR (Fuel Rod Internal Pressure)

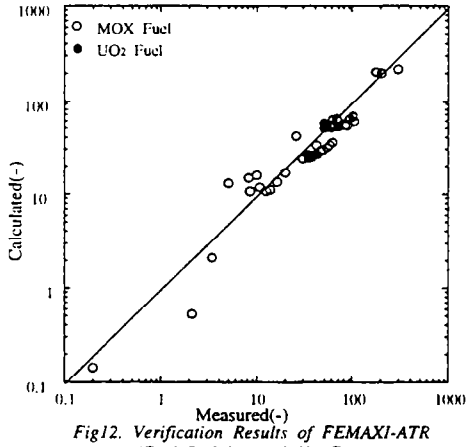

Fig12. Verification Results of FEMAXI-ATR (Fuel Rod Internal He Gas)

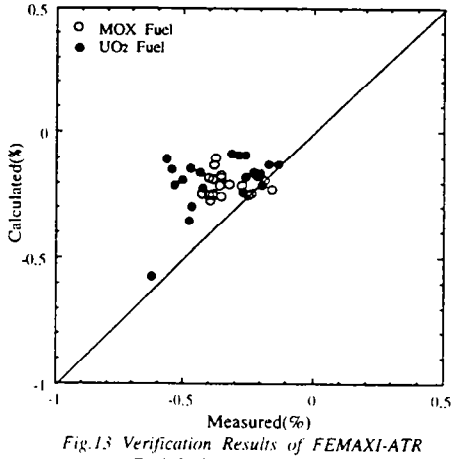

Fig.13 Verification Results of FEMAXI-ATR (Fuel Rod Outer Diameter Change)

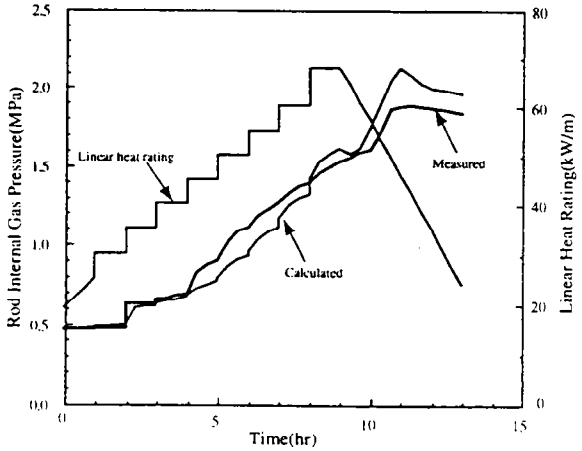

Fig.14 Validation result for rod internal gas pressure of IFA-591-6

"FEMAXI-ATR" code has been already applied to the design analysis of the ATR high burnup MOX fuel (maximum assembly burn-up of 55GWd/t). The structure and main specifications of the ATR high burnup MOX fuel are shown in Fig. 15.

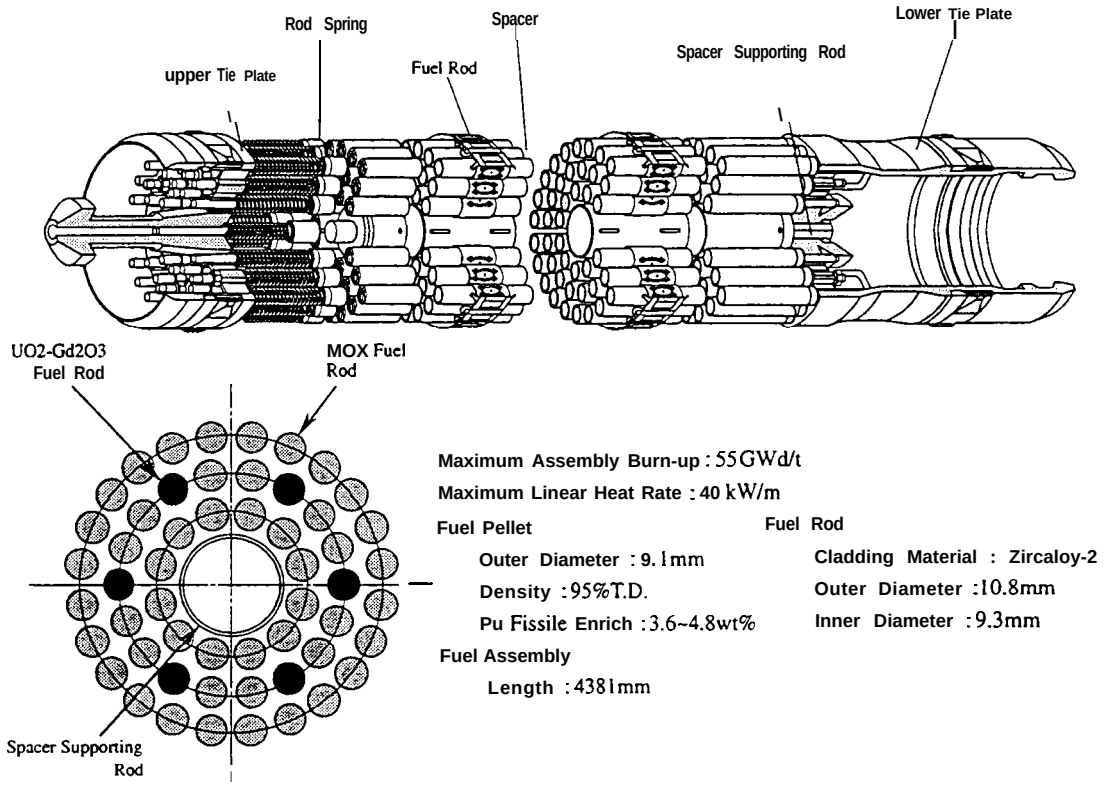

Fig. I5 ATR High Burnup MOX Fuel Assembly

5. Conclusion

JNC accumulated a lot of useful data on the irradiation behavior of the MOX fuel. Comparison of irradiation behavior between JNC MOX and UO_2 fuel shows that there is no distinguishable difference except for helium release, and the excellent homogeneity of JNC MOX fuel was confirmed by the post irradiation examinations.

MOX fuel rod performance evaluation code "FEMAXI-ATR" has been developed. Code verification was done by comparing calculated values with the results of the irradiation tests, and it was verified that "FEMAXI-ATR" can predict thermal and mechanical behavior of the MOX fuel accurately at a burn-up of up to about 60 GWd/t.

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PERFORMANCE OF MOX FUEL: AN OVERVIEW OF THE EXPERIMENTAL PROGRAMME OF THE OECD HALDEN REACTOR PROJECT AND REVIEW OF SELECTED RESULTS

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Abstract

The OECD Halden Reactor Project has defined an extensive experimental programme related to MOX fuels which is being executed with the objective to provide a performance data base similar to that available for UO_2. In addition to utilising fresh MOX fuel and re-instrumented segments from LWR irradiations to high burnup, the concept of inert matrix fuel is being addressed. The irradiation in the Halden reactor is performed in rigs allowing steady state, power ramping and cyclic operation. In-pile data are obtained from instrumentation such as fuel centreline thermocouples, pressure transducers, fuel and cladding elongation detectors, and movable gauges for measuring the diametral deformation. Various phenomena can be assessed in this way, e.g. thermal performance, swelling and densification, PCMI and fission gas release. .The paper describes the objectives of various experiments and provides examples of temperature, pressure and cladding elongation measurements performed on MOX fuel. Salient results are related to the threshold for the onset of significant fission gas release and the relaxation behaviour in a power ramp - PCMI situation.

1. INTRODUCTION

The generation of plutonium is inevitable in present-day reactors which use uranium as basic fuel. Although the idea of utilising this plutonium as initial fuel in fast breeder reactors has so far not been implemented as once conceived, most countries with reprocessing of spent fuel regard plutonium as an asset which for the time being can best be exploited by re-insertion and burning in existing reactors. This is also a viable way of disposing ex-weapon Pu. Mixed oxide (MOX) fuel has therefore received increasing attention within the experimental programme carried out at the Halden Reactor Project. Participants to this programme have expressed a need for establishing a data base on MOX fuel performance similar to that existing for Urania fuel, and the Halden Project has responded by defining an experimental programme to be executed in the Joint Programme of the 1997 - 99 programme period as well as the following 2000 - 2002 period. These programmes address the behaviour of MOX fuel from zero to high burnup (55 - 65 MWd/kg) in conditions of steady state and changing power. In parallel, Halden Project participants pursue their own investigations on a bilateral basis. These also encompass control materials which must be modified as the fraction of MOX in a core increases from the common one third up to whole MOX cores. Results from bilateral studies are often conveyed after some time to the general programme through publications and presentations in Enlarged Halden Programme Group Meetings.

The experimental programme at the Halden Reactor Project is aimed at providing data in support of a mechanistic understanding of phenomena associated with short and long term in-pile fuel performance

and property changes. To this end, irradiation techniques arid instrumentation have been developed [1] and over the years applied to several hundred instrumented fuel rods with numerous fuel types and variants. A considerable performance data base has been built up - mainly related to Urania fuel but in more recent years extended to MOX fuel. The data can be used for fuel behaviour model development and verification as well as in safety analyses and typically encompass fuel temperature, fission gas release, pellet-clad mechanical interaction (PCMI), fuel densification, swelling and creep.

The selected examples from separate effects and integral behaviour studies should give an impression of the experimental capabilities and, related to MOX fuel, elucidate thermal performance, aspects of fission gas release, and PCMI where the data also reflect creep properties. While similarities to Urania fuel properties are evident, there are also substantial differences which have to be incorporated in fuel behaviour models and codes.

2. EXPERIMENTAL CAPABILITIES

Investigations of fuel performance parameters, especially at high burnup, have to deal with a number of experimental problems, i.e. the time required for burnup accumulation, the demand on instrumentation to function reliably for the long time of in-core service, and the need for a separation of an increasing number of phenomena. The Halden Project has developed and applied techniques which make it possible to obtain reliable data for all relevant burnups, from beginning-of-life to ultra high exposure reaching 100 MWd/kgUO_2. The re-instrumentation of pre-irradiated fuel segments is of particular importance in this context [1].

While PIE ascertains the state at the end of irradiation, in-core instrumentation provides a full description of performance history, cross correlation between performance parameters, on-line monitoring of the status of the test, and direct comparison between different fuels and materials. Trends developing over several years, slow changes occurring on a scale of days or weeks, and transients from seconds to some hours can be captured by the same instrumentation. The availability of reliable instruments and irradiation rigs is therefore essential for fuel behaviour studies regardless of fuel type.

Data on fuel performance are typically obtained from:

- *fuel thermocouples* or *expansion thermometers,* which measure the fuel centre temperature from which also long-term changes such as conductivity degradation of the fuel can be deduced;
- *bellows pressure transducers,* which provide data on rod inner pressure and fission gas release;
- *fuel stack elongation detectors,* with which densification and swelling behaviour can be assessed;
- *cladding elongation detectors,* which provide data on the onset and amount of pellet - cladding interaction, permanent deformation, relaxation capabilities of fuel and cladding as a function of power and burnup, and even swelling of the fuel;
- *cladding diameter gauge* to determine radial deformations as a function of power, holding time after ramping, and burnup.

Fuel thermocouples, pressure transducers and cladding elongation detectors can be utilised for re-instrumentation of fuel segments irradiated in other reactors, often commercial LWRs. This technique has been applied in a number of tests and become more and more important for the investigation of high

burnup fuel performance. Three experiments currently being executed within the Halden Reactor Project experimental programme contain pre-irradiated, re-instrumented commercial MOX fuel.

The irradiation of instrumented fuel rods is carried out in specialised rigs according to test objectives, e.g. long term base irradiation, diameter measurements, ramp and overpower testing, load follow and automatic frequency control, and others. The MOX fuel testing at the Halden Project employs most of the above mentioned instrumentation and rig types. Steady state data are supplemented by information obtained from noise analyses of fuel thermocouples and cladding elongation signals. In addition, gas flow and hydraulic diameter measurements have recently been carried out on high burnup MOX fuel in a specialised rig designed for studying rod overpressure / clad lift-off under PWR conditions.

3. EXPERIMENTAL PROGRAMMES RELATED TO MOX FUEL

MOX fuel differs from Urania fuel on several counts related to nuclear and physical properties. Fuel temperatures are influenced by a somewhat lower conductivity, a more peripheral power generation due to more pronounced nuclear self shielding and, via gap conductance, a higher thermal expansion. The latter also influences PCMI behaviour as do better creep properties compared to Urania fuel. Fission gas release is inherently more complex in MOX fuels and may appear to be larger due to higher reactivity (higher power) at end-of-life for the same burnup as urania fuel.

These differences require attention and must be studied in experimental programmes. MOX fuel has therefore been tested in the Halden reactor on a bilateral basis for many years, and a series of MOX-related experiments is also being executed in the present joint programme. The synergism of the joint and bilateral programmes, also involving information exchange in Enlarged Halden Programme Group Meetings, contributes to an amplification of the benefits that members obtain from participation in the Halden Reactor Project.

In addition to participants' bilateral activities, an extensive experimental programme related to MOX fuel has been defined and is currently being executed as part of the joint programme of the Halden Project. It responds to the need expressed by participants that a database on MOX fuel performance be built up, similar to that accumulated over the years for Urania fuel. This programme draws on the general Halden Project testing and instrumentation experience and in particular on the re-instrumentation techniques which have been developed and applied to numerous rods of bilateral and joint tests.

The scope of the overall joint programme MOX testing includes:

- obtain data on basic thermal performance from low to high burnup, including assessments of changes of conductivity;
- assess fission gas release and release kinetics;
- derive information on fuel swelling and densification through evaluation of temperature data and pressure changes as a function of burnup;
- obtain data on PCMI behaviour and fuel relaxation capabilities;
- explore the rod over-pressure / clad lift-off effect for high burnup fuel
- produce high burnup (> 65 MWd/kg) MOX fuel through continued irradiation in the Halden reactor under PWR conditions and provide performance data (temperature, fission gas release, PCMI) for this high burnup;

- assess the in-core behaviour of fuel where plutonium ‘is carried in an inert matrix thus avoiding the generation of new Pu and allowing a more complete burning.

The following examples of experimental results elucidate some of the phenomena listed above. Although they are grouped under separate headings (thermal behaviour, fission gas release, PCMI), it must be kept in mind that the different phenomena are interlinked. Experiments with more than one type of instrumentation which allow the simultaneous measurement of e.g., temperature and rod pressure, are therefore of increased value. Most of the experiments presented below are of this type.

Thermal behaviour of MOX fuel

A good knowledge of the temperature distribution in a fuel pellet is essential for the prediction of fuel behaviour since most phenomena depend on temperature. The experience gained with standard urania fuel can to a certain extent be applied to MOX fuel as well. For example, it can be assumed that factors with a secondary influence on temperatures such as the slightly larger thermal expansion or the more peripheral power generation of MOX fuel are covered sufficiently through standard modelling. The more important thermal conductivity and in particular the change with burnup, however, need to be addressed with in-pile experiments. The inhomogeneous distribution of fissions may produce conductivity changes which differ from the development seen in Urania fuel. For the time being, the burnup dependent correlation derived for urania [2] is used at the Halden Project for MOX fuel as well, modified with a factor which reduces the conductivity by 8% as a recommended value. However, reductions of up to 20% have been seen in some experiments.

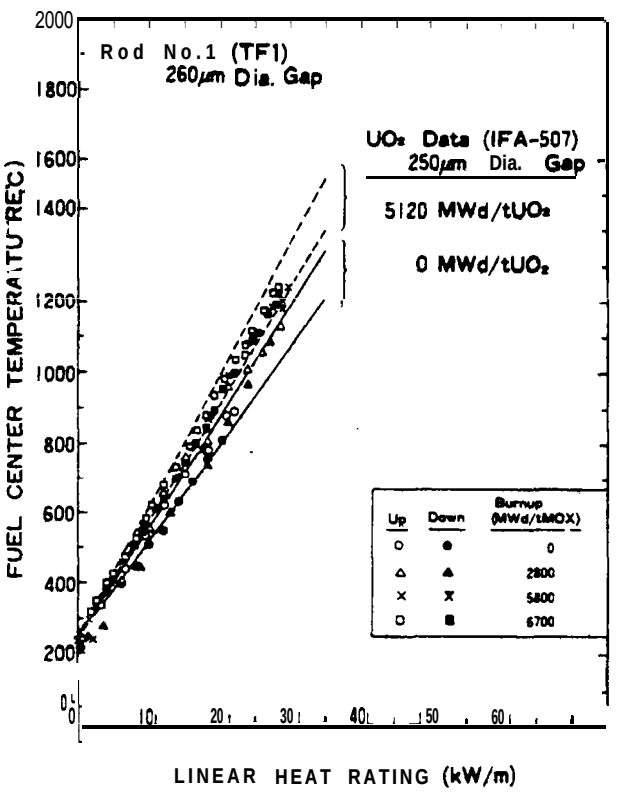

Fig. 1 Comparison of MOX and UO_2 fuel temperatures

The MOX fuel centreline temperatures shown in Fig. 1 originate from experiments (lFA-514 and IFA-529) originally conducted on a bilateral basis but also presented to joint programme participants. Extensive instrumentation (fuel thermocouples, fuel stack elongation detectors, cladding elongation detectors, rod pressure sensor) were employed and provided detailed data on fuel behaviour [3]. For burnups of 0 and 6.7 MWd/kg, the comparison with Urania fuel of similar design indicates somewhat higher temperatures in the MOX fuel.

Fission gas release

Fission gas release from MOX fuel with Pu agglomerates can be expected to differ from urania fuel because of two effects: the concentration of fissions (or burnup) in the agglomerates and the potential for higher end-of-life power and thus temperature of MOX fuel. In the experimental programme of the Halden Project, the temperature of onset of significant

fission gas release (> 1%) as a function of exposure and the release kinetics are being addressed. As an example, the in-pile pressure measurements performed in IFA-514 and IFA-529 showed that the empirical threshold of significant fission gas release (> 1%) derived from UO_2 fuel applies well to MOX fuel [4]. At around 20 MWd/kg, the fuel in these experiments was subjected to power cycling tests to simulate load following. During this time, measurements of rod internal pressure indicated that the release of fission gas was controlled by a diffusion mechanism and that there was no apparent increase in the release rate.

In a joint programme experiment which started with fresh MOX fuel, temperature and rod pressure are measured in two rodlets, one with hollow and one with solid pellet fuel. The objectives of this experiment are primarily to obtain temperature data as a function of burnup (up to 60 MWd/kg), and to establish the fission gas release threshold at various exposures. The latter is effected by temporary power increases and monitoring of the pressure response and the fuel temperatures. The result of an uprating at about 10 MWd/kgMOX is shown in Fig. 2. A definite pressure increase (gas release) is observed during the period at highest power.

The temperature of release onset related to the data of Fig. 2 is very similar to the threshold temperature of urania fuel. This is depicted in Fig. 3 together with the results of two other experiments. There is, however, an important difference to urania fuel. As can be seen from Fig. 2, the release seems to stop after returning to lower power, but the long-term development not presented here showed that the release, once triggered, continued even when temperatures were kept well below the onset level. This behaviour differs from experience with urania fuel where a re-sintering and discontinuation of fission gas release has been observed when temperature is kept below the release threshold for extended periods [5].

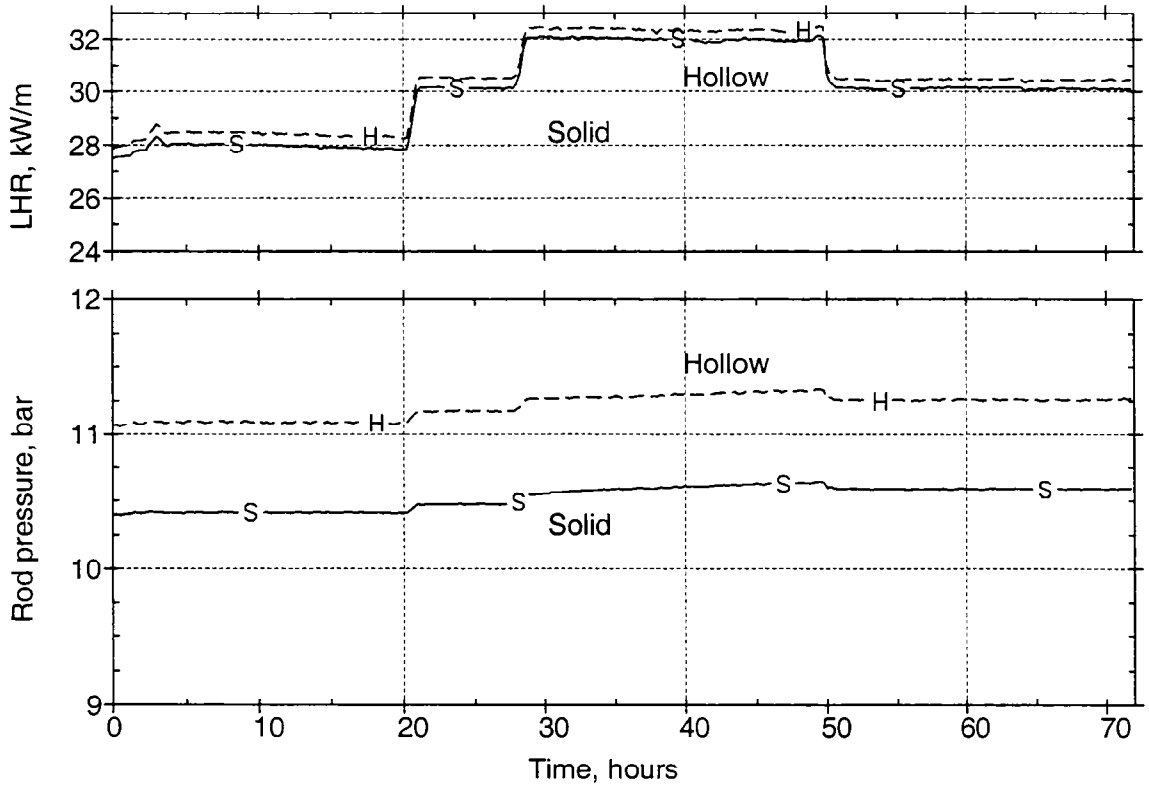

Fig. 2 Response of internal pressure to power uprating at 10 MWd/kg

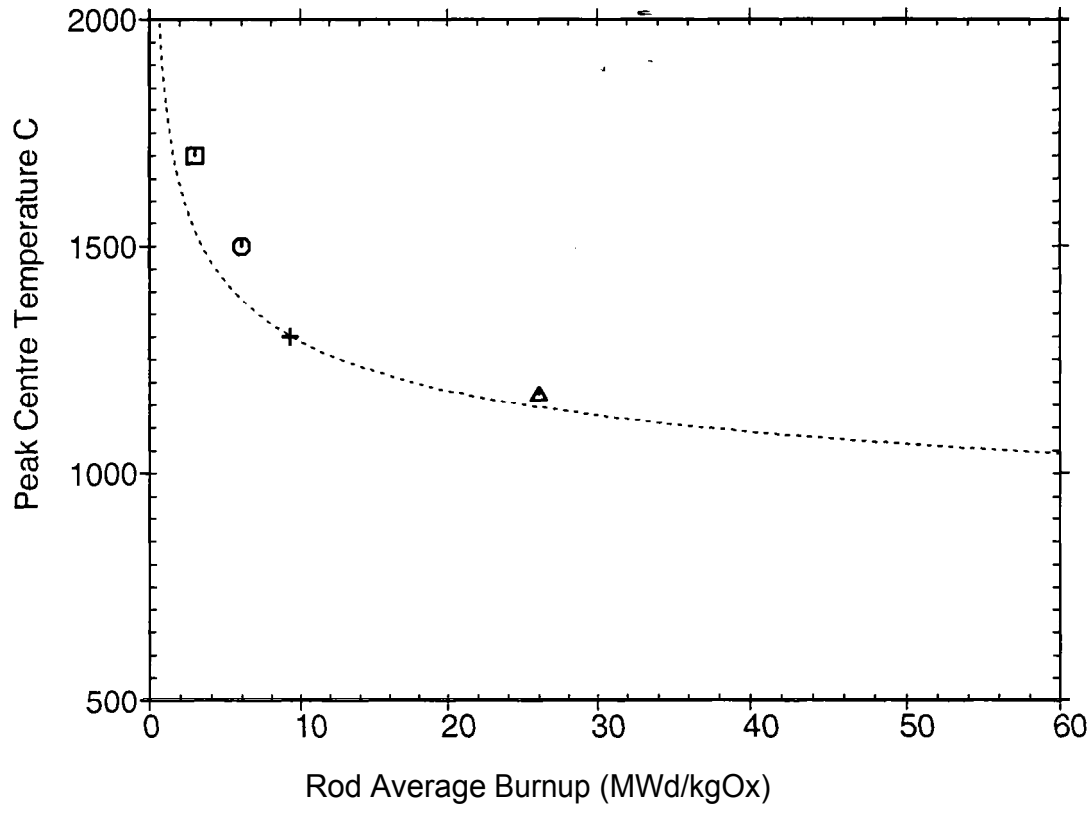

Fig. 3 UO_2 release threshold as a function of burnup and comparison with release onset of MOX fuel

Mechanical response, PCMI

An excellent example of how experimental techniques and the cross-correlation of phenomena through instrumentation combine to yield a more complete assessment of phenomena, is given with a test series that PNC (Power Reactor and Nuclear Development Corporation, Japan) has conducted in the HBWR. The objective of the tests was to study the Advanced Thermal Reactor (ATR) MOX fuel behaviour during transient operation and to determine the failure threshold [6]. To this end, fuel segments previously irradiated in the Fugen reactor were ramped in the HBWR using a ramp rig with He-3 power control. The instrumentation consisted of either cladding elongation detectors or rod pressure transducers.

The ramp rig allows various modes of power increases: continuous and gradual, staircase, and ramping with maximum speed. Ramps can be accurately repeated since the coil depressurisation/pressurisation is supported by a PLS control. Fig. 4 and 5 show the cladding elongation and rod pressure response to a staircase power increase [6]. The excellent relaxation behaviour of the MOX fuel during the 1 hour holding periods is evident. From the rod pressure measurements, the onset of fission gas release can be determined. An interesting point to note is an additional pressure increase during the final power decrease at about 9 hours into the transient. At the same time into the transient, the sibling segment equipped with a cladding elongation detector reveals a transition from fuel-cladding contact to free thermal expansion (contraction) of the cladding. The open gap then provides a pathway for released fission gas to the plenum. This phenomenon is often observed in conjunction with high burnup fuel where the gap is closed already at much lower powers than reached during the ATR ramp test.

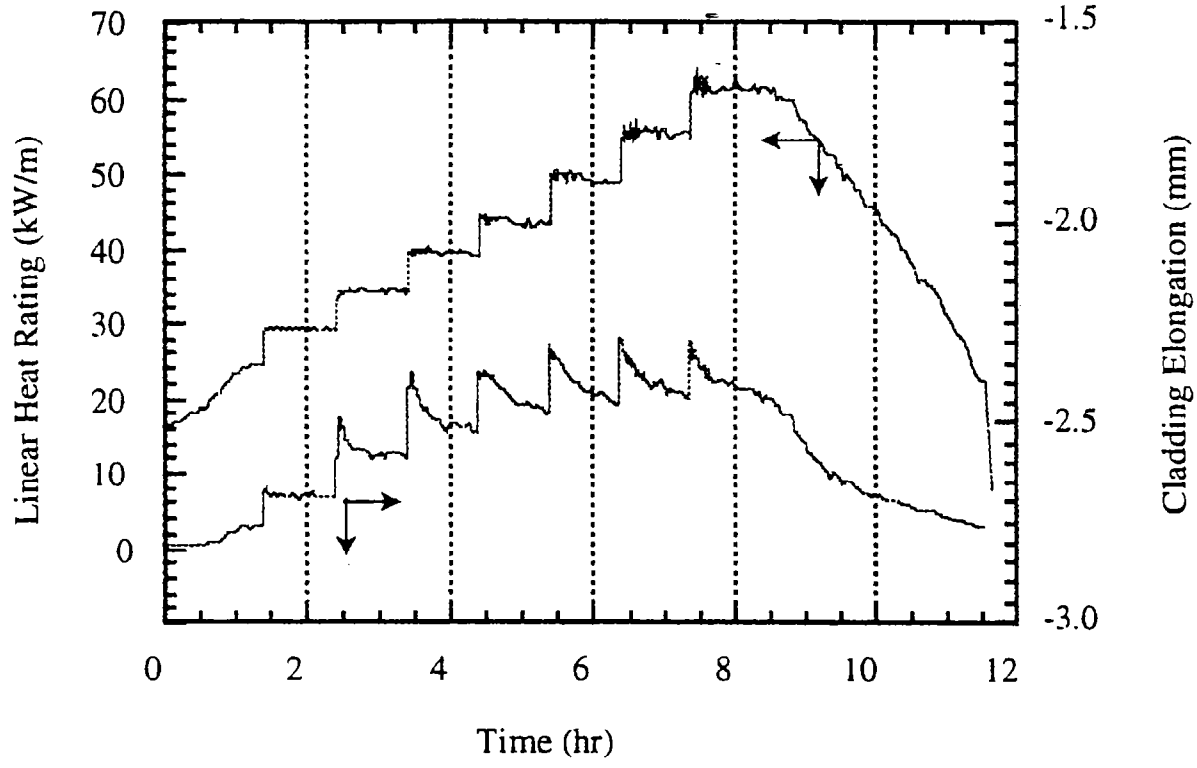

Fig. 4 Linear heat rating and cladding elongation of IFA-591 segment during ramp testing

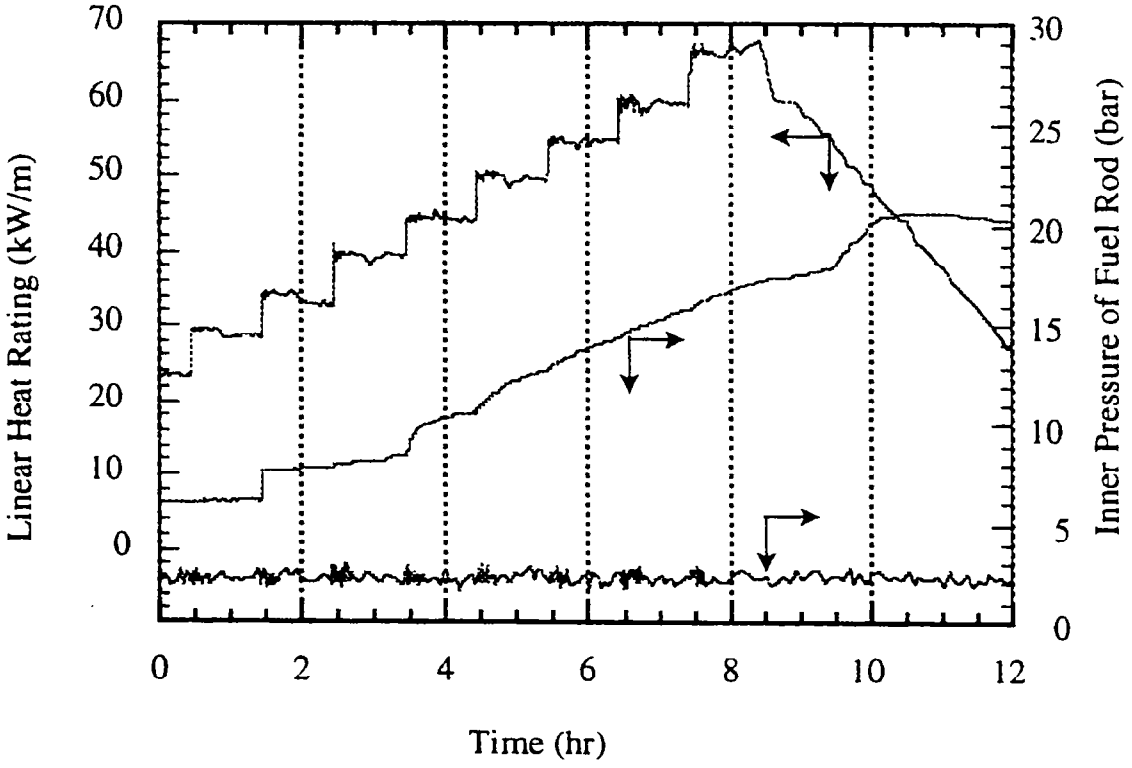

Fig. 5 Linear heat rating and rod pressure of IFA-591 segment during ramp testing

Re-instrumented MOX fuel segments are utilised in another experiment starting with a burnup of about 27 MWd/kg. This fuel had negligible fission gas release during irradiation in the commercial PWR and the objective of the experiment is to explore the release onset together with obtaining temperature and PCMI data. To this end, the power has been uprated with intermediate reductions to a base level as shown in Fig. 6. The upper part of Fig. 6 shows the cladding elongation response to the power changes and during the holding periods. Although the relaxation is not as pronounced as for the ATR fuel shown previously, it is still stronger than that of urania fuel. The comparison with other Halden Project experimental data indicates a MOX creep rate that is a factor of about 8 larger than that of UO_2 [7].

An interesting detail can be seen in association with the power cycle performed at about 4.5 days. The cladding elongation at the maximum of the cycle is slightly larger than at the end of the preceding holding period, indicating a PCMI racheting effect which, however, did not produce any discernible permanent strain. The 20% power change (30 - 24 - 30) is similar to the load follow range of a commercial reactor and it will be of interest to study the combination of relaxation/racheting during repeated cycles.

Miscellaneous tests

The *rod overpressure / clad lift-off* test is an ongoing experiment utilising re-instrumented MOX fuel pre-irradiated in a commercial PWR to an exposure of 55 MWd/kg. The objective of the test is to establish the overpressure that would lead to an opening of the fuel-clad gap, i.e. more cladding creep-out than fuel swelling and thus increasing fuel temperatures. The overpressure in this type of experiment is applied through an external pressurisation system which can produce overpressures of up to 450 bar above the PWR loop pressure of 160 bar. A similar experiment was successfully executed from July

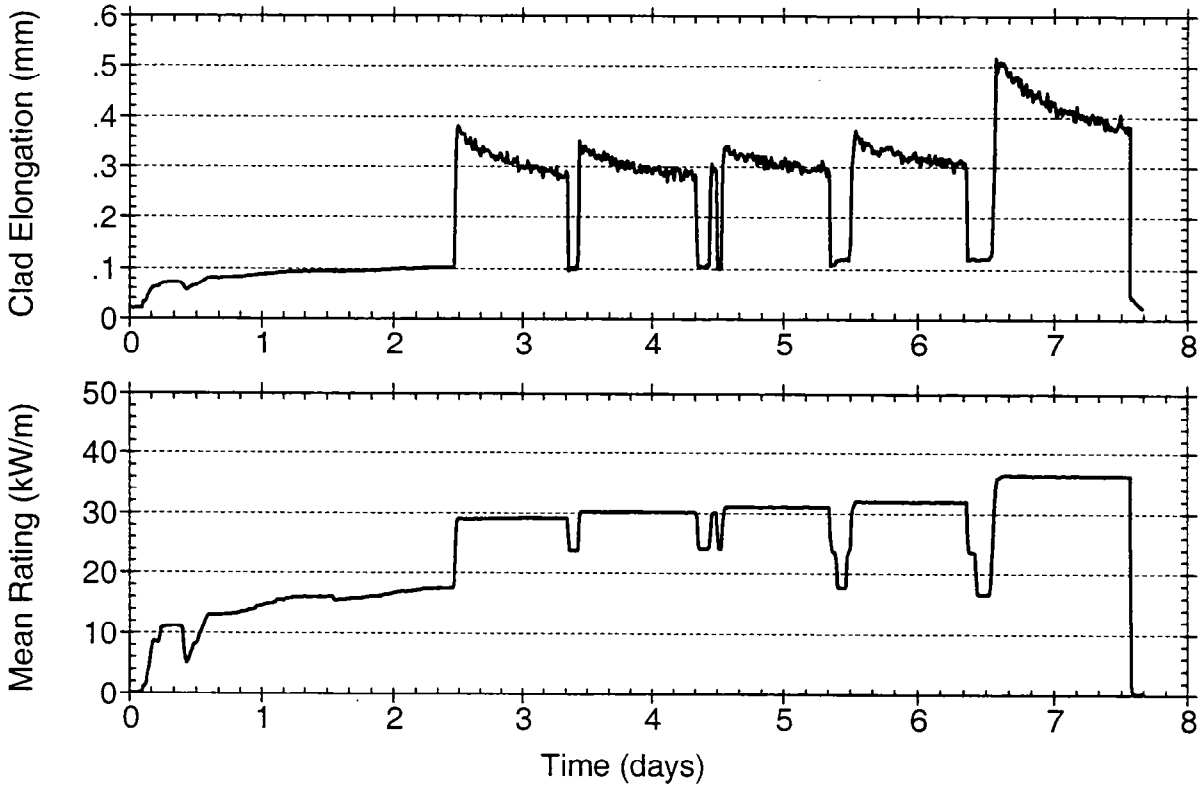

Fig. 6 Cladding elongation response to staircase power increase

1997 to May 1998 using UO2 fuel. The onset of clad lift-off was determined by monitoring the change of the fuel centre temperature at constant power over several periods (500 - 1000 hours each) with varying levels of overpressure. It seems that the currently investigated MOX fuel can endure a higher overpressure than UO_2 fuel in agreement with the superior creep properties of MOX fuel.

Halden Project Participants have expressed the need for studying MOX fuel behaviour at burnups currently not being reached in commercial nuclear power stations, but for which license applications are envisaged in the future. In order to extend the burnup of the four-cycle MOX fuel to at least 65 MWd/kg, a burnup extension rig has been designed that provides the required nuclear and thermal-hydraulic (PWR) conditions. The design includes in-core connectors for attaching the signal cables of re-instrumented fuel thermocouples and LVDTs for cladding elongation and rod pressure measurements. In this way, fuel performance data are already being obtained during the burnup extension period.

After burnup extension, a power uprating test of the fuel - conducted in the same manner as for the medium burnup MOX - will provide the basis for the following comparisons and assessments:

- Fuel thermal performance at -27, -55 and 65 MWd/kg, giving the basis for MOX fuel conductivity degradation at increasing burnup.
- Fission gas release onset at the burnup levels 27 and 65 MWd/kg.
- PCMI behaviour at the two burnup levels.

Finally, an experiment related to the burning of plutonium in an inert carrier matrix is being prepared in collaboration with organisations participating in the Halden Project. This experiment will also contain standard MOX for comparison. The foreseen instrumentation will allow measuring of temperature, pressure and dimensional changes of the fuel stack. A more detailed description is given elsewhere in this meeting [8].

The experiments described above together with parallel investigations of UO_2 fuel constitute an extensive experimental programme with the objective to establish a database of qualified high burnup data for fuel behaviour model development and validation as well as for application in safety assessments. The related work will be carried out not only in the present 3-years programme period from 1997 - 99, but also in the next period from 2000 - 2002 which is currently being defined in discussions with participants.

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HELIUM GENERATION AND RELEASE IN MOX FUELS

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Abstract

Measurement of the retained helium in MOX fuel matrices was carried out. Helium generation under irradiation was estimated by adding the released helium to the retained value. The measured helium generation agreed with the calculated one, when modified by adding ternary fission yield.

Helium pressure increase in a high burn-up MOX fuel rod can reach 30% or more of the FP gas pressure. This should contribute to higher internal rod pressure. The impact of generated helium on fuel performance requires further investigation.

1. INTRODUCTION

Helium is generated in fuel pellet matrices by alpha decay of trans-uranium nuclides, such as the Cm-242, (n,alpha) reaction, and ternary fission. The first one is the major source. Therefore, helium accumulation in the matrices, especially in MOX fuels, increases with the burnup, not linearly but exponentially. Helium, accumulated in fuel pellet matrices, is released into the free volume of the fuel rod and it increases the rod inner pressure along with the released fission product (FP) gas, when the fuel temperature rises.

Inner-pressure of a BWR fuel rod is one of the most important factors that restricts the life time of a fuel rod, and the release behavior of FP gas such as xenon and krypton is being studied widely. The volume fractions of helium in the total gas released into the free volume were reported to exceed over 30% in MOX fuels[1]. This is too large to neglect the effects on the mechanical design of MOX fuels. Therefore, it is important to clarify the release mechanism. The amount of helium released should be related to the generation rate and its release mechanism. However, there are few studies on helium release behavior, especially on its generation. The uncertainty on the calculated helium yield is quite large because of the complex reaction chains, the effect of operating history, and changes in void fractions of coolant during operation.

In this paper, as a first step to study the helium release mechanism, the reliability of helium generation calculations was evaluated. Helium volumes retained in pellets

were measured for samples from an MOX and a UO_2 fuel rod. Then helium behavior is discussed on the basis of data obtained from the puncture test, etc.

2. TEST CONDITIONS

2.1 Test Samples

The MOX fuel rod were irradiated to 30.4 GWd/t in Tsuruga Unit 1 and the UO_2 rod to 36.5 GWd/t in Fukushima Daini Unit 2. Those fuels had already been subjected to post irradiation examinations (PIES) including pin puncture test, before the present investigation. As shown Table 1 , the FP gas release rate was about 8 % for the MOX fuel rod, and 3.5 % for the UO_2 fuel rod.

Table 1 Characteristics of Test Fuel Rods

Fuel type	Pu enrich wt%	Burn-up GWd/t	FP gas release %
MOX	6.2	30.4	8
UO_2	–	36.5	3.5

Six samples were selected for the retained helium and FP gas analyses from the PIE archives. Those samples were localized at different levels from bottom to top of the fuel column of each fuel rod. As shown in Table 2, three samples were taken in 6.2 %Pu MOX fuel and three others in UO_2. The samples were irradiated locally up to 42 GWd/t. For each analysis, 2 to 4 single pellets were used.

2.2 Apparatus

Helium and FP gas remaining in the pellets were collected according to the following procedure. A retained gas collection apparatus which included pellet dissolution and gas collection parts, was set up in the NFD hot cell. The apparatus consisted of a flask, reserve tank, compressor pump, and gas sampling chamber. The flask was used for dissolving fuels, and was equipped with a vapor condenser to return boiling vapor to the solution. After precisely weighed specimens were put into the flask, the system was evacuated below 500 Pa. 100 ml 8M nitric acid was poured into the flask, and the sample was heated for over 2 hours to dissolve it. Nitric acid with 0.02M HF was used for the MOX fuel. To reduce inner pressure induced by NOx originating from the reaction between the fuel and nitric acid, H_2O_2 was blended into the solution. The system was kept below atmospheric pressure in order to prevent gas leakage. All gases released during the dissolution process were collected using the compressor pump (Toepler pump, etc.) in the apparatus. The gas stream was purified by cold trapping (-80 deg. C) and soda-lime absorbers were used to remove water and nitric oxides, before being transferred into the gas sampling chamber filled with a definite amount of standard composition gas. The standard gas was composed isotopes of helium, xenon and krypton. Hardly any He-3, Kr-78 and Xe-129 are found in fission product gases, so they were used for standard to estimate volumes of each isotope in the collected gas. The isotopic compositions of an allotment of the

Table 2 Sampling Matrix

Fuel type	Sample No.	Local burnup GWd/t
MOX	M1	25
	M2	31
	M3	36
U1	28	
U2	36	
U3	42	

mixture gas were analyzed with gas mass spectrometry and each gas volume retained in the pellets was evaluated. The evolution of the FP gas was followed by measuring the activity of the amount of Kr-85 passed through the cell ventilation duct when the collected gas was released after the measurement.

2.3 Calculation of Helium Generation

The primary sources of helium generation during irradiation of oxide fuel are (1) alpha decay of trans-uranium elements, primarily Cm-242, (2) alpha particles formed by ternary fission, and (3) (n,alpha) reaction between O-16 and fast neutrons. The (n,alpha) reaction is the dominant source of helium generation in low burn-up UO_2 fuel loaded in commercial BWRs, and the same phenomenon occurs in MOX. The ternary fission has a yield of 0.2 to 0.3% helium atoms per fission in both U and Pu[2]. All of the trans-uranium elements except Np-239 and Am-242 undergo some alpha decay. But most of the elements have a long half-life, so about 90% of the alpha decay of trans-uranium elements would come from Cm-242, which has a 163-day half-life.

Table 3 Calculated Helium Generation

Fuel type	Sample No.	Local burnup (GWd/t)	Helium generation (cm^3/g-fuel)
MOX	M1	25	0.06
	M2	31	0.08
	M3	36	0.10
UO2	U1	28	0.02
	U2	36	0.02
	U3	42	0.026

In the calculation with the ORIGEN code[3], reaction yields were corrected for the neutron spectrum of a Japanese typical commercial BWR. The ORIGEN code calculates helium generation by alpha decay and the (n,alpha) reaction. The results of the ORIGEN calculation were modified by adding ternary fission yield, estimated by using the fission yield compiled by Rider[2]. The ternary fission yield contributes about 10% to total helium generation.

Table 3 presents the results of the calculated helium generation of the MOX and UO_2 fuel. The calculated helium production of the MOX fuel is about 4 times that of the UO_2 fuel because of shorter chain to transpose plutonium to Cm-242 than for uranium.

3. RESULTS

3.1 Fuel Dissolution

After the sample has been put into the flask, the system was evacuated, and nitric acid was poured into the flask after the system pressure was below 500 Pa. The solution was kept at room temperature in the first step of dissolution.

For the UO_2 fuel, no bubbles which form by reaction between fuel and nitric acid in dissolving process were observed. After the start of heating, bubble formation on the fuel surface was observed when solution temperature reached 40 deg.C. After that, solution temperature exceeded the heating temperature, and spontaneously increased when the heater was turned off. Solution temperature reached 60 deg.C 30 minutes from the starting of the temperature increase, and then began to decrease. Solution temperature decreased to room temperature after 2 to 4 hours from the start of the dissolution process. After the solution had cooled, it was reheated for 2 hours to make

sure the sample was completely dissolved. The solution was kept just the below boiling temperature in order to keep inner pressure below atmospheric pressure.

For the MOX fuel, immediately after adding the nitric acid, bubble formation was observed at room temperature and solution temperature spontaneously increased to 55 deg.C. after 30 minutes. After the solution had cooled, it was reheated for 2 hours.

Table4 Residual FP gas and Helium

Sample No.	Residual gas volume(cm^3/g-fuel)		
	Xe	Kr	He
M1	0.333	0.016	0.017
M2	0.402	0.023	0.026
M3	0.560	0.025	0.038
U1	0.661	0.070	0.009
U2	0.995	0.124	0.009
U3	0.683	0.049	0.011

After the dissolution process, the solution was filtered and the remaining residue was dried and weighed. The weight of the residue was about 4% of the fuel. This value was not different from the predicted weight of fission products.

3.2 Residual FP Gases and Helium

Table 4 presents the results of the recovered amounts of FP gas and helium retained in the fuel. The Xe/Kr ratio of residual FP gas of the UO_2 fuel rod gives a mean result of 9.6 which is not very different from 8.5 obtained by the free FP gas determination of the same fuel rod in the puncture test. The Xe/Kr ratio of the MOX gives a mean result of 20.2 which is larger than 13.8 obtained in the puncture test. Kr-85/Kr ratio of the MOX fuel gives a mean result of 0.039 which is about 35 % less than the value of 0.06 obtained in the puncture test. It must be taken into account that the residual FP gas composition determination was carried out a long time after the puncture test and that Kr-85 has a half-life of about 10.3 years. All the released FP gas during the MOX fuel dissolution was followed by Kr-85 activity measurement and estimated as 0.45cm^3/g-fuel. The obtained residual fission gas (xenon and krypton) volumes of the MOX fuel spread from 0.349cm^3/g-fuel to 0.585cm^3/g-fuel, but a mean result of 0.467 agrees well with the total released gas volume.

Table 4 Residual FP Gas and Helium

4. DISCUSSION

4.1 FP Gas Generation

Table 5 presents the amounts of FP gas such as xenon and krypton generated in the MOX fuel. The measured total FP gas generation was got by adding the measured average released volume (the puncture test results) to the measured local retained volume (the results in this work). The average released FP gas obtained by the puncture test is shown in Table 6. The measured total FP gas generation of the MOX fuel samples was lower than the predicted value by the ORIGEN code. However, the MOX specimens were kept in sealed capsules for a long time after their preparation, and additional gas releases were

Table 5 Comparison between Measured and Calculated FP Gas Generation

Sample No.	Local burnup (GWd/t)	FP gas generation (cm^3/g-fuel)	
		measured	calculated
M1	25	0.41	0.62
M2	31	0.49	0.77
M3	36	0.65	0.90

detected when the specimens were taken from the capsules. The additional gas release can be explained by the following consideration.

Immediately after the irradiation, the generated gas (excluding the released part) might be present in both the matrices and bubbles on grain boundaries. The residual gas of the MOX fuel measured in this work might be mainly correspond to that remaining in the fuel matrices in which the gas atoms could not move during long term storage. Therefore, the additionally released gases of the MOX fuel might originate from grain boundary bubbles and be released through splits such as cracks which were caused by handling during the long period post-PIE storage.

Table 6 Average Released Gases

Fuel type	Sample No.	Local burnup (GWd/t)	Gas release(cm^3/g-fuel)	
			FP gas	Helium
MOX	M1	25	0.06	0.03
	M2	31		
	M3	36		
UO2	U1	28	0.03	0.01
	U2	36		
	U3	42		

The volume of additional gas release measured by Kr-85 radioactivity when the capsule was opened was estimated as 0.23cm^3/g-fuel. This is equivalent to 30% of the calculated generation. The corrected FP gas generation in the MOX fuel samples with the additional gas release, are almost equal to the calculated one.

By adding the released FP gas fraction at the puncture test and the additionally released one during its long period storage after the puncture test, the above mentioned presumption leads to the conclusion that about 40% of the FP gas generated accumulated in the grain boundaries.

Because of the freshness of the UO_2 sample in this work, the retained FP gas fraction probably includes amounts remaining both in the matrices and on the grain boundaries. Because of the similarity of irradiation conditions between the MOX and the UO_2 fuel rods, it can be presumed that the fraction of the FP gas on the grain boundaries in the UO_2 was about 40% of generation, too.

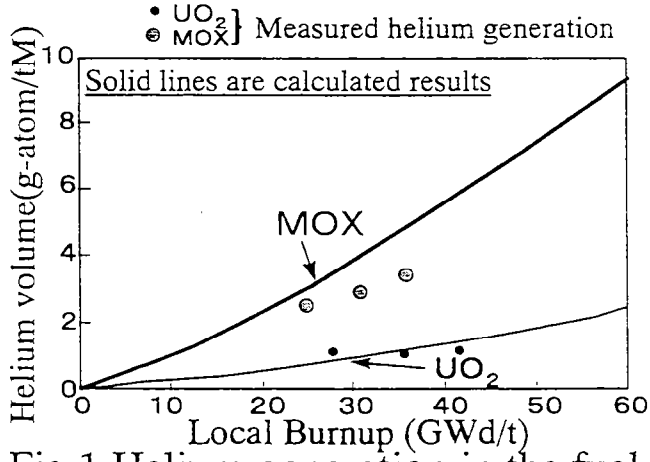

Fig.1 Helium generation in the fuel

4.2 Helium Generation

The measured and calculated helium generation can be compared in Table 7 and Fig. 1. The total generated gas volumes of helium estimated in a similar way to the FP gas generation, coincided with the values calculated by the ORIGEN code for the UO_2 fuel rod. The results for the UO_2 fuel show that the helium generation has a tendency to be underestimated with increasing burn-up. It must be taken into account that the helium

Table 7 Comparison Between Measured and Calculated Helium Generation

Sample No.	Local burnup (GWd/t)	Helium generation (cm^3/g-fuel)	
		measured	calculated
M1	25	0.05	0.06
M2	31	0.06	0.08
M3	36	0.07	0.10
U1	28	0.02	0.02
U2	36	0.02	0.02
U3	42	0.02	0.02

generation was determined by adding the measured local retained helium to the average released helium, and the local release at low burn-up (low power, low fuel temperature and low generation) position would be expected to be lower than the average.

A discrepancy is noted between the measurement and calculation for the MOX fuel samples. However, as mentioned above, additional gas releases of about 30% were detected when the MOX specimens were taken from the sealed capsules after the long time storage.

The helium generation corrected by the additional gas release, was almost equal to the calculated one. The proportions of helium generation to the FP gas generation of MOX and UO_2 are 0.1 and 0.025, respectively.

Table 8 Helium Release Rate

Fuel type	Sample No.	Helium release(%)	
		Average	Local
MOX	M1	40	71
	M2		67
	M3		60
UO2	U1	50	52
	U2		52
	U3		58

The average helium release rate obtained by the puncture test and the local helium release rate measured in this work can be compared in Table 8. The average helium release rate was estimated by dividing the released helium volume obtained from the puncture test by the calculated generation. The local helium release rate was estimated by subtracting the measured local retention from the calculated local generation.

The local helium release rate scarcely depends on the local burn-up (and maybe temperature). The local release rate (measured in this work) of the UO_2 fuel agrees well with the average helium release rate (measured in the puncture test). However the local helium release rate of MOX fuel is about 20 to 30% higher than the average helium release rate. The differences in the helium release rate between the average and the local of the MOX fuel can be explained by the above mentioned additional release.

Notice must be taken of the similarity in the helium release rate in the puncture test between MOX and UO_2. Those values are almost the same as the fraction of FP gas collected on the grain boundaries, too. Those similarities may suggest that the released helium originates from the collected FP gas area.

4.3 Helium Release Behavior

The average FP gas release rate of the MOX fuel rod in the puncture test was about 8 %. However, the average helium release rate in the puncture test was about 40%. The average helium release rate of the MOX was roughly five times as large as the average FP gas release rate. The data obtained earlier[4] from UO_2 suggest that the average helium release rate was about four times as large as that of FP gas release. And the data obtained from MOX suggest that average helium release rate was about two[5] to three[6] times or more larger than FP gas release rate. The average helium release rate for the MOX is fairly consistent with those reports.

The average FP gas release rate of the UO_2 fuel rod in the puncture test was about 3.5 %. However, the average helium release rate at the puncture test was about 50%. Therefore, the average helium release rate was roughly fifteen times as large as the average FP gas release rate. This difference between the helium release rate and FP gas release rate in the UO_2 fuel is not very different from that of earlier reports.

The difference between average FP gas release rate and average helium release rate was also reported for UO_2 fuel earlier[4][7] and explained[4] by considering the following process. At the temperature of interest, the gas is probably released by a

diffusion mechanism. Because of its small mass: helium has a diffusion coefficient about 1000 times as fast as xenon, thus it was about a 30 times higher release rate than xenon on the basis of the Booth model in which gas release rate is proportional to the square root of the diffusion coefficient. Therefore, the helium release fraction might be expected to be apparently much higher, and the helium retention in matrices might be much lower than that of fission gas such as xenon and krypton.

The FP gas fraction in the MOX is estimated as about 55 to 65 % and the helium fraction is 29 to 40%. In spite of the significant differences of the retention in matrices predicted by larger helium diffusion coefficient, no significant differences can be seen in the fractional retention in matrices between the helium and the FP gas.

For the above diffusion mechanism, the helium release fraction of MOX fuel might be expected to be higher than that of UO_2[4]. The helium production depends on plutonium content, plutonium quality, and age (americium content), and fuel bum-up. Therefore, helium generation at the periphery of a pellet is expected to be much higher than at the center. The higher helium concentration near the fuel surface makes helium atoms more readily available for release than xenon or krypton atoms which are distributed more uniformly through the fuel. In MOX fuel, the helium production is higher and radial power distribution is relatively sharper than that of UO_2. Therefore, the helium release fraction of MOX fuel might be expected to be higher than that of UO_2. However, no significant differences can be seen in the fractional helium release rate between the MOX fuel and the UO_2 fuel.

The above mentioned experimental results suggest it is difficult to describe the large difference in the gas release behavior between helium and FP gas which appeared in their release rates using only their diffusivities.

The data on UO_2 and MOX fuel reported earlier[4][6][7] suggest that helium and the FP gas have a similarity in their release behaviors. Little helium and FP gas were released when fuel rods had been irradiated at a lower linear heat rate than the threshold identified in FP gas release. When the fuel rod had experienced a higher linear heat rate than the threshold, the helium release rate increased with growth of the FP gas release rate. This seems to indicate that both gases have almost the same threshold which may correspond to the tunnel formation of gas bubbles on pellet grain boundaries.

According to the present observations, it seems that the retained helium fractions and FP gas fractions in both the MOX and UO_2 matrices are practically the same. The helium atoms do not have a potential to form bubbles independently on a grain boundary because of their relatively low number density, and exist together within the FP gas bubbles. Before tunnel formation is accomplished, the large diffusion coefficient of helium atoms moves them in an area about 300 micron in all directions as compared to a 10 micron area for xenon atoms, but the former is not enough to reach the free surface. For nearly all areas in about a 10,000 micron diameter fuel, generated helium atoms can not reach the free surface because of the relatively long distance to the free surfaces and trapping by intragranular bubbles. Because of a lack of paths to the free surface, the generated helium atoms distribute homogeneously in the matrices the same as the FP gas, and the difference in diffusion coefficients between helium and the FP gas is not important. When and where the tunnel formation and the short-cut to free surface were realized in both MOX and UO_2 fuel, helium in grain boundary bubbles may be promptly released into the free volume. After that, new helium atoms released into the grain boundary bubbles in which other helium atoms have gone away.

At the boundary area between grain boundarg bubble formation and none, grain boundary-bubbles are small, and frequent exchange of gas atoms between matrices and bubbles caused by trapping by fission gas bubbles and re-solution by fission fragments makes the proportion of helium to FP gas in the bubbles the same as that in the matrices. This means that helium and FP gas volumes on the grain boundary (and/or in the matrices) are proportional to each generation, and the proportion of helium to FP gas in each area coincides to that of its yield. Additionally released FP gas of MOX fuel might be originate from this area and include helium of almost the same proportion.

The similarity of the helium release fraction between MOX and $U0_2$ means that the released helium volume of each fuel is strongly proportional to their helium generation. The proportion of helium generation to the FP gas generation is about 0.1 for MOX, and 0.025 for $U0_2$. This difference between MOX and $U0_2$ leads to greater importance of helium release in MOX fuel. As mentioned above, if the helium release in MOX and U02 relates to FP gas release rate and is about five times larger than that of FP gas release rate, the proportion of released helium to the released FP gases always is about 0.5 and 0.125 for MOX and $U0_2$, respectively. Helium pressure increase in a high burn-up MOX fuel rod can therefore reach 30% or more of the total pressure increase. This should contribute to higher internal rod pressure.

5. CONCLUSIONS

Measurement of retained helium in MOX fuel matrices was carried out. Helium generation under irradiation was estimated by adding the released helium to the retained amount. The measured helium generation coincided with the calculated one, modified by adding ternary fission yield.

Helium pressure increase in a high bumup MOX fuel rod could reach 30% or more of the FP gas pressure increase. This should contribute to higher internal rod pressure. The impact of generated helium on fuel performance should be being investigated further.

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DEVELOPMENT AND VALIDATION OF THE ENIGMA CODE FOR MOX FUEL PERFORMANCE MODELLING

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Abstract

The ENIGMA fuel performance code has been under development in the UK since the mid-1980s with contributions made by both the fuel vendor (BNFL) and the utility (British Energy). In recent years it has become the principal code for UO_2 fuel licensing for both PWR and AGR reactor systems in the UK and has also been used by BNFL in support of overseas UO_2 and MOX fuel business. A significant new programme of work has recently been initiated by BNFL to further develop the code specifically for MOX fuel application. Model development is proceeding hand in hand with a major programme of MOX fuel testing and PIE studies, with the objective of producing a fuel modelling code suitable for mechanistic analysis, as well as for licensing applications. This paper gives an overview of the model developments being undertaken and of the experimental data being used to underpin and to validate the code. The paper provides a summary of the code development programme together with specific examples of new models produced.

1. INTRODUCTION

ENIGMA is a fuel performance code which has been under development by BNFL and British Energy (initially in collaboration but since 1991 in parallel) for more than a decade [1],[2]. The code attempts to model the fuel and clad properties and in-reactor processes which determine fuel behaviour, and has been used successfully in support of UO_2 and MOX fuel licensing both in the UK and overseas. For UO_2 fuel modelling the ENIGMA code is considered to be amongst the most up-to-date and most mechanistically based of any code currently available in the world. A particular feature of the ENIGMA development programme was a commitment to make use of essentially all of the experimental and commercial reactor data available to the UK industry. The result is a code which has been validated against over 500 rod irradiations and incorporates the lessons learned from many of the most recent and important fuel research programmes (including those at Halden, Risø, Studsvik, BR2, and many others).

For BNFL, current efforts are strongly focused on MOX fuel issues, with the intention of developing a state-of-the-art MOX fuel code as part of a wider strategy for Short Binderless Route (SBR) MOX product development and qualification. The present paper considers the current status of the code with respect to MOX fuel modelling, highlighting some of the experimental results which have influenced current thinking. It also identifies areas of on-going and future study, although in the limited space available only a sub-set of the issues are considered.

Much of the MOX data used for benchmarking the code to-date has come from international programmes which have studied MOX fuels from a variety of different sources. Relatively little data has existed on BNFL's own SBR fuel product. Many programmes are however now in place or are being planned and the SBR performance database is beginning to grow rapidly. PIE has been completed on prototype fuel [3] and the first PIE of commercially irradiated SBR fuel is presently underway, as considered in a separate paper at this meeting [4]. A lot of valuable in-pile data is also coming from test irradiations in Halden, from both bilateral and joint programme experiments.

BNFL is committed, as a MOX fuel vendor, to testig, understanding and proving its product, The programmes to achieve this have been defined and initiated, and the teams of scientists and engineers required to oversee the work have been assembled. The development of the ENIGMA code is one key element in this overall strategy.

2. MOX FUEL MODELLING

2.1. Nuclear Physics Models

Radial Profile Evolution. The RADAR model [5] is implemented in ENIGMA to calculate the within pin radial power distribution. Previous code versions have been based on simplified calculations of the depletion of the ^{235}U, ^{239}Pu and ^{241}Pu isotopes in a fixed number of radial fuel annuli. The start-of-life ^{239}Pu and ^{241}Pu contents were previously combined into an 'effective plutonium content', which was depleted as ^{239}Pu. The calculations were tuned to match predictions from the neutronics code WIMS. The radial power distributions determined in this way were satisfactory for MOX fuel at low burnups and with low plutonium contents, but recent studies have shown some deficiency at higher burnups and/or high plutonium contents, in particular in the soft spectrum of the Halden reactor.

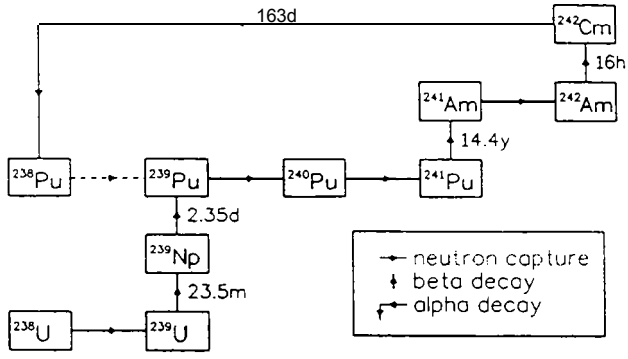

FIG. I. Isotope Model for Calculation of Power and Heavy Metal Concentration Profiles

To address this, an improved version of RADAR has been developed which extends the depletion equations to cover all of the relevant heavy metal isotopes, as illustrated in Figure 1. Cross-sections used have been based loosely on those from the JEF library, with the depletion calculations tuned to match those from the lattice code CASMO-4 [6]. The revised model has been benchmarked for commercial PWR, BWR and Halden reactor conditions. The improved version of RADAR gives accurate radial power profile predictions at both high burnups and high Pu contents. Additional work is planned to provide further benchmarking of the routine against data from EPMA and/or SIMS.

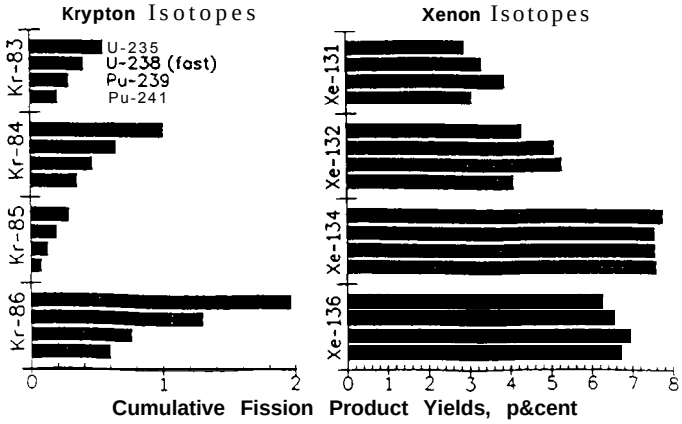

FIG. 2. *Fission Gas Yields from Uranium and Plutonium Fissions*

Fission Gas Generation. To complement the improved WAR model, a new fission gas generation model for MOX fuel has also been developed. The generation model calculates the net number densities through life of each of the krypton and xenon isotopes ^{83}Kr to ^{86}Kr and ^{131}Xe to ^{136}Xe, plus the important precursors ^{131}I and ^{132}Te, in each of the fuel annuli.

Generation, neutron capture and decay are computed based on the power history and the uranium and plutonium isotope distributions. The new model allows a more accurate calculation of the net fission gas generation rate which takes account of the different krypton and xenon isotopic yields of the various fission events, as shown in Figure 2. This is then used as the basis for the development of a more advanced fission gas release model. The new generation model also allows the isotopic composition of the fission gas in the fuel-clad gap to be evaluated. This may be important in gap conductance calculations and the predictions can be compared with measured data from PIE gas puncture tests and from gas flow experiments.

Helium Generation. Helium generation in fuel occurs via two main mechanisms – alpha-decay and ternary fission. Although many of the heavier isotopes present in fuel are alpha active, it is found that by far the dominant contributor is ^{242}Cm, which has a half-life of only 163 days. In UO_2 fuel relatively little ^{242}Cm is generated at typical current burnup levels; the main source of helium therefore tends to be from ternary fission, and overall helium generation is generally negligible. By contrast, in MOX fuel the build-up of ^{242}Cm can be significant, particularly for fuel derived from aged or low quality plutonium sources. Helium production can then be significant, especially at high burnups. The new version of the WAR routine (see above) incorporates the calculation of the ^{242}Cm build-up. Determination of the alpha decay and ternary fission yields is then straightforward, giving the code a reasonable estimate of the total helium generation.

2.2. Fuel **Materials Properties**

In collaboration with its customers, BNFL has undertaken a substantial programme of materials properties measurements on SBR MOX fuel [7]. For several properties, including thermal expansion coefficient, specific heat capacity and elasticity, the evidence points to no significant difference between MOX and UO_2 characteristics within the range of parameters studied. Fuel melting point is clearly reduced by the presence of plutonium, but this is not a modelling issue since the design limit used as a failure criterion is sufficiently conservative to cover all fuel types including MOX. The two physical properties which are most MOX-specific are thermal conductivity (discussed below) and creep (discussed in Section 2.5).

Fuel Thermal Conductivity. The low temperature behaviour of the fuel thermal conductivity is well modelled by an equation of the form $1/(a+bT)$, where *a* describes the phonon-impurity scattering, *b* represents the phonon-phonon scattering and T is the absolute temperature. This expression describes the behaviour of both standard urania fuel and MOX fuel and the effects of burnup can be accommodated by an increase in the phonon-impurity scattering term arising from the effects of the build-up of fission products in the matrix.

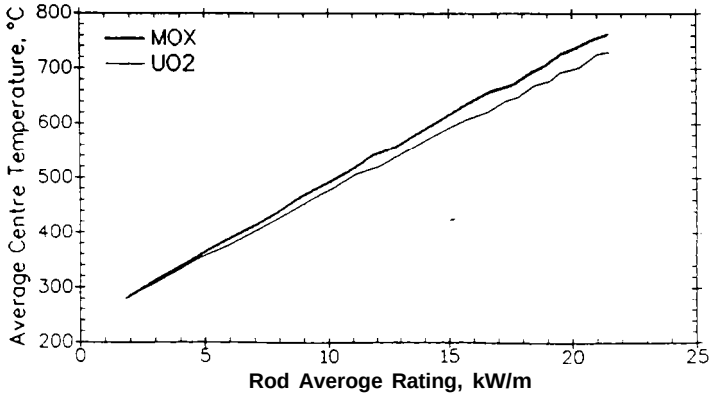

FIG. 3. Comparative Start-of-Life Temperature Measurements on UO_2 and MOX Fuel Rods

The range of plutonium concentrations envisaged for commercial irradiations is not expected to exceed 10 wt% so it is in this region where thermal conductivity is pf interest. It is sensible to ask whether the transition behaviour from standard Urania to MOX fuel takes place gradually or abruptly. Gibby [8] has measured the thermal conductivities using a laser-flash thermal diffusivitymethod for a range of urania-plutonia solid solutions with 0, 5, 12, 20, 25 and 30 wt% PuO_2. These data can be re-analysed by plotting the inverse conductivity against absolute temperature. This should yield a linear dependence of slope, *b,* and intercept, a. The effect of plutonia appears to modify both the intercept value, a, and the slope, *b, indicating* that there is both an impurity effect on phonon scattering as well as a modification to the phonon spectrum which impacts on the phonon-phonon scattering term. However, the dependence on plutonia content (except at the 30 wt% level) is found to be weak and it is therefore possible to take the plutonium dependence out of the denominator and move it into the numerator. The MOX effect is consequently applied in ENIGMA as a fixed 8% reduction in the numerator of the conductivity expression. Comparative MOX versus UO_2 start-of-life temperature measurements from a Halden experiment are shown in Figure 3, supporting the choice of the 8% reduction.

Conductivity Degradation with Burnup. The effect of irradiation on the fuel is the gradual replacement of fissile atoms with two or more fission product atoms. To a first approximation this increases the phonon-impurity scattering and hence the *a* term. Analysis of out-of-pile annealed laser-flash specimens indicates that the increase in the *a* term is accompanied by a decrease in the *b* term *[9],* but the latter effect is small and in practice a forced-fit with an increased *a* term only can be used. To represent the effects of fission products the conductivity in ENIGMA is modified to an equation of the form $1/(a_0+a_1B(1-f)+bT)$ where B is the burnup andfis the fraction of fission gas atoms not in solution in the matrix, i.e. the fractional sum of all gas which is either in bubbles or has been released.

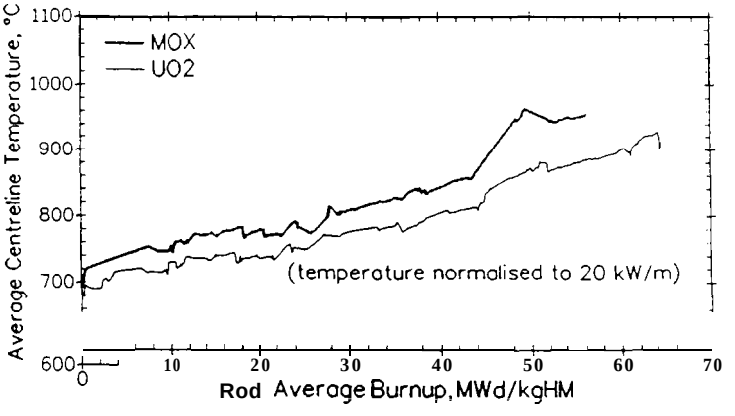

FIG. 4. Comparative Fuel Conductivity Degradation Rates in UO_2 and MOX Fuel Rods

For MOX fuel, the code assumes the same value of the degradation parameter a_1 as for UO_2. Figure 4 shows some in-pile experimental data which confirms that the evolution of normalised temperature with

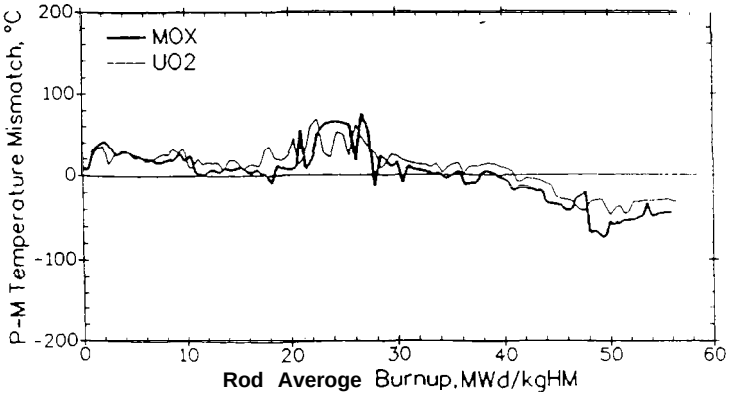

FIG. 5. ENIGMA Fuel Centreline Temperature Predictions for MOX and UO_2

burnup for MOX and UO_2 rods is essentially parallel, supporting the above assumption. As Figure 5 shows, the code's temperature predictions are found to be satisfactory for both MOX and UO_2 through to high burnup. The gradual trend toward underprediction at high burnups could be associated with some subtlety in the degradation process or in the modelling of fission gas porosity; experiments and theoretical work are in progress to address these issues.

2.3. Fuel Microstructural Changes

Fuel Densification. The code contains models for the sintering of the as-fabricated porosity and for the gradual swelling of the fuel due to solid and volatile fission products. The densification model [10] is formulated in terms of fast and slow densification processes, each an inverse exponential in burnup and also a function of temperature and grain size, with a temperature-dependent partitioning between the two. The model was fitted to a database of in-pile UO_2 fuel stack length measurements from Halden. Although the corresponding database of MOX fuel measurements is currently very limited, the existing model generally appears to perform satisfactorily, once the porosity, grain size and temperature differences are allowed for. However, further work is planned in this area.

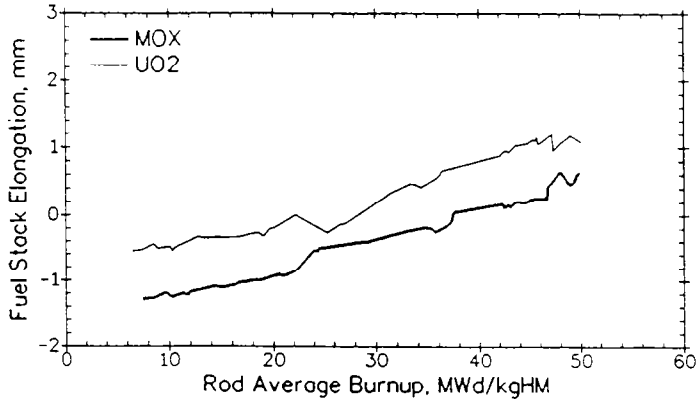

FIG. 6. Comparative MOX and UO_2 Stack Elongation Trends with Burnup

Fuel Swelling. The swelling of the fuel due to solid fission products is modelled as a linear function of burnup. Since much of the swelling arises from elements which are volatile at higher temperatures, the model is also coupled to the gas release model so that the swelling is reduced as fission products are released. The additional swelling caused by the formation of gas bubbles is then modelled separately. For benign irradiation conditions solid fission product swelling is the dominant effect. ENIGMA currently assumes a figure of 0.5 vol% per atom% burnup, although a broad range of swelling rates (0.2% to 1.0%) have been found in different experiments. This variability in swelling may be associated with the interactions between swelling, densification and creep. In a comparative MOX versus UO_2 irradiation in Halden the evidence suggests similar swelling rates for the two fuel types, as shown by the parallel trends in Figure 6.

The code's predictions of stack length changes, both from in-pile and post-irradiation measurements, appear to be satisfactory, and unbiased as a function of burnup, as shown in Figure 7. While the densification and swelling models are currently assumed to be isotropic, diametral swelling is found to be underpredicted in some instances; again, further studies are planned in this area.

Fuel Grain Growth. At sufficiently high temperature fuel grain size can increase during operation, particularly early in life before the grain boundaries become pinned by fission gas bubbles. Accurate modelling of the grain size is important because of its influence on fission gas release, densification and creep behaviour. The ENIGMA model for this process was tuned using data exclusively from UO_2 irradiations [11]. When initial grain size differences are taken into account, along with differences in operating temperature, the model is also found to perform satisfactorily for MOX fuel. At present,

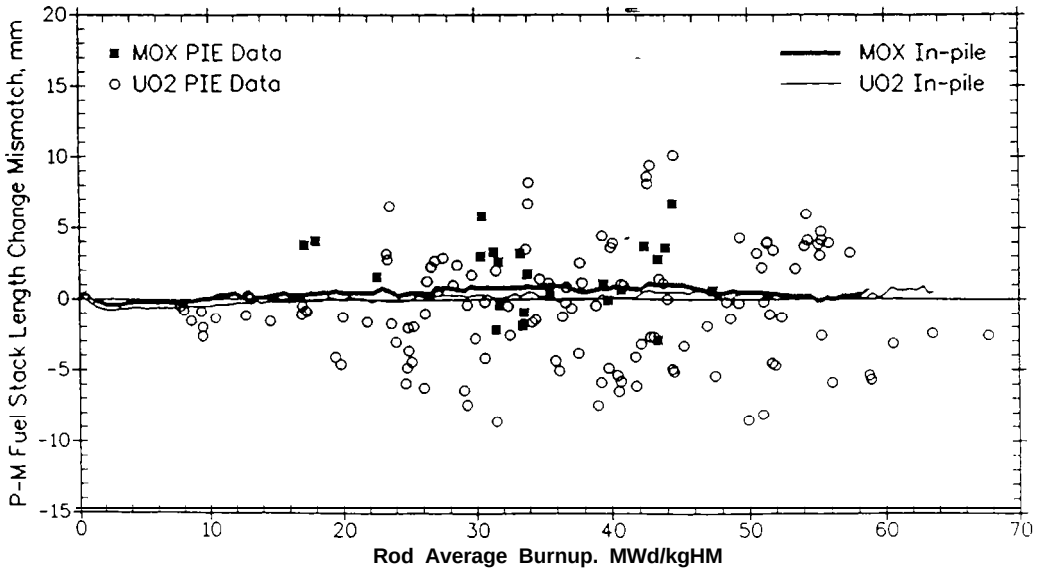

FIG. 7. ENIGMA Stack Length Change Predictions as a Function of Burnup

however, PIE cases where positive grain growth has been observed in SBR fuel are currently limited to those seen under the fairly extreme conditions experienced in the Callisto tests [3]. Further evaluation of the modelling is therefore planned as new PIE data become available.

Homogeneity Changes. Except for rim formation in high burnup fuel, standard UO_2 remains an essentially homogeneous material, and the current ENIGMA code models fuel in this way. In contrast, MOX fuel begins as a heterogeneous material containing small islands of plutonium-rich material. The combined effects of inter-diffusion and plutonium burnout/generation gradually homogenise the material such that in the high burnup limit MOX fuel becomes largely indistinguishable from UO_2. The impact on performance depends strongly on the degree and scale of the initial heterogeneity. For SBR fuel, which has relatively small plutonium agglomerations compared with some other MOX fuels, the need for explicit modelling of the homogenisation process is uncertain. This issue is being assessed as part of the on-going development of a mechanistic fission gas release model for MOX fuel.

Rim Effect. The code contains a simple empirical model to account for the observed gas release and fuel conductivity effects resulting from the formation of the refined, highly porous microstructure at the periphery of high burnup fuel. The model was fitted using UO_2 data but is currently applied equally to MOX fuel. The validity of this assumption is questionable, but study of the rim effect is an active area of research and the development of a more mechanistic treatment should become possible in due course. At present the code also makes no attempt to model explicitly the development of the clad inner bore corrosion layer and the resulting fuel-clad bonding that can occur at medium to high burnups. Since plutonium containing fuels may be more oxidising than standard UO_2, these effects could be more pronounced and possibly more significant in MOX fuel.

2.4. Fission Product Release

Stable Gas Release and Swelling. The ENIGMA code embodies a sophisticated mechanistic fission gas release and swelling model [12,13,14]. The model was formulated solely on the basis of UO_2 fuel measurements and has been validated against a broad range of experimental and commercial irradiations. A histogram showing the number of rod irradiations (plus post-irradiation puncture tests) used for validation purposes is included in Figure 8 as a function of burnup. For MOX fuel, the size of

the database is clearly much reduced, but, as the main plot B-l Figure 8 shows, the degree of agreement between predicted and measured gas release is found to be as good for MOX as for UO_2 fuel.

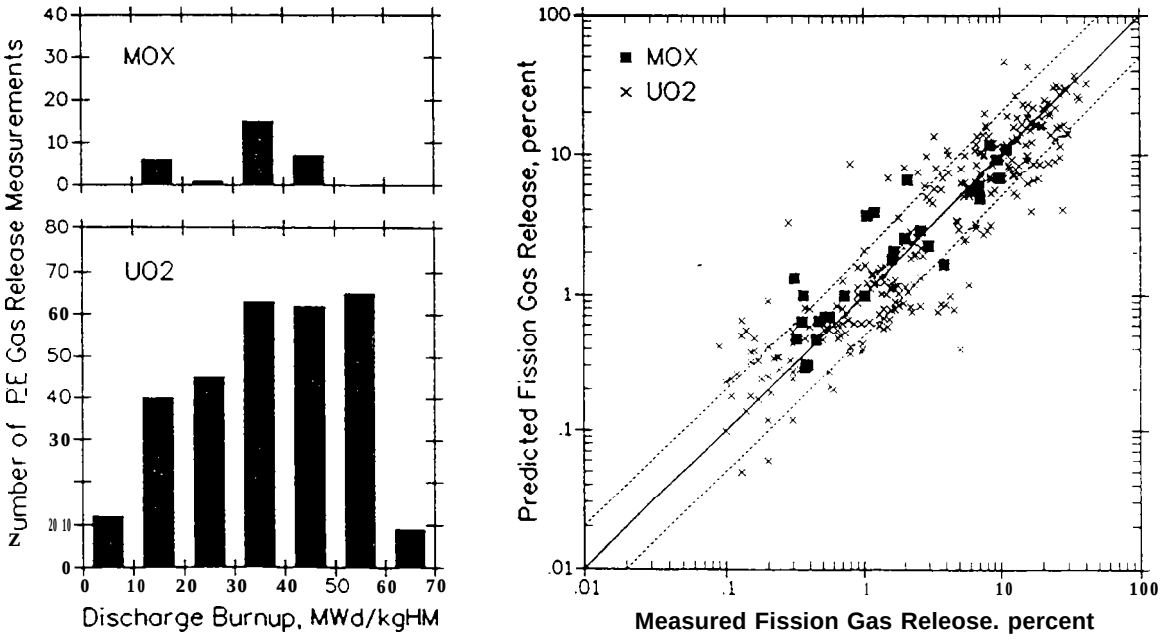

FIG. 8. ENIGMA Fission Gas Release Database for UO_2 and MOX Fuel

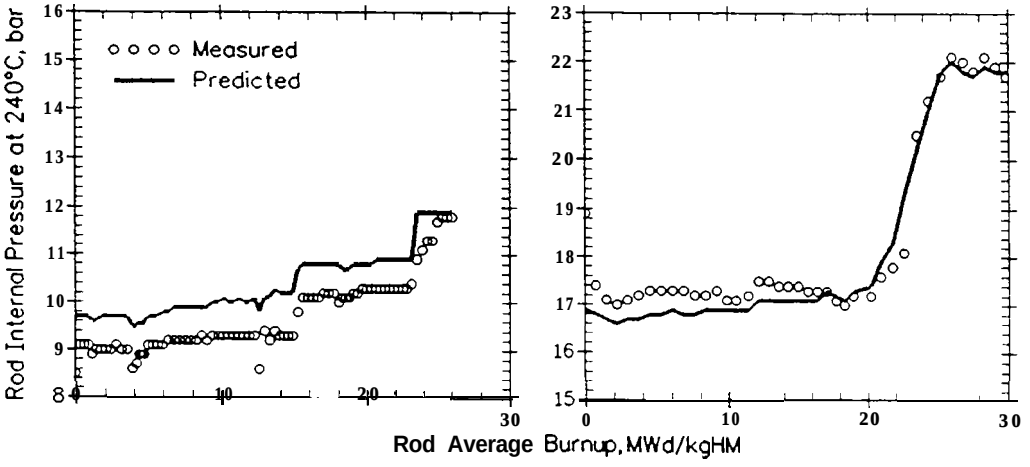

FIG. 9. Fission Gas Release Onset Predictions in two Halden MOX Experiments

Moreover, the existing model provides good predictions for the onset of gas release, as revealed in Figure 9 which shows pressure measurements in an instrumented rod in Halden. However, this may be somewhat fortuitous, particularly for some of the older heterogeneous fuels (e.g. from the PRIM0 programme), since the details of the release process may be quite different. There is evidence that the existing model can significantly underpredict the levels of gas bubble swelling under certain ramp test conditions. The development of a mechanistic gas release and swelling model specifically for MOX fuel is therefore envisaged. Some components of the model, for example a MOX-specific fission product generation routine (as discussed in Section 2.1) are already in place. Other components will emerge as part of the programme of experimental studies, which will include in-pile measurements of fresh and re-instrumented fuel rods, and SEM/TEM of irradiated, annealed and ramp tested fuels.

Helium Release. The helium generated from ternary fissk% and alpha decay (as discussed in Section 2.1) is lost from the fuel matrix by a diffusive process Diffusion of the small helium atoms is considerably more rapid than that of krypton or xenon. However, reliable diffusion coefficient measurements have not been reported and hence the code currently uses a simplified empirical release model. The helium released is assumed to escape immediately to the fuel-clad gap, with no hold up in the grain boundary bubbles. The acceptability of these simplifications would become questionable for situations where the amount of helium released were significant compared with that of the krypton and xenon, e.g. for very high burnup or for aged or recycled plutonium sources. However, the current approach is considered acceptable for existing MOX fuel types and fuel duties, although the validation database is rather small.

Short-Lived Fission Product Release. Gas flow experiments in Halden have produced measurements of iodine and other short-lived fission product release levels [15], and the code contains a simple, conservative model to predict iodine release levels under steady state conditions. All measurements to-date have been on UO_2 fuel. Since ^{131}I yields are significantly higher from plutonium fissions than uranium, the applicability of the model for MOX fuel has yet to be established. To remedy this, a new date have been on UO_2 fuel. Since ^{131}I yields are significantly higher from plutonium fissions than uranium, the applicability of the model for MOX fuel has yet to be established. To remedy this, a new bilateral gas flow experiment has recently begun at Halden which will compare release levels in MOX and UO_2 fuels. The start-up phase of the test was completed successfully in March 1999. Assessment of the data and the implications on short-lived fission product release modelling is underway.

2.5. Pellet Mechanical Response

Thermal Creep. In u&-radiated conditions, fuel undergoes thermal creep in response to compressive stress. The thermal creep strain rate is generally modelled for both UO_2 and MOX as the sum of a linear term proportional to applied stress, which is consistent with diffisional creep, and a power-law term proportional to stress to the power 4.5, which is consistent with dislocation climb – annihilation creep. Recent proprietary tests comparing the fuel creep behaviour of UO_2 and SBR MOX fuel have shown that at high stresses and/or low fuel temperatures, creep strain rates proportional to stress to the power n, where n is significantly larger than 4.5, are observed, as illustrated in Figure 10. Thus, new thermal creep strain rate equations for both UO_2 and MOX fuel have been developed to fit both the proprietary and open literature fuel creep data which include additional power law creep terms proportional to stress to a high power.

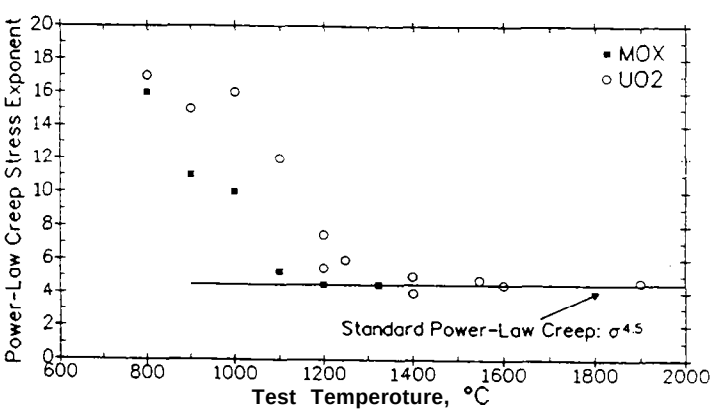

FIG. IO. Power-Law Creep Stress Exponent: Proprietary and Open Literature Data

Irradiation Creep. Under irradiation fuel linear thermal creep rates are found to be enhanced by a substantial factor (this phenomenon is referred to as irradiation-enhanced creep). On top of this, an additional creep component arises which is linear in stress and independent of temperature and which occurs only in the presence of irradiation (irradiation-induced creep). This latter creep component is generally dominant under most conditions. The available database on irradiation creep has recently been reviewed. Irradiation-enhanced creep terms were modified for both UO_2 and MOX fuel to include a

fission rate dependence Also, the terms for irradiation-induced creep were optimised against the available database. Compared with the previous model the revised formulation employs roughly a five-fold increase in the irradiation creep rate under typical fuel irradiation conditions. The effects of this increase are illustrated in Figure 11 which shows rod diameter variation at a constant linear rating of 20 kW/m as a function of burnup. After the minimum diameter is reached at the point of gap closure, the effect of the increased creep is to reduce the clad outward strain rate due to improved pellet accommodation. Significant effects of the change in creep modelling are also observed in ramp cases. The increased creep observed with MOX fuels is a factor in its improved PCI resistance.

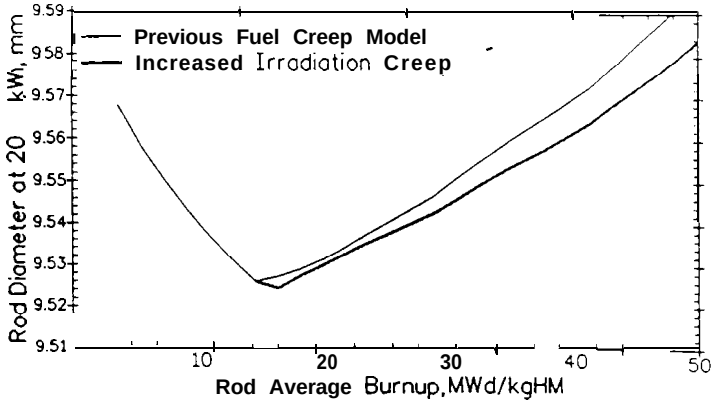

FIG. I I. Effect **of** *Increased Irradiation Creep Rate on Steady State Rod Diameter Predictions*

Pellet Cracking and Relocation. The code employs simple empirical adjustments to account for the thermal and mechanical effects of pellet cracking and relocation behaviour. These also take in the effects of pellet-clad eccentricity and clad ovality. The gap conductance model [16], as well as containing classical terms for gas conductance and radiation, also includes an additional empirical component to account for relocation effects. In the absence of specific data, the applicability of these empiricisms to MOX fuel currently has to be assumed, but it is possible that differences in pellet properties (surface roughness, plasticity, cracking patterns, etc) could necessitate the re-benchmarking of some aspects of the empirical modelling. However, no major shortcomings have been identified from the observations made so far.

Pellet Wheatsheafing and Ridging. The effects of thermal gradients and cracking result in the pellet adopting a wheatsheaf shape, such that after clad creep-down has occurred the cladding profile displays a characteristic ridging pattern. The code contains models for the size of the wheatsheaf height and its change during a power increase or decrease, and for the response of the cladding to changes in pellet shape. Testing of these models against prototypic data for MOX fuel is currently very limited. There is some evidence to suggest that ridging patterns observed on SBR MOX fuel may be a little different to those found on UO_2 fuel, with less well-defined primary ridges and a greater propensity for secondary ridges, but much more data is required to substantiate this.

PCI Performance. During power ramps, pellet fragments expand and move apart, widening the cracks between them. Under fuel-clad contact conditions this leads to azimuthal stress concentration in the cladding and potentially to clad failure by stress corrosion cracking. ENIGMA contains models to predict pellet-clad interaction behaviour, although application of the code is generally confined at present to assessments of conditioning and de-conditioning effects, with failure propensity determined by reference to the empirical database of PCI ramp test results. The existing model was developed for UO_2 fuel. Its applicability to MOX could be affected by a number of factors, for example, higher fuel creep rate, differences in cracking patterns, or differences in fuel-clad bonding behaviour and hence in fuel-clad friction coefficients. The limited **experimental** evidence available so far points to failure propensity being significantly lower in MOX fuel than in standard fuel (see Figure 12), although the reasons for this are not fully understood. Further ramp test studies on MOX fuel are planned in the near

future, both as part of international collaborative projectS and in BNFL's own programmes. These should provide the raw material for improved mechanistic modelling of MOX fuel PCI behaviour over the next few years. Nevertheless, it must be recognised that providing a mechanistic treatment of PCI failure is perhaps the ultimate challenge for a fuel performance code.

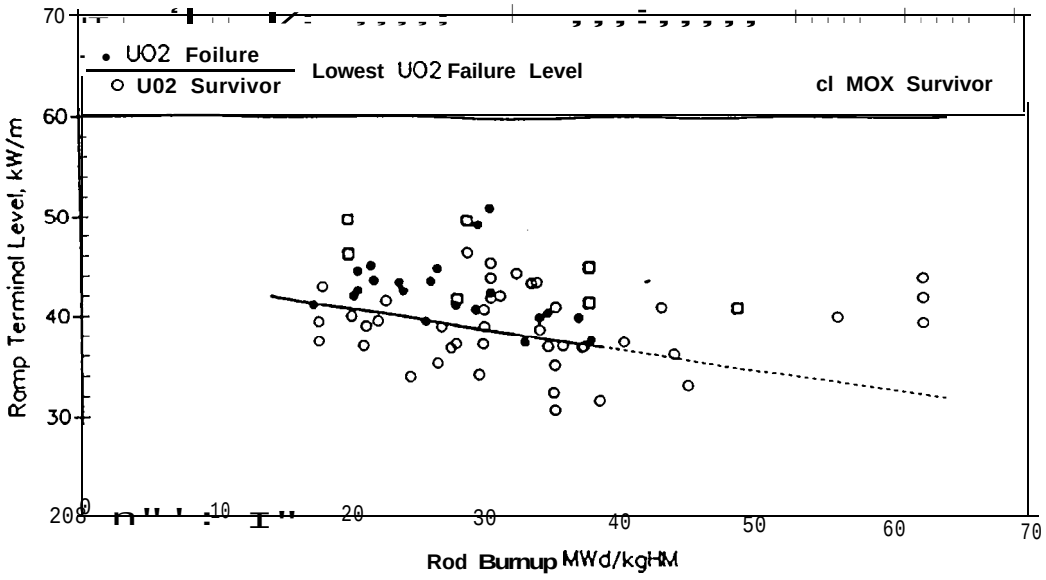

FIG. 12. *Comparative PCI Failure Levels in UO_2 and MOX Fuel*

3. SUMMARY AND CONCLUSIONS

As part of a wider strategy for SBR MOX fuel testing and qualification, a fundamental m-evaluation of the ENIGMA fuel modelling code is being undertaken in order to provide a state-of-the-art tool for MOX fuel behaviour assessment and licensing. Many of the code's component sub-models are being reviewed and further developed where required, including sub-models for:-

- evolution of the radial profile of heavy metal isotopes
- calculating fission product spectra
- helium generation
- evolution of the fuel microstructure including rim structure formation
- fuel thermal conductivity and its evolution with burnup
- fuel densification and solid fission product swelling behaviour
- irradiation and thermal creep properties
- stable fission gas release and gas bubble swelling behaviour
- fuel grain growth
- short-lived fission product generation and release
- ramp test behaviour and PCI resistance.

The paper provides a status report on the work being performed and highlights some of the key issues involved.

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FUEL ROD DESIGN BY STATISTICAL METHODS FOR MOX FUEL

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Abstract

Statistical methods in fuel rod design have received more and more attention during the last years. One of different possible ways to use statistical methods in fuel rod design can be described as follows: Monte Carlo calculations are performed using the fuel rod code CARO. For each run with CARO, the set of input data is modified: parameters de-scribing the design of the fuel rod (geometrical data, density etc.) and modeling parameters are randomly selected according to their individual distributions. Power histories are varied systematically in a way that each power history of the relevant core management calculation is represented in the Monte Carlo calculations with equal frequency. The frequency distributions of the results as rod internal pressure and cladding strain which are generated by the Monte Carlo calculation are evaluated and compared with the design criteria. Up to now, this methodology has been applied to licensing calculations for PWRs and BWRs, UO_2 and MOX fuel, in 3 countries. Especially for the insertion of MOX fuel resulting in power histories with relatively high linear heat generation rates at higher burnup, the statistical methodology is an appropriate approach to demonstrate the compliance of licensing requirements.

1. INTRODUCTION

The insertion of MOX fuel assemblies in LWRs can be guided by different aims, e.g. maximum reduction of Pu stockpile, maximum operational flexibility, no modification of the existing fuel management strategy, no safety penalties and if possible, no economic penalties, c.f. [1] to [4]. In any case it is important for design and licensing purposes to describe the expected behavior of the MOX fuel rods as precisely as possible based on appropriate methods and codes.

It was found that design analyses with statistical methods, using the fuel rod code CARO, fulfill the requirements on accuracy and reliability not only for UO_2 fuel, but also for MOX fuel, especially for modern core designs with higher enrichments and more demanding fuel rod power histories.

In the following, the features of the fuel rod code CAR0 with respect to MOX fuel will be described and an overview of the statistical methodology, its application to MOX fuel rods and the current status of its introduction into licensing will be given.

2. MOX FUEL CHARACTERISTICS AND THEIR MODELLING WITH CAR0

The insertion of MOX fuel assemblies in a reactor core normally takes place after insertion of already licensed UO_2 fuel assemblies.

To perform design calculations, all aspects of MOX fuel which could lead to a different behavior, have to be considered: material properties, neutronic de-sign characteristics, and irradiation behavior. The following points have to be addressed:

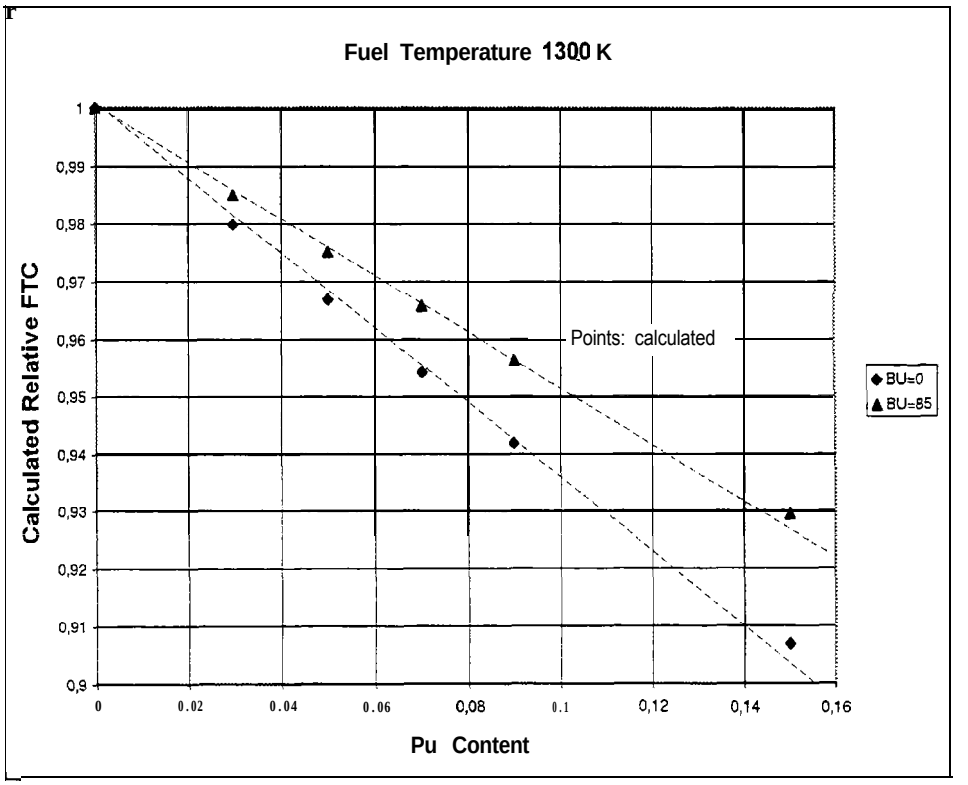

FIG. 1: Relative fuel thermal conductivity reduction versus Pu content

a) material properties of MOX fuel
- meiting point as a function of burnup
- fuel thermal heat conductivity as a function of burnup
- grain size, porosity distribution
- thermal elongation

b) neutronic design characteristics
- radial power density distribution in the pellet
- power histories
- neutron flux

c) irradiation behavior
- fission gas release, ratio of Xe to Kr
- densification / swelling of the fuel
- He release

Examples of the modeling of these effects in the fuel rod code CAR0 are given in the following Figures. Figure 1 shows the reduction of the fuel thermal conductivity versus the Pu content. The reduction decreases with burnup due to the saturation of the influence of the defect concentration on the fuel thermal conductivity. In Figure 2, the radial power density distribution of MOX fuel compared to UO_2 fuel is given for a burnup of 0, 30, and 70 MWd/kgM. These two examples show minor differences between UO_2 and MOX fuel, but the next feature has significant influence: the evolution of linear heat generation rate with time. Figure 3 indicates two power histories of equal burnup, one for UO_2 fuel and one for MOX fuel, with the typically slower decrease of reactivity for MOX fuel. This, in turn, leads to a relatively higher fission gas release of MOX fuel rods. It can be asked if this typical difference in power histories explains the observed higher fission gas release of MOX fuel.

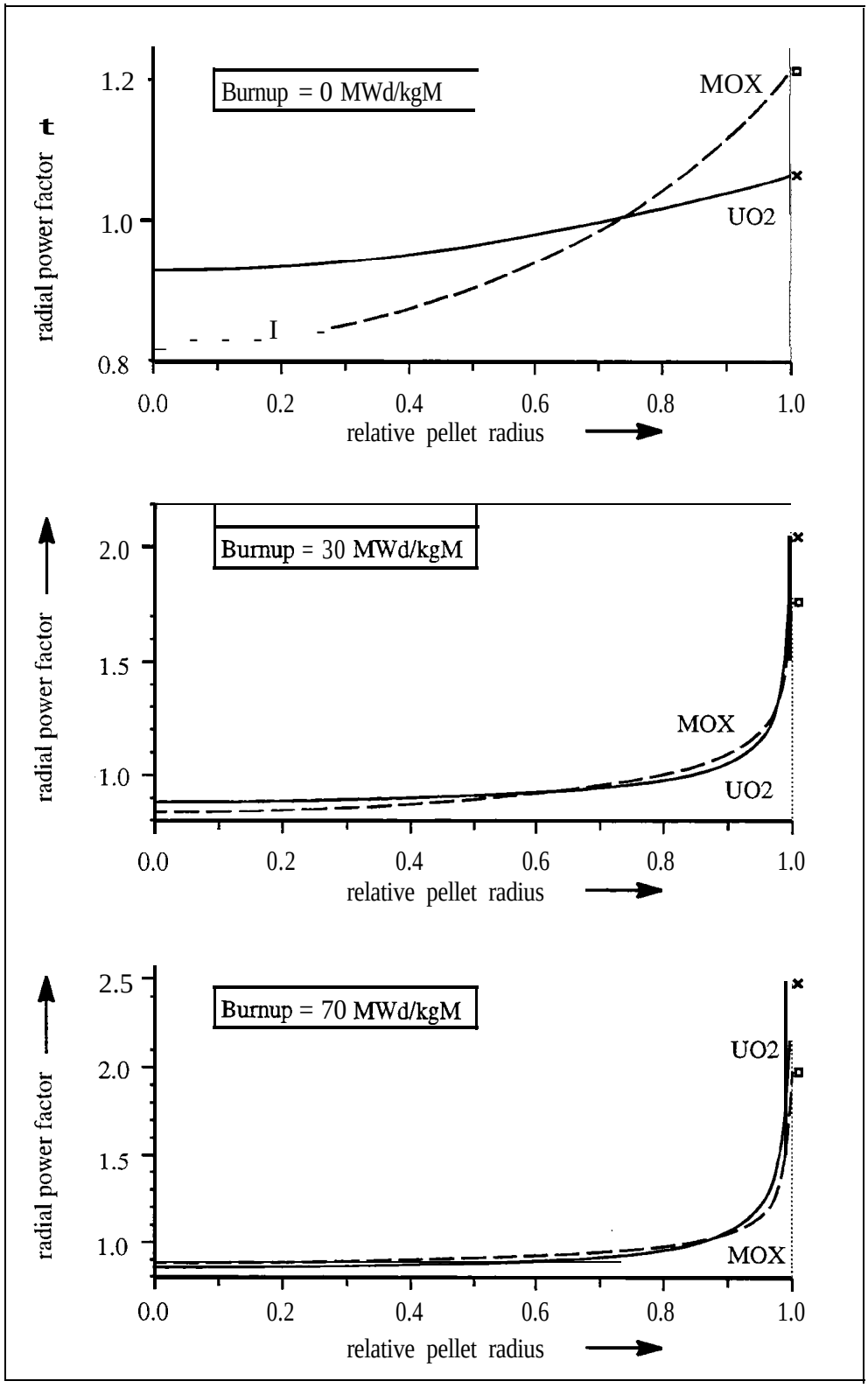

FIG. 2: Radial power density distribution for different burnups

For CAR0 this question can be answered as follows: Taking into account the above mentioned MOX features explicitly via modeling of the separate effects or implicitly via validation by a broad data base of measured values, see Table 1, it is demonstrated by Figure 4, that there is no systematic deviation between calculated and measured fission gas release values for UO_2 and MOX fuel, using the same set of modeling parameters. This is also confirmed by [5], [6], [7].

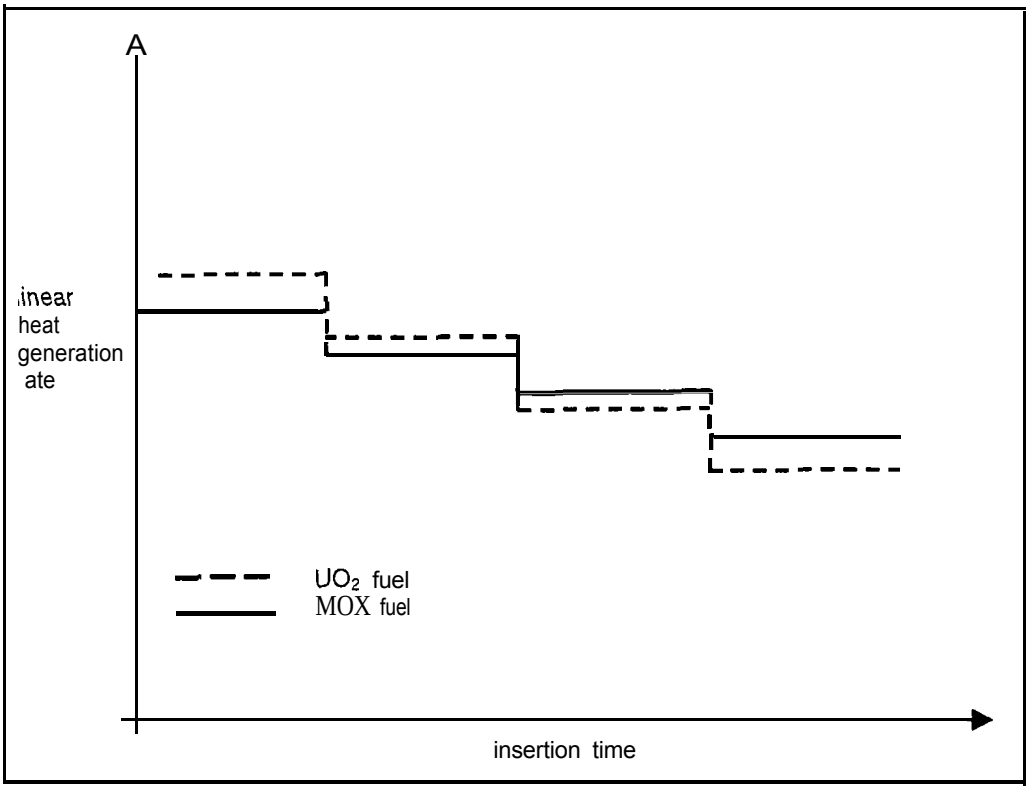

FIG. 3: *Typical power histories for UO_2* and *MOX fuel* with *equal* burnup

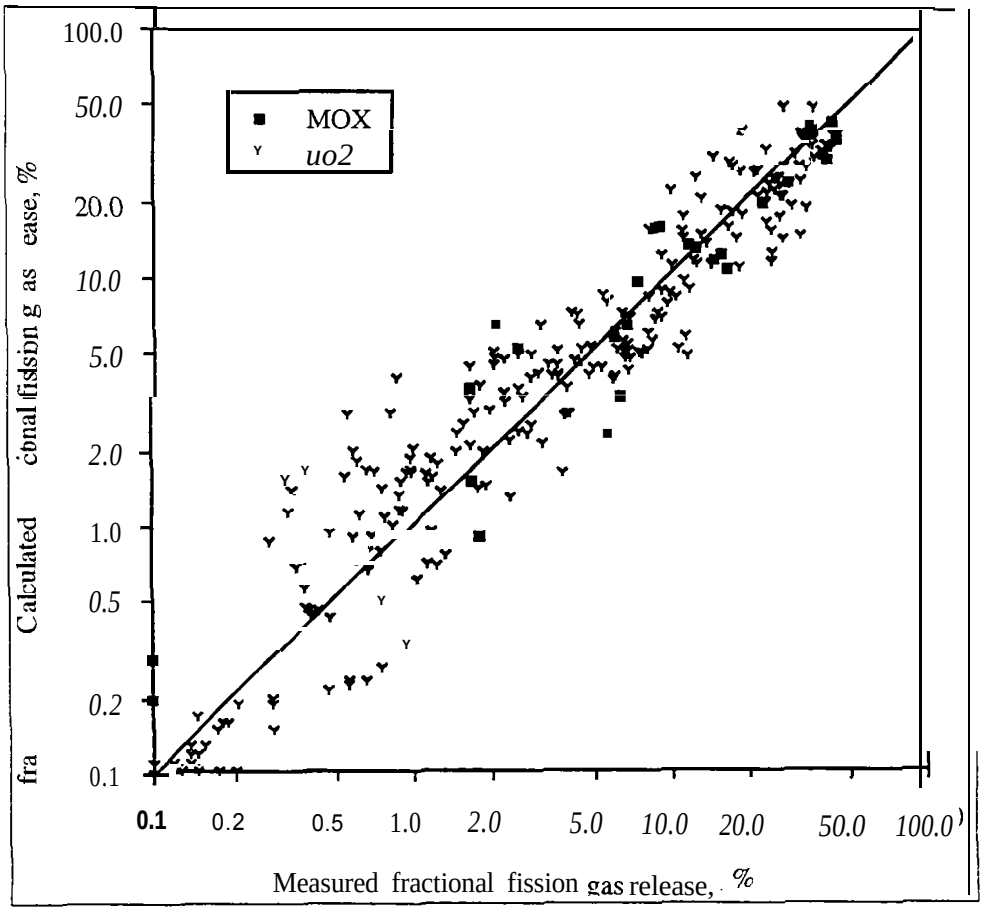

*FIG. 4: Validation of the **fission** gas release model for PWR fuel rods: Comparison measured/with CAR0 calculated values*

TABLE 1. VALIDATION DATA BASE FOR CARO: FISSION GAS **RELEASE VALUES FOR PWR** FUEL

	1990	1996	1999
Number of fuel rods	114	267	316
Maximum fuel rod burnup [MWd/kg]	53.5	77.8	90
Number of fuel rods with burnup > 60 MWd/kg		4	19
Number of fuel rods with MOX fuel	**13**	**28**	**40**

3. STATISTICAL METHODS

Statistical methods in fuel rod design have received more and more attention during the last years. This is demonstrated not only by some recent publications on this subject, c.f. [8] to [13], but also by the fact that the American National Standard for Light Water Reactors, Fuel Assembly Mechanical Design and Evaluation [14], quotes explicitly the option to use "probability analyses in which the variances of independent parameters are statistically combined".

Of course there are different ways to use statistical methods in fuel rod design, as described e. g. in [9] or [11]. One method is presented in short in the following. It has been applied to fuel rod licensing for several projects, both PWR and BWR, UO_2 and MOX fuel.

3.1 Description of the method

The scheme of the statistical method is shown in Figure 5 (a description in more detail is given in [15]): Monte Carlo calculations are performed using the fuel rod code CARO. For each run with CARO, the set of input data is modified: parameters describing the design of the fuel rod (geometrical data, density etc.) and modeling parameters are randomly selected according to their individual distributions. Power histories are varied systematically in a way that each power history of the relevant core management calculation is represented in the Monte Carlo calculations with equal frequency. (The complete set of all occurring power histories is provided by nuclear core design via interface data file, which is automatically transformed into an input data file for the fuel rod code.)

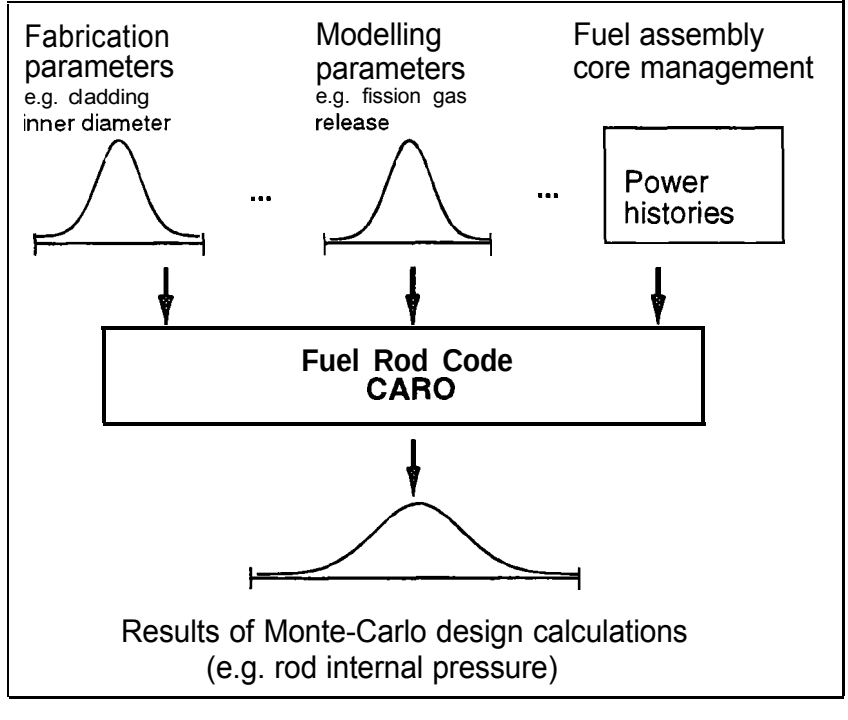

FIG. 5: Scheme of statistical fuel rod design

The frequency distributions of the results as rod internal prasure and equivalent cladding strain which are generated by the Monte Carlo calculations are evaluated and compared with the design criteria.

By this methodology, two aspects of fuel rod design are getting higher importance than within the frame of conservative deterministic methods: the distributions of fabrication parameters and the consideration of power histories.

3.2 Treatment of fabrication parameters

For conservative deterministic design calculations, only the tolerance limits of fabrication parameters are important. For statistical design calculations, certain assumptions on the distribution of the most important fabrication parameters are necessary.

It was shown by exhaustive statistical analyses of quality control data, e. g. of pellet diameter and cladding inner diameter [16], that the following assumption is justified: all distributions of fabrication parameters are represented by a Gaussian distribution, cut off at 3 σ, where the $\pm$ 3 o-limits coincide with the two tolerance limits.

3.3 Treatment of power histories

As statistical calculations are based on a set of real given power histories from a certain core management calculation, it has to be checked if the design analyses are valid for following cycles, too.

Additionally, variations of the set of power histories from cycle to cycle should be covered and margins for core management purposes should be available. Therefore, the Monte Carlo calculations are performed with modified power histories: The original fuel rod power factors of each cycle, resulting from core management calculations, are multiplied by chance and independent of each other with a constant factor. This factor has a Gaussian distribution around 1 with a scattering of 2 σ (the distribution is cut off at 2 σ).

By this method the batch averaged burnup stays about constant. The maximum fuel rod burnup increases slightly because some power histories which have already a relatively high burnup are multiplied by chance by factors greater than 1 in the average. The power jumps between two cycles, important for fission gas release, are partly increased also.

An example for a typical 1300 MW PWR is given in Figure 6: In this case, the fuel rod powers are multiplied by a value of + 20 %. This means that the factors are in the interval [0.8, 1.21. For each burnup interval (steps of 0.1 MWd/kg(U)) the maximum fuel rod averaged power (including the described statistical variation) of each fuel assembly is plotted against fuel rod burnup, together with an upper envelope. With this set of power histories, the design analysis is performed and the design criteria have to be fulfilled.

In the same Figure, a dashed curve is integrated which is the upper envelope of the maximum powers without statistical variation. The gap between the two curves provides the desired margins..

4. APPLICATION OF THE STATISTICAL METHODOLOGY TO MOX FUEL RODS

As discussed in Chapter 2, higher fission gas release can be expected for MOX fuel rods compared to UO_2 fuel rods, if burnup and fuel management strategy are the same. Especially in case of a fuel rod design analysis for high burnup in connection with highly demanding power histories, a realistic assessment of the expected behavior of the fuel rods is important. This realistic assessment can be assured by using the statistical design methodology, based on a fuel rod code which is validated against a broad database of experience feedback up to high On the basis of a realistically calculated distribution of the expected maximum rod internal pressure values the decision on the feasibility of a fuel reload scheme is much better justified than on deterministic worst results with unknown margins, not taking into account the expected frequency of the occurrence of such results.

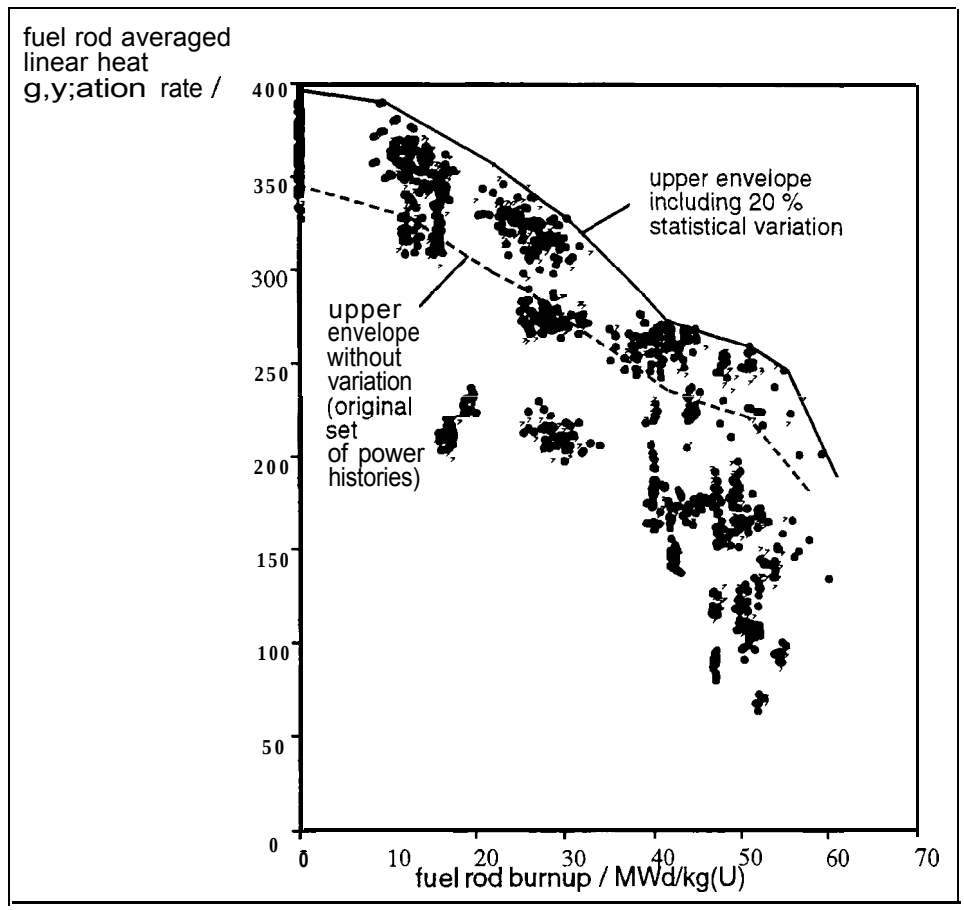

FIG. 6: Comparison of maximum averaged fuel rod linear heat generation rate with and without statistical variation

In Figure 7, two examples of maximum fuel rod internal pressure distributions are shown for two different plants A/B, containing fuel rods with/without lower plenum. The corresponding burnup distributions, depending on the individual optimized reload strategy for each plant, include values up to 69 resp. 67 MWd/kg(M).

From the results of the Monte Carlo calculations (several thousand runs with CAR0 in each case) it can be derived, that the internal pressure of a certain limited number of rods exceeds the coolant pressure with a certain probability, but the small number of extreme values of the distribution do not violate the design criteria.

5. STATUS OF INTRODUCTION INTO LICENSING

The confirmation of the statistical methodology as an appropriate tool for design analysis was given by the fact, that this methodology has been accepted by our customers, see e.g. [3], and successfully applied for licensing. The Swiss Federal Nuclear Safety Inspectorate (HSK) published its positive assessment of our methodology in [17].

Up to now licensing analyses with statistical methods have been performed for 10 different plants in 3 countries, for PWR and BWR, and for UO_2 and MOX fuel rods. Another analysis, using statistical methods, the calculation of core damage extent during a hypothetical loss of coolant accident, is currently being licensed.

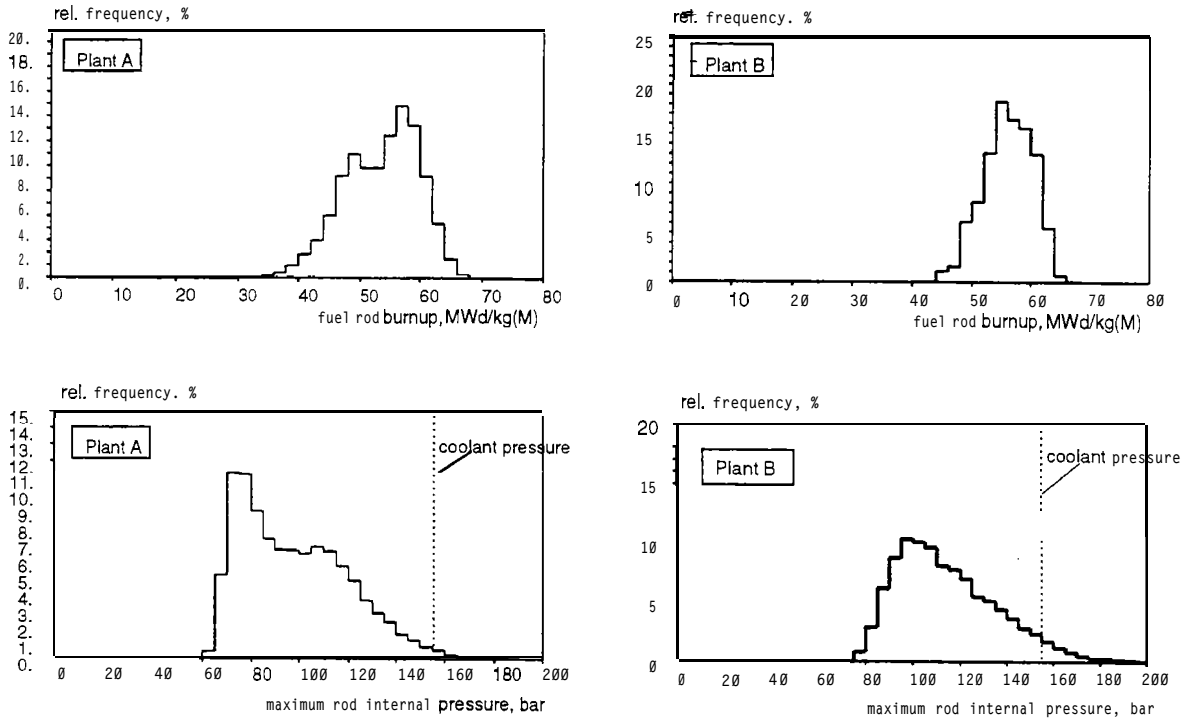

FIG. 7: Frequency distributions of fuel rod burnup and maximum rod internal pressure for MOX fuel rods in two plants

The introduction of the new methodology revealed interesting discussions with customers and licensing authorities which led to improvements and further development of the statistical methods.

6. SUMMARY AND OUTLOOK

The advantages of the fuel rod design using statistical methods in connection with a modern fuel rod code (burnup dependent fuel thermal conductivity, mechanistic fission gas release model and validation against a data base up to high burnups) have been largely discussed in [15].

The improvements of fuel rod design consist of:

- consistent treatment of code uncertainties and distributions of fabrication parameters
- possible sensitivity assessment of fabrication parameter effects and power history influences
- realistic assessment of margins, especially for higher burnups and more demanding core management strategies, e.g. for MOX fuel
- possible on-line fuel rod design with core management calculations
- possible extension to class-2 and core damage extent analysis
- possible consistent extension to dry storage design analyses

The statistical design methodology is a powerful tool to realistically assess the behavior of the fuel rods in a reactor core. It has the capability of characterizing the degree of conservatism through the statistical evaluation of numbers of fuel rods coming close to a design limit or by making statements about the statistical certainty for the actual occurrence of extreme cases. Since fuel insertion conditions are becoming more and more demanding as well as knowledge about performance affecting mechanisms and experience data bases has been increasing, the classical deterministic method should be replaced by a statistical one which provides more, especially more differentiated information about fuel rod behavior.

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FABRICATION AND PERFORMANCE TESTING OF CANDU MIXED-OXIDE FUEL

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Abstract

AECL's mixed-oxide fuel fabrication activities are performed in the Recycle Fuel Fabrication Laboratories (RFFL) at the Chalk River Laboratories. Since the start-up of the RFFL in the mid-1970s, several fabrication campaigns have been conducted in the facility, producing various types of mixed-oxide (MOX) fuel, which were used for both irradiation and physics testing. More recently, CANDU® fuel bundles containing 0.5 w-t % plutonium in natural uranium, produced in the RFFL, were successfully irradiated in the NRU reactor at powers up to 65 kW/m and to burnups ranging from 13 to 23 MW•d/kg HE. Two of the bundles had power histories that bound the normal powers and burnups of natural UO_2 CANDU fuel (<65 kW/m, burnups of 13 to 15 MW•d/kg HE). These bundles exhibited sheath strain and fission-gas release (FGR) typical of those observed in similarly operated UO_2 fuel. Significantly more grain growth was observed than that expected for UO_2 fuel; however, this increase in grain growth had no effect on the overall performance of the fuel. Two other bundles operated to extended burnups of 19 to 23 MW•d/kg HE. Burnup extension above 15 MW•d/kg HE only had a small effect on FGR.

1. INTRODUCTION

Research and development activities on Pu-containing mixed-oxide (MOX) fuel have been conducted by Atomic Energy of Canada Limited (AECL) at its Chalk River Laboratories (CRL) site since 1960, and they remain a strategic part of AECL's advanced fuel-cycle program. The program includes MOX fuel fabrication development, irradiation testing, post-irradiation examination (PIE), as well as reactor physics and fuel-management studies.

2. CANDU MIXED-OXIDE FUEL FABRICATION

2.1. The RFFL

AECL's mixed-oxide fuel fabrication activities are performed in the Recycle Fuel Fabrication Laboratories (RFFL) at the Chalk River Laboratories (CRL). The RFFL facility is designed to produce experimental quantities of CANDU mixed-oxide fuel for reactor physics tests or for demonstration irradiations [1].

Since the start-up of the RFFL in the mid-1970s a number of fuel fabrication campaigns have been conducted in the facility, producing various quantities of fuel with different compositions including MOX (Table I). The fuel elements and bundles were used for test irradiations in the NRU experimental reactor and for physics tests in the zero-power ZED-2 reactor. To date, about 5000 individual fuel elements, equivalent to over 160 bundles and containing close to 3 t of MOX, were fabricated in the RFFL [2].

Subject to restrictions imposed by the presence of Pa (essentially all operations are done inside ventilated and filtered glove boxes and fume hoods), the processes employed in the RFFL follow conventional natural-UO_2 practice (Figure 1). Weighed amounts of the starting powders (UO_2 or ThO_2 and PuO_2) are milled either separately or as a master mix using a vibratory mill. Blending could then be achieved using the high-intensity mixer followed by a turbula blender. The blended MOX powder is pre-pressed into compacts, which,,, in turn, are fed into a granulator. The resulting free-flowing granules are then suitable for final pressing into green pellets by an automatic hydraulic press. The green pellets are loaded into a batch furnace, where sintering is performed under a dilute hydrogen cover gas. Sintered pellets are then centreless ground to a specified diameter and surface finish. Acceptable pellets are loaded into sheaths that are subsequently end-closure welded using a tungsten-inert gas (TIG) welding system. The sealed elements are helium leak-tested, scanned for surface alpha contamination, and dimensionally inspected before being assembled into bundles.

2.2. Inspection Techniques on MOX Fuel

Standard inspection techniques-such as finished dimensions, surface finish, and immersion density of the pellets-are applied to obtain physical characteristics of the MOX fuel. Also, standard metallographic procedures are used to measure microstructural grain size of sintered pellets. Chemical analytical methods are employed to determine fuel properties, including oxygen-to-metal ratios, assay and isotopic composition of fissile components, and impurity contents of the finished pellets.

TABLE I. FUEL FABRICATION CAMPAIGNS CONDUCTED IN THE RFFL

Experiment	DATE	FUEL TYPE	QUANTITY (kg MOX)
BDL-419	1979-80	(U, 0.5% Pu)O_2	15, 36-element bundles (320 kg)
BDL-422	1981-83	(Th, 1.75% Pu)O_2	6, 36-element bundles (120 kg)
BDL-43 0	1982	Natural ThO_2	1, 36-element bundle (20 kg)
WR1-1012	1982	(Th, 1.8% ^{235}U)O_2	2, 21-element bundles (20 kg)
WR1-1012	1982	(Th, 2.3% Pu)O_2	2, 21-element bundles (20 kg)
WR1-1010	1982-85	(Th, 2.3% Pu)O_2	1332 elements (650 kg)
BDL-432	1986-88	(Th, 1.4% ^{233}U)O_2	1350 elements (700 kg)
ZED2-96	1996-97	(U, 0.3% Pu)O_2	37, 37-element bundles (810 kg)

One inspection technique of interest is alpha auto-radiography that is used in combination with image analysis to determine Pu particle size and distribution. This method provides a quick and practical means of quantitatively determining the extent of plutonium homogeneity of MOX fuel in a production environment. A typical image from an auto-radiograph is digitized and enhanced for analysis, as shown in Figure 2. Information on particle size and inter-particle distances are then obtained to determine a measure of Pu homogeneity.

Another use of the image analyzer is to employ the local composition differences to convert the contrast between the plutonium-rich particles and the matrix into gray-levels. If a relationship between the gray-level and composition can be established, plutonium and

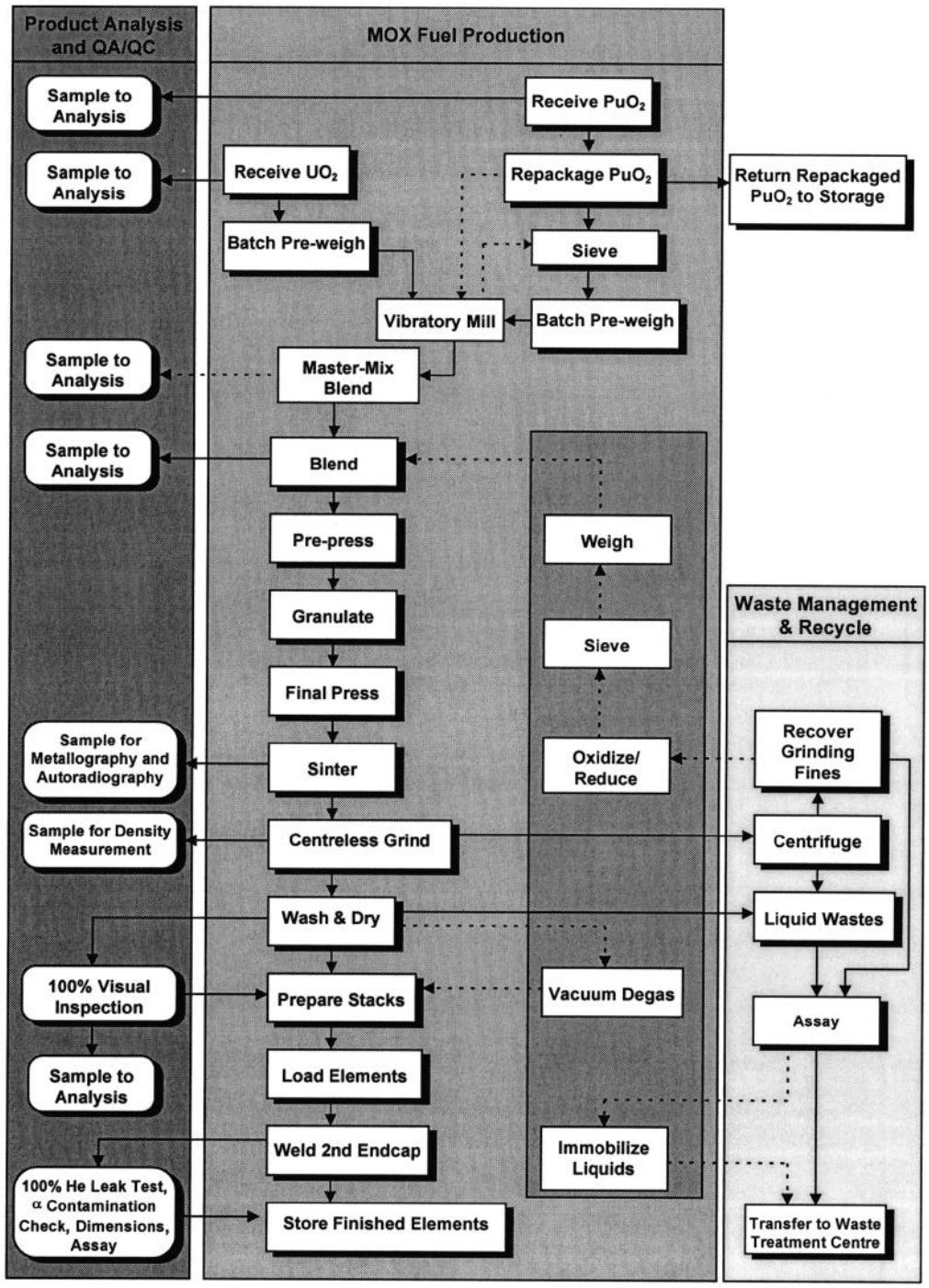


FIGURE 1. Flow sheet for the MOX fuel fabrication process employed in the RFFL.

uranium distributions can be determined directly by image analysis, making it practical and economical for a MOX fuel fabrication facility. Development work is being conducted at AECL to accomplish this procedure.

Some MOX fuel samples were analyzed using a quantitative X-ray dispersive spectrometric (WDS) technique [3]. The analysis used a WDS spectrometer attached to the shielded JEOL-840 scanning electron microscope (SEM) at CRL. Point analysis with a 2-μm interval was conducted diametrally across the Pu-rich particles to obtain the uranium and plutonium distribution profiles, which all exhibited a similar shape. A compositional

transition band, ~10 μm in width, was observed between the particle and the matrix, indicating plutonium and uranium interdiffusion had occurred. The individual measurements are consistent and accuracy is satisfactory.

The gray-scale trace of a Pu-rich particle obtained from image analysis of an autoradiograph is compared to the concentration profile obtained from WDS in Figure 3. As can be seen, the correlation between the two profiles is very good. Further work is being conducted to compare uranium and plutonium concentrations in particles with corresponding alpha autoradiographic images to provide a bench mark for routine alpha autoradiographic analysis.

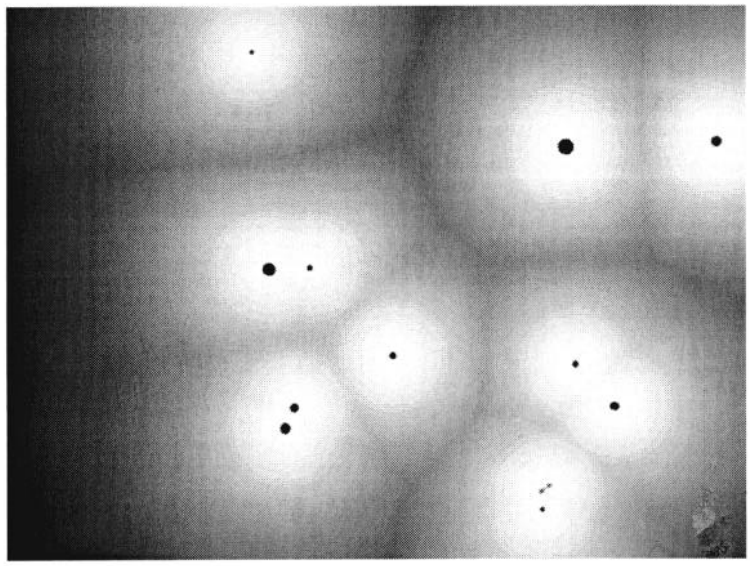


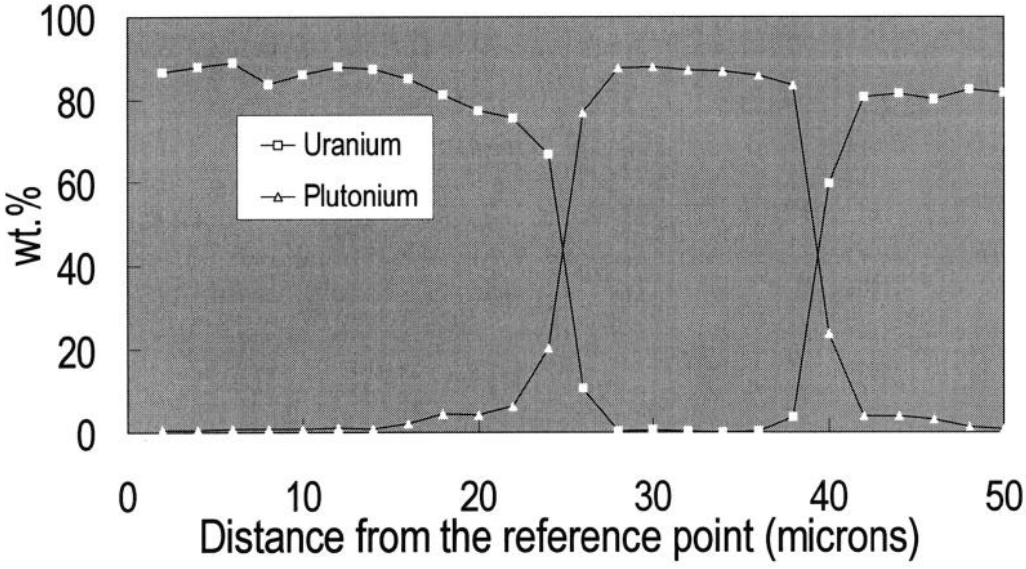
FIGURE 2. Photograph showing a digitized and enhanced image of a typical alpha autoradiograph of an unirradiated MOX fuel. The dark spots are Pu-rich areas.

3. CANDU MIXED-OXIDE FUEL PERFORMANCE TESTING

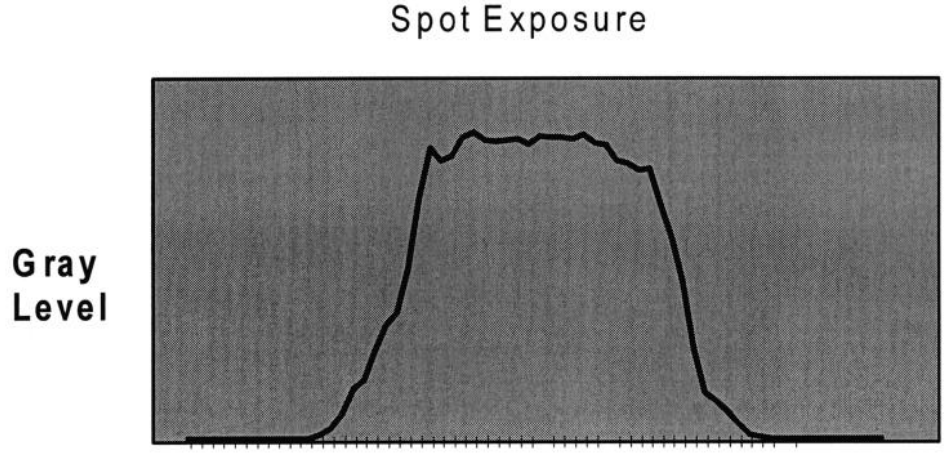
3.1. The BDL-419 Experiment

Irradiation testing and PIE of Pu-containing MOX fuel at AECL have progressed from testing of several fuel elements—to investigate basic ceramic properties of MOX fuel—to currently demonstrating the multi-bundle irradiation performance of Canadian-fabricated MOX fuel. One of the more recent tests is the BDL-419 experiment. It has involved the fabrication, irradiation, and post-irradiation examination of MOX fuel bundles of the same 37-element geometry as is currently used in commercial CANDU power reactors. Details of this experiment, including the irradiation history and PIE results, are reported elsewhere [4]; the highlights are discussed here.

The main objectives of the BDL-419 experiment were (a) to demonstrate that (U, Pu)O_2 fuel fabricated in the RFFL is capable of sustaining powers and burnups typical of CANDU UO_2 fuel, that is, at peak powers <65 kW/m to burnups of approximately 10 MW•d/kg HE; and (b) to investigate the performance of CANDU MOX fuel at extended burnups (>18 MW•d/kg HE).



(a)



(b)

FIGURE 3. Plots of (a) uranium and plutonium concentration profile obtained by microprobe spot analysis across a Pu-rich area, and (b) a comparable gray-scale trace of digitized and enhanced image of a similar area as it appears on an alpha auto-radiograph.

3.2. Fuel Design And Irradiation

The fuel elements for BDL-419 were fabricated in the RFFL [1,2]. The (U, Pu)O_2 pellets, produced by blending natural UO_2 with PuO_2, contained 0.5 wt % fissile plutonium in heavy element. The elements were assembled into bundles that were essentially the same as those used in the Bruce commercial power reactors.

A summary of the PIE results of four BDL-4 19 bundles-designated ABB, ABC, ABD and ABE-is presented here. Bundles ABD and ABE operated at powers up to 65 kW/m and to burnups ranging from 13 to 15 MW•d/kg HE, thus bounding the normal powers and burnups of natural-UO_2 CANDU fuel. Similar powers were observed in bundles ABB and ABC, but to extended burnups ranging from 19 to 23 MW•d/kg HE. These bundles were irradiated under conditions similar to those of commercial CANDU power reactors. A summary of power and burnup data is shown in Table Il.

TABLE II. SUMMARY OF POWER HISTORY, STRAIN AND FISSION-GAS RELEASE DATA FOR BDL-4 19 MOX FUEL

Bundle Identity	Measured Outer-Element Burnup (MW•d/kg HE)	Outer-Element Power at BOL (kW/m)	Outer-Element Midpellet Strain (%)	Outer-Element Fission-Gas Release (%)
ABB	18.7 - 20.4	50 - 55	-0.1 to 0.0	4 - 5
ABC	22.1 - 22.8	60 - 62	-0.1 to +0.1	12 - 13
ABD	13.0 - 13.3	63 - 65	-0.2 to +0.2	lo- 11
ABE	14.4 - 15.0	54 - 56	-0.3 to 0.0	7 - 9

3.3. **Results of the PIE**

Midpellet residual sheath strains of -0.3% to +0.2% were observed in the outer elements of the four BDL-419 bundles (Table II). These strains are within the range that is normally observed in CANDU UO_2 fuel [5].

Fission-gas release (FGR) of 4% to 13% was observed in the outer elements of the four BDL-4 19 bundles. This FGR is within the range that is expected for similarly designed and operated UO_2 fuel [5-7]. As in the case of UO_2 fuel [7], the MOX fuel FGR dependence on burnup is weak relative to the effect of power.

Alpha auto-radiography of an outer-element fuel pellet cross-section illustrated that the periphery of the fuel retained the as-fabricated inhomogeneous microstructure of the fuel, containing regions with plutonium-rich particles as well as regions low in plutonium content. Plutonium homogenization was observed in the central region of the fuel at powers >55 kW/m, and was accompanied by columnar grain growth. Columnar grain growth is not normally observed in CANDU UO_2 fuel below powers of 65 kW/m. The progression in burnup from 13 to 23 MW•d/kg HE did not appear to have any effect on the observed microstructural changes; this finding indicates that the degree of grain growth observed was due to high power operation at BOL (beginning-of-life), not burnup extension.

The degree of grain growth observed in the BDL4 19 MOX fuel relative to the degree expected of UO_2 fuel is indicative of higher operating temperatures or different grain-growth kinetics, or of both. The columnar grain growth observed in the BDL-419 MOX fuel does not appear to have contributed to increased strain or FGR, both strain and FGR- as noted above-were in the normal range for comparable UO_2 fuels.

4. SUMMARY AND CONCLUSIONS

Since the start-up of the RFFL in the mid-1970s several fabrication campaigns have been conducted in the facility, producing various types of CANDU mixed-oxide fuel, which

were used for both irradiation and physics testing. To date, about 5000 individual fuel elements, equivalent to over 160 bundles and containing close to 3 t of MOX fuel, were fabricated in the RFFL.

Development work is being conducted at AECL on MOX fuel fabrication and characterization. One characterization technique is alpha auto-radiography that is used in combination with image analysis to determine Pu particle size and distribution. Using a quantitative X-ray dispersive spectrometric (WDS) technique, a correlation between the gray-level and composition can be established, and plutonium distributions can be determined directly by image analysis. This method would provide a practical and economical means of quantitatively determining the extent of plutonium homogeneity of MOX fuel in a production environment.

CANDU mixed-oxide fuel bundles that contained 0.5 w-t % plutonium in natural uranium were successfully irradiated from outer-element BOL powers of 50 to 65 kW/m to burnups ranging from 13 to 23 MW•d/kg HE. The power histories observed in some bundles bound the normal powers and burnups of natural-UO2,CANDU fuel (up to 65 kW/m, to burnups ranging from 13 to 15 MW•d/kg HE). These bundles exhibited sheath strain and FGR typical of those observed in similarly designed and operated UO_2 fuel. Significantly more grain growth was observed than that expected for UO_2 fuel; however, this increase in grain growth had no effect on the overall performance of the fuel.

Other bundles operated to extended burnups of 19 to 23 MW•d/kg HE at powers similar to those at which other bundles operated. Burnup extension beyond 15 MW•d/kg HE had no apparent effect on sheath strain and grain growth, and only had a small effect on FGR (similar to that observed in UO_2 fuel). This extended burnup had no effect on the performance of the bundles.

The BDL-4 19 tests demonstrate that MOX fuel produced in the RFFL is capable of sustaining powers and burnups typical of natural-U02 fuel currently operating in CANDU reactors and that its performance at extended burnups is similar to that expected of UO_2 fuel.

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RIA TESTS IN CABRI WITH MOX FUEL

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Abstract

Three MOX-fuel tests have been successfully performed within the framework of the CABRI REP-Na test program. From the experimental findings which are presently available, no evidence for thermal effects resulting from the heterogeneous nature of the fuel can be given. There are very clear hints however that fission gas effects are enhanced with regard to the behaviour of UO_2. The clad rupture observed in REP-Na 7 is of different nature than the failures observed in Cabri tests with UO_2 fuel. Failures of UO_2 fuel rods only occurred when the clad mechanical properties were severely affected by the presence of hydride blisters, while in REP-Na 7 a clear indication is made that the loading potential of the MOX fuel pellets was high enough to break a sound cladding. Concerning the transient fuel behaviour after reaching the critical heat-flux under reactor typical conditions (pressure, temperature and flow), no data base could be provided by the tests in the present sodium test loop (as for the UO_2 fuel behaviour). The IPSN project to implement into the Cabri reactor a pressurised water loop which will allow to simulate the complete RIA accident sequence under PWR reactor typical conditions, aims at providing this missing data base.

1 - INTRODUCTION

Rapid power excursions, which may result from reactivity transients or Reactivity-Initiated Accidents (RIAs), may represent an unacceptable threat to the safety of nuclear power reactors. The ejection of a control rod assembly, under the effect of the reactor system pressure following the rupture of the housing of a control rod drive, has been chosen as a design-basis accident for Pressurized Water Reactors (PWRs) in most countries.

Based on experimental programs that were performed in the USA up to the end of the 1970s RIA safety criteria were defined and are still valid in several countries, including France (for high burnup fuel these criteria are presently considered as inadequate). The data base for justification of the validity of these criteria was originally limited to burnup up to 33GWd/t in mean assembly and for UO_2 fuel and no additional safety experiments were performed when licensing extensions were approved by the regulatory authorities.

The continuing search for economic competitiveness and also for the optimized use of nuclear fuel are the driving forces for changes in fuel management modes and in particular the trend to still higher burnup. In addition, in several countries the use of plutonium as fissile material (MOX fuel) has been introduced. High burnup phenomena, like cladding corrosion, fission product accumulation, and structural changes of the fuel as well as heterogeneous nature and low reactivity decrease of the MOX fuel, are liable to reduce the safety margins which were known to be large at the time of the definition of the criteria.

An important research program was initiated in France by'IPSN in collaboration with EDF at the end of the 198Os, essentially the REP-Na tests in Cabri including three MOX-fuel tests at different burnup levels. Furthermore, a set of support tests for key properties and phenomena, and the development of the specific RIA fuel behaviour code, SCANAIR are part of this program. More recently, the research effort has been extended to neutronic calculations with use of coupled thermal-hydraulic and 3D neutronic kinetic computer codes, with the aim to improve the evaluation of the safety margins by comparison with test results. The entire research activity is characterized by a significant international cooperation [6,10].

In the present paper are presented the specific aspects of the three MOX-fuel tests of the CABRI REP-Na program, the test objectives, the choice of the test parameters, the major results and the present state of understanding.

2 - TEST OBJECTIVES

The fuel test program CABRI REP-Na has been started in 1993 with the aim to extend the data base of Light Water Reactor fuel behaviour under the conditions of the reactivity initiated accident (RIA) of PWRs, i.e the ejection of a control-rod bundle under hot zero-power conditions.

The main objective of this program was the investigation of potential high burn-up effects and the verification of the RIA safety criteria for classical UO_2 fuel, corresponding to the request of the french safety authority DSIN, in view of the intention of Electricité de France (EDF) to increase the burnup limit from 47 to 52 GWd/t (mean assembly).

In addition and beyond the UO_2 objectives, the investigation of MOX fuel behaviour under RIA conditions was included in the definition of the REP-Na test matrix, in anticipation of future licensing requests concerning the behaviour of high burnup MOX fuel under RIA conditions [1].

The most commonly used MOX fuel is elaborated through the MIMAS fabrication process which is characterised by the mechanical mixing of natural or depleted UO_2 with a masterblend powder of $(UPu)O_2$. This process produces a slightly heterogeneous final product with mixed oxide agglomerates (or clusters), imbedded in the UO_2 matrix. The as fabricated plutonium concentration in the clusters is the one of the masterblend, approximately 30%.

The mean size of these agglomerates is rather small, - 20 microns, however the fabrication specifications allow that a small but non-negligeable number of the clusters reaches the size of several hundreds of microns.

The heterogeneity of the MOX represents the most significant difference with regard to UO_2 fuel. Its potential effect under the conditions of the fast transient heating under the RIA has been considered and investigated since long time [2,3,4]. The result of these investigations is taken into account in the MOX fuel fabrication specifications.

The early concern however was related only to the aspect of the thermal behaviour of the plutonium-rich clusters and only fresh fuel experiments were performed. More recent results, from CABRI UO_2 and from NSRR tests, give rise for arguments which lead to postulate a specific high burnup MOX behaviour, resulting from fission gas effects.

The three MOX tests in CABRI were therefore defined in such a way that possible effects, both thermal and high burnup clad - loading effects could be investigated:

- In the test REP-Na 9 with a 2 cycle rod, the mean burnup is low (28 GWd/t). In the clusters the plutonium content is still high and the cluster burnup is already significant and, accordingly a quite important fission product inventory has build up. At this level, the combined thermal and fission gas effect might be maximised.
- In the 3 cycle test rod REP-Na 6 (47 GWd/t) the transient overheating of the clusters has decreased and transient loading resulting from fission gas becomes predominating.
- In REP-Na 7 finally, the mean fuel burn-up reaches 55 GWd/t and, at this level, a large fraction of the fissions under nominal operation and in the transient is to be attributed to the newly formed plutonium, i.e. in the bulk of the UO_2 matrix. The transient energy deposition becomes more

homogeneous but still, the original clusters contain high amounts of fission gas in the intergranular bubbles and inside the neighbouring porosity.

The expected contributions from thermal effects firstly and from structural and gas effects secondly are therefore changing in a characteristic manner from REP-Na 9 to REP-Na 7. At the time of the definition of the tests it was expected that the global and detailed test results would allow to evaluate, at least qualitatively, whether the thermal or the fission gas effect resulting from the fuel heterogeneity would be predominant. It appears at present that this goal has been reached, as will be seen from this paper.

Table 1: The MOX-fuel tests of the Cabri REP-Na test matrix:

Test ('date))	*Tested rod*	*Pulse (ms)*	*Energy at pulse end (cal/g)*	*Clad Corrosion (μ ZrO_2)*	*Results and remarks*
IUa-6 (3/96)	**EDF MOX, 3c span 5** 47 **GWd/t**	**40**	**165 (at 1.2 s) (690 J/g)**	**35**	**No** rupture **Hmax = 148 cal/g** $\Delta\phi/\phi$: **2.65 %** (mean max) FGR = 21,6 %
I?la-7 **(2/97)**	**EDF MOX, 4c span 5 55 GWd/t**	**40**	**175 (at 1.2 s) (732 J/g)**	**50**	**Rupture at 120 cal/g Hmax = 140 cal/g Pressure peaks Fuel dispersal Examination currently carried out**
IUa-9 (4/97)	**EDF MOX** 2 c **span 5 28 GWj/t**	**34**	**241 (at 1.2 s) (953 J/g)**	**<20**	**No** rupture **Hmax = 210 cal/g** $\Delta\phi/\phi$ **= 7.3 % mean max** FGR ≈ **35% (to** be confirmed) Examination currently carried out

***Note** : the burn-up indicated in the table is the maximum burn-up of the test rod/et : the father rod has a burn-up 10 to 72% lower.*

In table 1 are presented the major test parameters and a few significant experimental results.

All test rods are refabricated from sections of industrial MOX-fuel rods, irradiated in commercial Nuclear Power Plants. The refabrication procedure in all cases was the FABRICE routine [7] as in all the REP-Na tests. In all cases also, the father rod was sectionned at the level of span 5, the penultimate intergrid span at the upper end of the fuel rod. At this level, the burnup is rather constant and the outer clad corrosion is close to rod maximum. The refabricated rodlets were filled with He at 0.3 MPa pressure, equivalent to the sodium pressure in the coolant channel. Last but not least, all MOX tests were performed with a power-pulse-width close to 40 ms, a value which is a better approach of reactor pulse when loaded with MOX fuel.

3 - MAIN RESULTS OF THE CABRI-REP-Na MOX FUEL TESTS

Three tests have been realized in the CABRI-REP Na programme, using MOX fuel rods with various burn-up levels (28, 47 and 55 GWd/t) and standard Zircaloy-4 cladding.

The main characteristics and results are given in table 1.

3 - **1. REP-Na 9 TEST**

The REP-Na 9 test used a 2-cycle MOX fuel rod irradiated in St Laurent B1 EDF power plant. The test rod was reconditioned from the 5th span of an industrial rod with 28 GWd/t burn-up and a low clad corrosion ($\approx 10\,\mu m\ ZrO_2$).

The power transient of 34 ms half width did not lead to rod failure although the high energy injection (241 cal/g at 1.2 s) resulted in a maximum mean fuel enthalpy of 210 cal/g. An evaluation of the maximum fuel temperature with the SCANAIR code [5] with homogeneous description indicates that it probably reached in the periphery 2400°C in the first ms and 2700°C later on, in the central part. This high mean maximum temperature may have produced local melting of a fractional of the higher enriched MOX clusters.

The maximum fuel and clad elongations are 8 mm and the residual clad elongation amounts to 5 mm.

The transient evolution of the inlet and outlet sodium flow rates showed rapid variations (« TOP effect ») which is known to result from the transient radial deformation of the rod, the thermal expansion of the sodium and the heating of the outer channel wall.

The profilometry of the rod after test showed a very high mean plastic circumferential strain (the maximum value ever obtained in the CABRI REP-Na tests) with maximum strain near pellet ends. At the peak power level, the hoop strain reached 8% at the pellet edge and 6% in the pellet middle height, showing an hourglass type pellet deformation with increase of the primary local strains up to 100 µm in diameter (fig1).

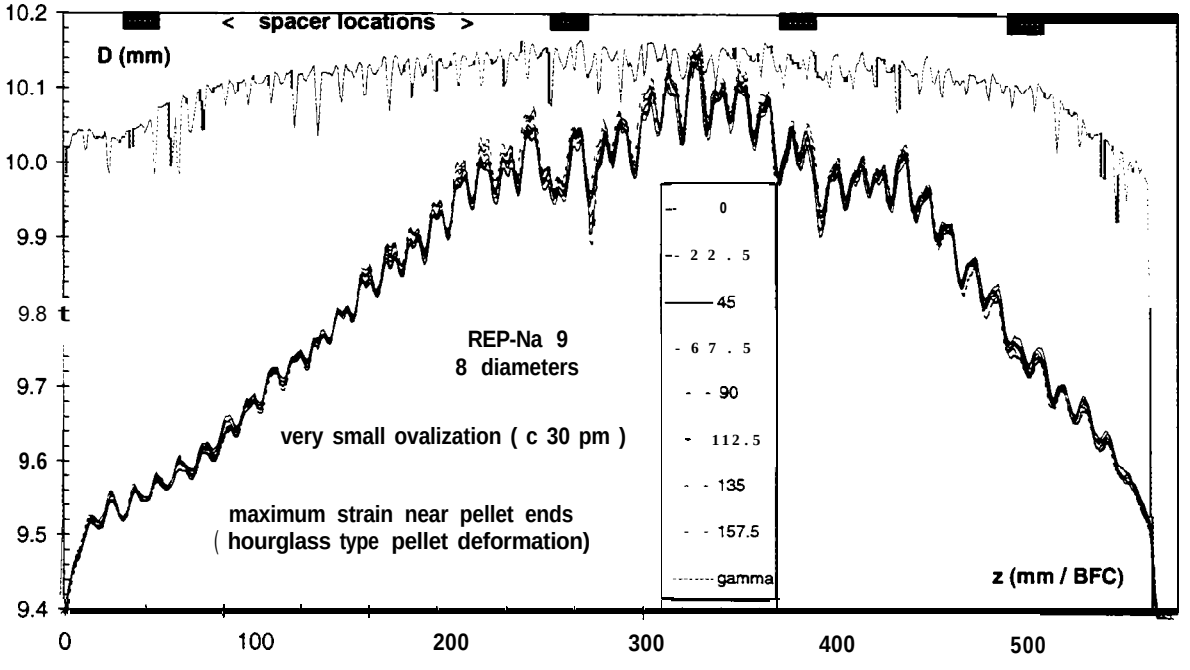


Fig. 1 : REP-Na 9 profilometry : rod diameters versus axial position

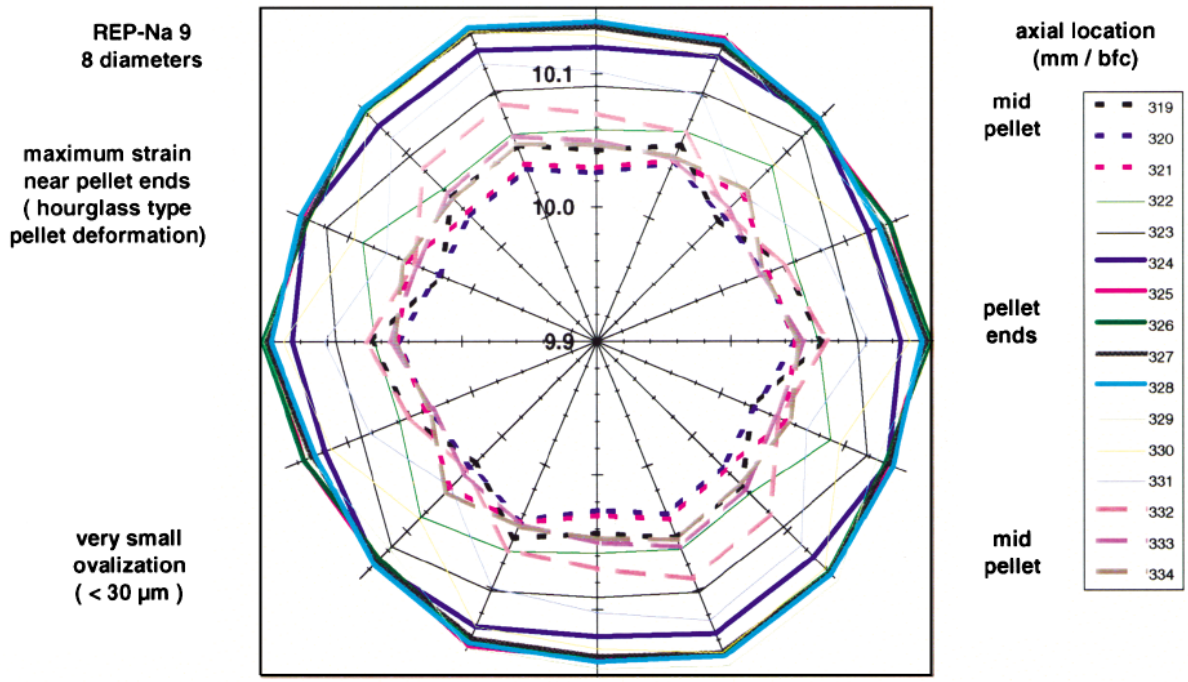


Fig. 2 : REP-Na 9 profilometry : polar representation of rod diameters versus azimuth

Based on the profilometries on 8 diameters, no significant ovalization (< 30 µm) has been evidenced (fig.2).

The non destructive examinations (gamma scanning and x-ray radiography) do not show any filling of the dishings (as observed in REP-Na 2 [5]) by the fuel, but this has to be confirmed by the destructive examinations.

A preliminary information from the rod piercing indicates a high level of gas release with probably a significant contribution of Helium release. Rough estimation leads to a fission gas release of around 35%, to be confirmed after the analysis of the released gas.

The REP-Na 9 test with MOX fuel can be compared to the REP-Na 2 test performed with an UO2 rod and a similar energy deposit : BR3 rod at 33 GWd/t, low corrosion thickness (4 µm), power pulse of 9.5 ms half width with 211 cal/g total energy injected and a maximum mean fuel enthalpy evaluated to be 210 cal/g.

Concerning the clad hoop strain, the REP-Na 2 test also resulted in an hourglass shape with a mean value of 3.5% at maximum power level (4.1% at pellet edge and 2.9% at mid-pellet) without any ovalization.

Such high level of cladding deformation in both tests is explained by the combined effect of the fuel thermal expansion and by fission gas induced swelling.

This last phenomenon is linked to vacancy diffusion induced by fission gas bubble pressure increase and is activated when the fuel temperature remains higher than 2100 K during a sufficient time which is the case in the center part of the pellets in both tests with high energy injection.

The hourglass shape of the clad straining as shown by profilometry is a clear confirmation of the fuel loading under parabolic radial temperature profile as obtained in the late phase of the transient (fuel cooling down phase).

In spite of the similar energy injection, the higher deformation obtained in REP-Na 9 compared to REP-Na 2 [5] can be explained by the different mechanical properties of the cladding material : indeed, in the temperature range covered by the tests (maximum inner and outer clad temperatures respectively around 900°C and 500°C), the yield stress of the standard zircaloy-4 of REP-Na 9 cladding is strongly reduced in opposition to BR3 rod cladding (by a factor ≈ 2).

Another topic for comparison is the fuel behaviour through the filling of the dishings : complete in REP-Na 2 and attributed to fuel visco-plasticity at temperature close to melting, not clearly evidenced in REP-Na 9. If such result is confirmed by the REP Na 9 destructive examinations, it seems to be in opposition to what is observed in slow power ramps where the MOX fuel shows an enhanced creep behaviour compared to UO_2.

Lastly, the fission gas release rate (FGR) is found much higher with the MOX fuel rod than with the UO_2 rod : however, it is important to notice the EPMA results of a 2 cycles MOX fuel rod which tend to indicate an amount of around 25 % of fission gases in intergranular and porosity bubbles (not seen by EPMA) at the end of irradiation (high gas retention inside the porosity in $UPuO_2$ agglomerates). This high quantity is thus available to be released during the power transient in correlation with the grain boundary separation occurrence as already deduced from the analysis of high burn-up UO_2 fuel submitted to a RIA [5]. The total FGR result after the power transient, if confirmed, is thus consistent with the assumption of the contribution of initial porosity and intergranular bubbles associated to grain boundary separation and is increased by intragranular bubbles migration at such high temperature levels as in REP-Na 2 and REP-Na 9.

3 - 2. REP Na 6 TEST

The REP-Na 6 test has been performed with a 3 cycles MOX fuel rod irradiated in the St Laurent B2 EDF power plant. The test rod reconditioned from the 5th span of a reactor rod was characterized by a burn-up level of 47 GWd/t and a maximum clad corrosion thickness of 35 µm.

The CABRI power transient of 40 ms half width injected 165 cal/g at the peak power level (at 1.2s) resulting in a maximum mean fuel enthalpy of 148 cal/g (SCANAIR evaluation).

The rod did not fail but a significant residual clad straining (fig. 3) has been obtained (2.65% mean value at peak power node, PPN) together with a strong ovalization (100 µm in diameter at PPN, fig. 4).

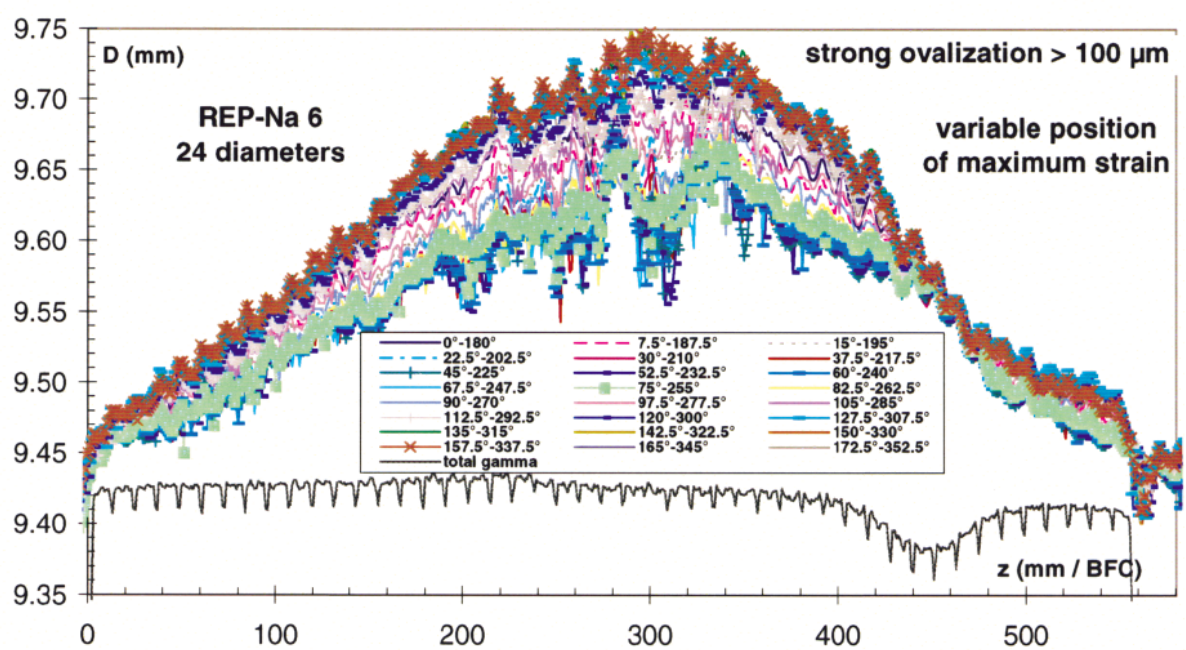


Fig. 3 : REP-Na 6 profilometry : rod diameters versus axial position

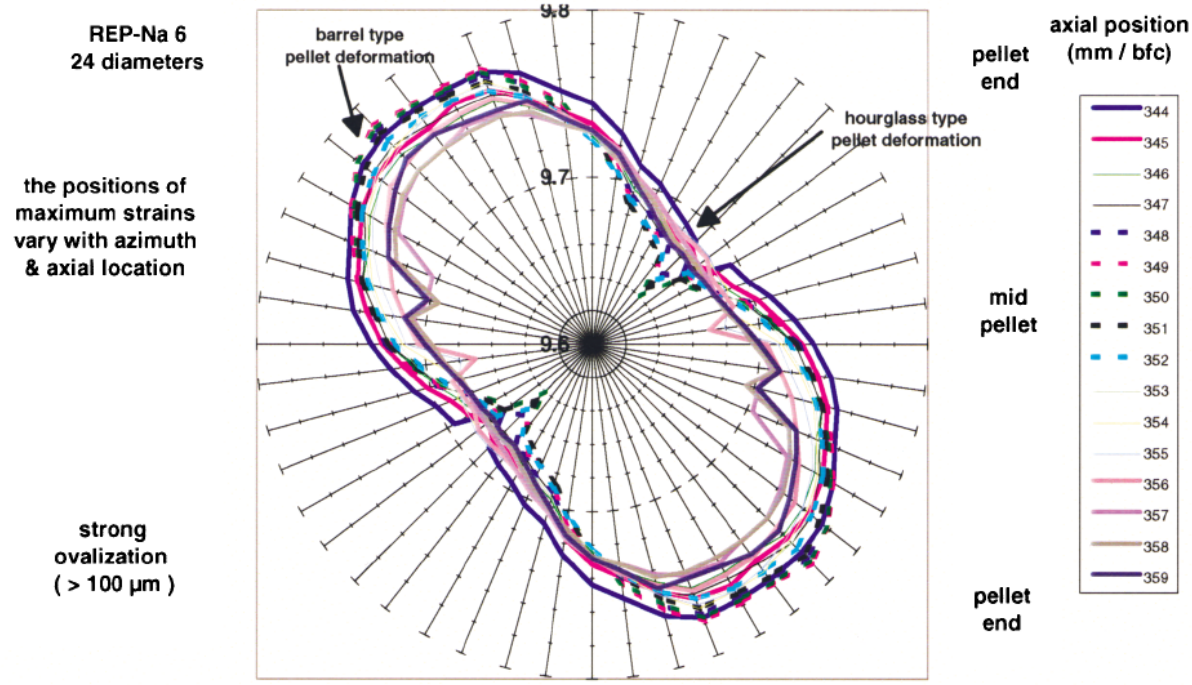


Fig. 4 : REP-na 6 profilometry : polar representation of rod diameters versus azimuth

The origin of such ovalization is not yet understood and studies are underway in order to identify whether this can be due to fuel or clad heterogeneity (microstructure, gas retention, corrosion), or to asymetrical energy deposit in the CABRI reactor (neutron absorption due to instrumentation devices). The fact that both tests with higher energy deposit and lower burn-up such as REP Na 2 and REP Na 9 do not lead to straining with ovalization is also to be considered (deformation with contribution of the fuel center part, more homogeneous).

The measurement of the zirconia thickness after test showed some transient spalling close to peak power node (PPN) and to the top of fissile length : this phenomenon already observed in some REP-Na UO_2 tests (REP-Na 3 - Na 4) is linked to the transient clad straining and is favoured by the presence of a thick initial oxide layer as in REP Na 4 (80 µm initial thickness, very large spalling due to CABRI test, [5]).

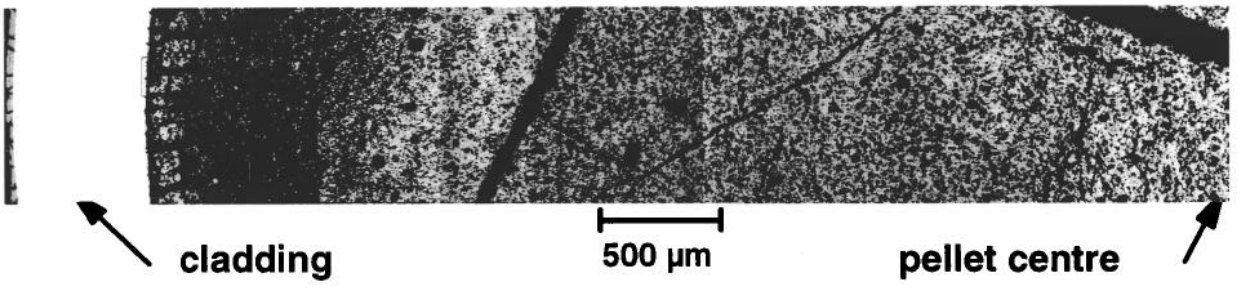
The fission gas release due to the transient amounts to 21.6 % of the retained gases : this high release is consistently understood if we take into account the results of EPMA examinations before the test indicating that a large quantity of gases is present in intergranular and porosity bubbles already at the end of irradiation.

In addition, the metallographic examinations performed on radial cuts (see fig.5) clearly show that the power transient resulted in a high degree of grain boundary separation in the UO2 matrix, particularly at the pellet periphery as previously seen in the other REP-Na tests.

Such effect is linked to the radial power distribution leading to maximum temperature in the outer zone of the pellet in case of rapid power transients. The grain boundary fragmentation together with clad straining allow gas to escape through paths and contributes to the high level of transient fission gas release.

Another point issued from post test examinations, is the evidence in the central part of the pellet, of inter and intragranular gas precipitation with presence of channels at grain boundaries which confirms the contribution of the center fuel part to the fission gas release.

Finally, the examinations did not highlight any specific thermal behaviour of the Pu aggregates which did not undergo local melting (even when located at the pellet periphery), nor significant morphology modification as compared to pre-irradiation state.



REP-Na 6 micrography along a radius

detail at 90% of the pellet radius showing fragmentation of the UO2 matrix

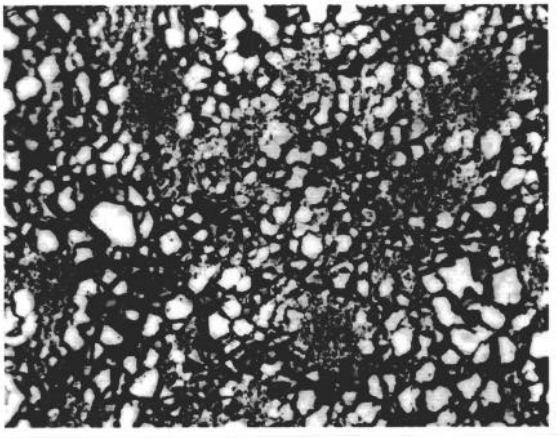


Fig. 5 : REP-Na 6 radial cut at peak power location

3 - 3. REP-Na 7-TEST

The REP-Na7 test has been performed using a 4-cycle MOX fuel rod (55 GWd/t) reconditioned from the 5th span of a PWR rod irradiated in Gravelines 4 power plant and with a clad corrosion thickness of 50 µm.

The neutron-radiography before test did not exhibit any hydride accumulation (so called « blister ») nor spalling of the oxide layer.

The power transient of 40 ms half width led to rod failure (at 452 ms) for an injected energy of 109 cal/g at peak power node (PPN).

According to calculations with the SCANAIR code [5] the rod failed at the time when a mean fuel enthalpy of 120 cal/g at PPN was reached.

The failure was immediately followed by a strong sodium flow ejection and high pressure peaks in the channel (200 b at inlet, 110 b at oulet) and by the voiding of the coolant channel (fig.6).

From the microphones and flowmeter signal analysis, the failure has been located around PPN (26 cm from bottom of fissile length). However, the hodoscope did not give any evidence of fuel motion at that time due to its low sensitivity with low enriched PWR fuel.

A second event, in the lower part of the test rod, occurred 18 ms later (seen by hodoscope, flowmeter, pressure transducers), clearly indicating fuel motion in the lower part of the channel : at this time, which could be considered as the latest one for the onset of fuel ejection, the maximum fuel enthalpy is evaluated to be 130 cal/g.

The large amount of fuel motion is confirmed by the low residual sodium flow (5% of its initial value) indicating an almost complete channel blockage. This point is corroborated by the non-destructive examinations showing loss of fuel in the lower part of the fissile column and fuel relocation in the filters.

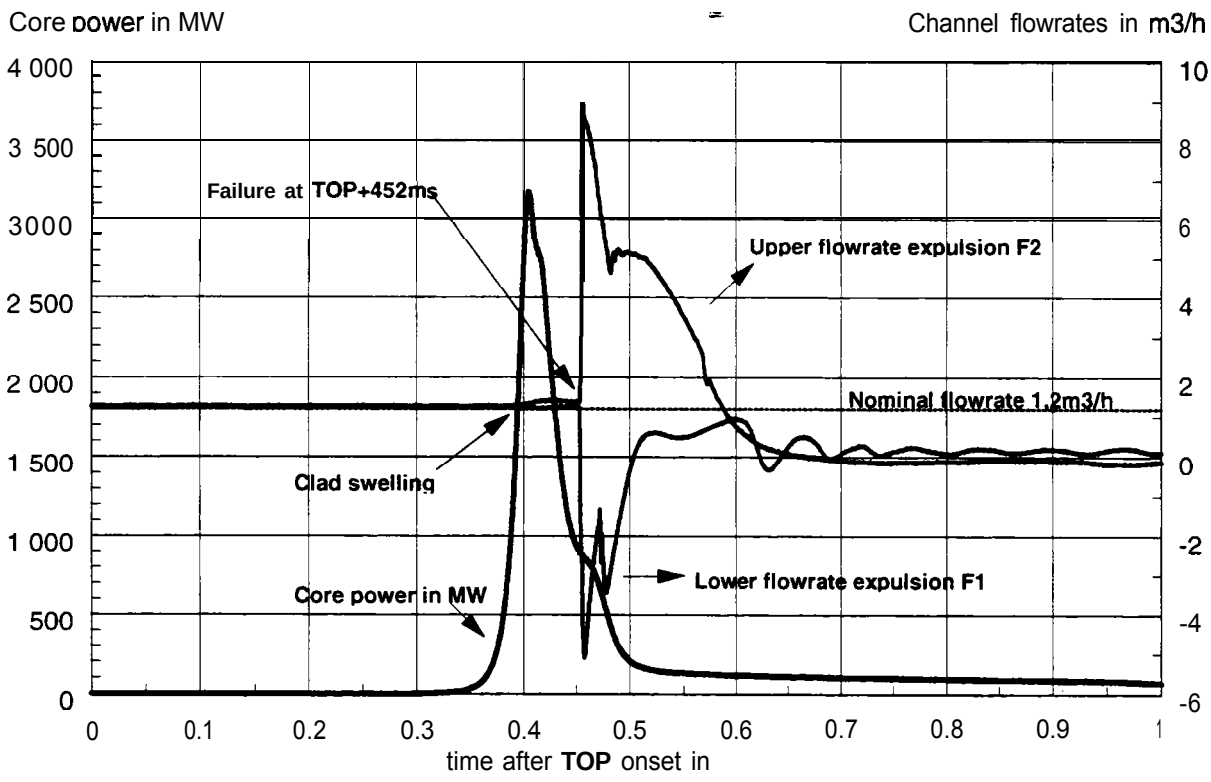


Fig 6 : Flow rate ejections in the test consecutive to rod failure in Rep-Na 7

At the present time no additional information from post-test examinations is available due to the delay of hot cell work.

The main striking point is the rod failure which occurred in spite of a limited corrosion level and absence of spalling of the cladding in opposition to the UO_2 fuel REP-Na failed rods characterized by a high burn-up level (60 GWd/t) and a thick corrosion layer (80 - 130 µm) with initial spalling leading to the presence of « blisters » (hydride accumulation).

First of all, in REP-Na 7, the axial location of the first failure at peak power level tends to eliminate any effect of fuel heterogeneity due to $UPuO_2$ agglomerates which is stochastic and could trigger the failure at any axial level. Moreover, the comparison of REP-Na 7 and REP-Na 6 with a similar power pulse leading to a maximum fuel enthalpy of 145 cat/g without failure, suggests an important influence of burn-up on the clad loading.

On the other hand, an evaluation with the SCANAIR code assuming an homogeneous behaviour of the MOX fuel leads to an equivalent plastic strain of 2 % at the time of failure.

Taking into account the results of the mechanical tensile tests realized in the PROMETRA program [5] on similar cladding as REP-Na 7 rod which show values of 1% and 30% for uniform and total elongations respectively, we can deduce that the clad failure in REP-Na 7 results from the contribution of fission gas pressure loading. Indeed, in the MOX fuel, the high concentration of fission gases in the UPu02 agglomerates over the whole section can be compared to the local rim zone of high burn-up UO_2 fuel and may be responsible for gas overpressure (the agglomerates represent roughly 20% of the fuel mass compared to 5% for the rim zone of a 5-cycles UO_2 rod).

The high level of confinement and concentration of gases could also explain the violent flow ejection at failure time.

However, the precise description of the MOX fuel behaviour by the SCANAIR code is not validated. Additional information from future post-test examinations is needed for better understanding.

3 - 4. MAIN OUTCOMES COMPARED TO UO_2 FUEL BERAVIOUR

From the preliminary analysis of these three CABRI REP Na tests with MOX fuel, the following points can be deduced.

- There is no evidence of a direct impact of the $UPuO_2$ agglomerates with regard to local thermal effect for rod failure in spite of the high energy deposit (REP Na 9) and as it could be expected from the fuel heterogeneity.

- The occurrence of the transient clad spalling due to clad straining is confirmed as it already occurred with UO_2 fuel rods in REP-Na 3 and REP-Na 4. Consequences on the clad-coolant heat transfer in reactor situation might be expected from this behaviour (enhancement of the risk of boiling crisis followed by clad heat-up)

- Similarly to what has been observed with UO_2 fuel CABRI tests, the contribution of intragranular gases to fission gas induced fuel swelling is confirmed at high temperature level (mainly in REP-Na 2 and REP-Na 9) leading to significant clad straining.

- A significant increase of fission gas release is found with the MOX fuel compared to UO_2 fuel at similar burn-up ; this effect is linked to the presence of a higher quantity of gases in intergranular and porosity bubbles associated to the $UPuO_2$ agglomerates behaviour under irradiation and is much increased at high burn-up. As a consequence of a rapid power transient with fuel heat-up and gas overpressure leading to grain boundary separation, a larger amount of gases can be released and be available for clad loading under gas pressure.

- The failure of the REP-Na 7 test rod with a sound cladding and a low corrosion level may be explained by the contribution of gas pressure on clad loading and suggests a high burn-up effect with MOX fuel.

5 - CONCLUSION AND FURTHER NEEDS

The three MOX-fuel tests of the CABRI REP-Na test program have allowed to investigate the consequences of the combined thermal and fission gas effects on a MOX fuel rod submitted to a RIA.

From the experimental findings which are presently available, no evidence for thermal effects resulting from the heterogeneous nature of the fuel can be given.

On the other hand, there are very clear hints that fission gas effects are enhanced with regard to the behaviour of UO_2.

These increased fission gas effects are to be explained by the significant difference of the gas retention in MOX fuel during nominal operation. The heterogeneous nature of the MOX and also the slightly higher fuel temperature during nominal operation produce, for a given gas release fraction, a higher gas retention fraction in intergranular bubbles and in porosities. These gas retention sites produce undelayed pressure effects during rapid transient heating which lead to fuel fragmention and associated clad loading.

The failure of REP-Na 7 is to be considered with particular attention because it is of a fundamentally different type than the failures of UO_2 fuel rods which only occurred when the clad mechanical properties were severely affected by the presence of hydride blisters. Such cladding behaviour has largeiy deviated the attention from the contribution of the high burnup pellets effect on the origin of the clad loading.

In REP-Na 7, the loading potential of the fuel pellets was high enough to break a sound cladding.

One of the frequently asked questions concerning the Cabri tests is related to the test conditions and the representativity of the test fuel [8]. It has been demonstrated earlier [e.g.9] that the loading conditions during the PCMI phase are representative and any failure occurring during this phase is directly transposable to the reactor case if the critical heat flux is not reached before the failure event. The rapid increase of the clad temperature after the boiling crisis might restaure the ductility of the corroded and hydrided brittle clad material. However the mechanical resistance is decreasing rapidly

and the internal rod pressure increases due to the high transient fission gas release. For this field of questions, no data base is presently available because no experimental facility exists in the world, allowing to expose irradiated fuel to reactor typical power excursions under the boundary conditions of the PWR (pressure, temperature and flow).

The IPSN project to implement into the Cabri reactor the pressurized water loop aims at providing this missing test facility for such a complete investigation.

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Last but not least we acknowledge the efficient and fruitful co-operation with our industrial partners EDF and FRAMATOME.

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HIGH BURNUP IRRADIATION PERFORMANCE OF ANNULAR FUEL PINS IRRADIATED IN FAST REACTOR PFR

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Abstract

The UK Prototype Fast Reactor (PFR) has irradiated MOX annular pelleted fuel pins clad with PE16 up to burn-up of over 20 % heavy atom (ha) without failure, these high burn-up fuel pins can provide the valuable data for the study of high burn-up capability. Thus, post irradiation examinations (PIE) have been performed on PFR high burn-up fuel pins, and the irradiation performance is evaluated focusing especially on the mechanical and thermal performance at high burn-up. The fuel pins from LVD and ANT assemblies were irradiated up to 23.2 and 18.9 %ha (at peak burn-up). The results of LVD test pins have been evaluated, which demonstrate that these fuel pins have excellent mechanical and thermal performances at high burn-up because of the high swelling resistance of PE16, the maintenance of initial annular geometry up to high burn-up and the behavior of Fuel to Clad Joint (JOG) formation. In this paper, the newly obtained results of ANT test pins with different O/M ratio (ANT: 1.985, LVD: 1.965) are added, and compared with the LVD pins. The ANT results indicate that FCCI becomes larger and the fuel swelling behavior is different at high burn-up. However, the effects are evaluated not to be severe for the capability of high burn-up (-20 %ha). Therefore, we conclude that MOX annular pelleted fuel pins clad with low swelling material have high burn-up capability in O/M ratios ranging from 1.965 to 1.985.

1. INTRODUCTION

In recent years, the economics of the fast reactor system as a whole has been given considerable attention and it has been shown that increasing fuel burn-up offers significant cost advantages. Annular fuel, which lowers the smear density to compensate for pellet swelling as burn-up increase and maximizes heat rating for a prescribed margin to fuel melting, is one of the promising design for fast reactor driver fuel.

UK Prototype Fast Reactor (PFR) has irradiated MOX annular pelleted fuel pins up to burn-up of over 20 % heavy atom (ha). The PFR high burn-up fuel pins can provide the valuable data for the study of high burn-up capability. Therefore, post irradiation examinations (PIE) have been conducted at CEA/Cadarache under the collaborative program between UKAEA, BNFL / CEA / JNC, and the irradiation performance of MOX annular pelleted fuel at high burn-up is evaluated.

The test fuel pins were selected from LVD and ANT assemblies designed for high burn-up experiments, which had achieved burn-up of 23.2 and 18.9 %ha (at peak), respectively without failure. The PIE of LVD test pins had been almost completed and the results were reported[1]. This paper adds the newly obtained results of ANT test pins with different O/M ratio (LVD : 1.965, ANT : 1.985), and discusses and compares the results.

After briefly outlining the design of these pins, we describe the main PIE results concerning high burn-up fuel pin behavior and the analytical evaluation results focusing on mechanical and thermal performance at high burn-up.

2. FABRICATION PARAMETER AND IRRADIATION

LVD was a cluster type assembly containing 12 fuel pins, and ANT was a fuel assembly containing 256 fuel pins. The pins of both assemblies were supported by honeycomb type grid spacers. These fuel pin designs employ 6.58 mm diameter fuel pin containing annular pellets clad with high nickel content steel alloy PE16[2], which is highly resistant to irradiation swelling. Figure 1 shows the schematic diagram of LVD and ANT test fuel pins. Both pins have a lower gas plenum, the lower axial blanket (LAB) and fissile column are supported by the platforms located by means of crimps within the cladding tube. Fabrication parameters of both test pins are summarized in table I, there is a difference between the O/M ratios of both pins. LVD and ANT assemblies were irradiated up to 23.2 and 18.9 %ha burn-up with peak cladding damage doses of 144 and 148 dpa-NRT(Fe), respectively. The peak linear heat ratings (LHR) of LVD and ANT pins are 580 and 480 W/cm. The irradiation parameters are summarized in table II.

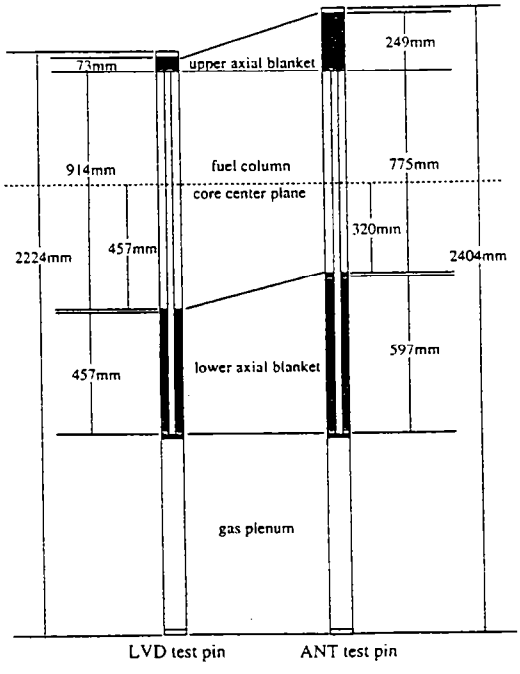


Fig.1 Schematic diagram of LVD and ANT test pins

TABLE I Fabrication parameters (LVD and ANT test pins)

	LVD test pins	ANT test pins
fuel pin total length (mm)	2224	2404
fuel column length (mm)	914	775
upper axial blanket length (mm)	73	248.5
lower axial blanket length (mm)	457	597
smear density (%TD)	86	88
gas plenum volume (cm^3)	19.7 (including central void)	21.2 (including central void)
pellet height (mm)	~7.5	~7.5
pellet outer diameter (mm)	5.56 (average)	5.56 (average)
pellet inner diameter (mm)	1.50 (minimum)	1.50 (minimum)
pellet density (g/cm^3)	10.6	10.9
Pu enrichment (wt.%)	32.3	25.2
O/M ratio (-)	1.965	1.985
cladding material	Nimonic PE16	Nimonic PE16
cladding outer diameter (mm)	6.58 (average)	6.58 (average)

TABLE II Irradiation conditions (LVD and ANT test pins)

	LVD test pins	ANT test pins
irradiation period (EFPD)	1095 (PFR Runs 11A to 29)	990 (PFR Runs 12 to 29)
peak burn-up (%ha)	23.2 (all pins)	18.9 (all pins)
peak dose (dpa-NRT)	144	148
max. linear heat rating (W/cm)	580 (at BOL) 360 (at EOL)	430 (at BOL) 480 (at peak LHR) 350 (at EOL)
max. cladding mid-wall temperature (°C)	670 (at BOL) 600 (at EOL)	580 (at BOL) 590 (at peak LHR) 570 (at EOL)

3. RESULTS OF POST IRRADIATION EXAMINATIONS

Non destructive examinations such as X-ray radiography, external visual examinations, gammascannings, profilometries and eddy currents were performed both on the LVD and on the ANT pins. Then destructive examinations starting with pin puncture followed by ceramography, fuel density, gas retention measurements and electron probe microanalyses (EPMA) were conducted on one pin of each type.

3.1. Non destructive examinations

The X-ray radiography examinations all along the pins showed the stability of the annular fuel and the blanket columns, the central void remaining large all over the columns. A small quantity of dense material was detected in the bottom end caps. This is not an unusual observation on PFR pins, that has no effect on the pin performances.

Large cesium displacements from the centre of the fissile columns towards its ends and the blankets were measured by gammascanning. It led to very high quantity of cesium between the lower axial blankets and the fissile columns. Moreover some interpellets locations exhibit peaks in cesium content, this is linked to the annular shape of the pellets. The estimated cesium concentrations of the two pins that underwent destructive examinations are presented in fig.2 and fig.3 together with the EPMA measurements in the oxide. A comparison with the cesium created allows an evaluation, for the different levels, of the amount of cesium displacement.

The diameter profilometries presented in fig.4 and fig.5 showing the low strains of these pins, the maximum value being less than 1%ΔD/D. LVD and ANT pins had similar cladding evolutions, the highest deformations occurring in the lower part of the fissile column and at the top of the

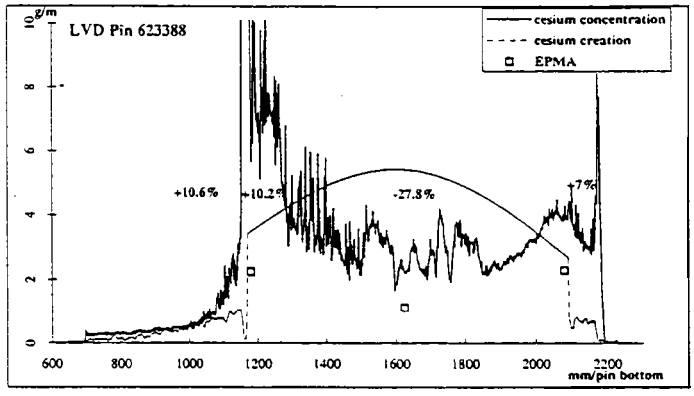


Fig.2 Cesium profile along LVD 623388pir

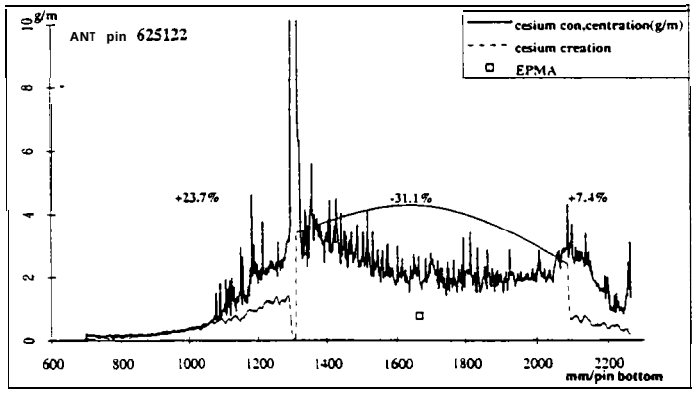


Fig.3 Cesium profile along ANT 625 122 pin

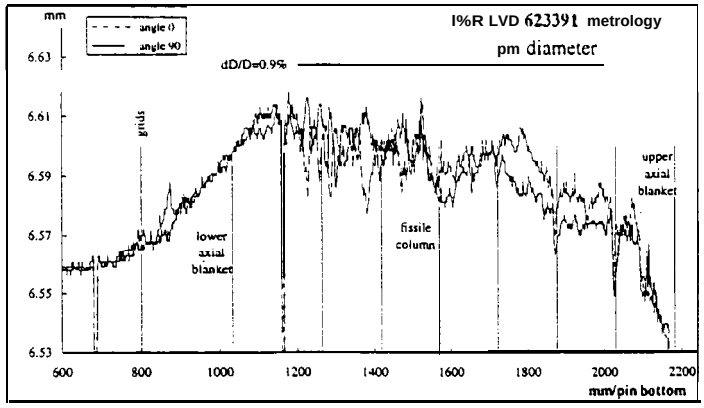


Fig.4 Profilometry of LVD 623391 pin

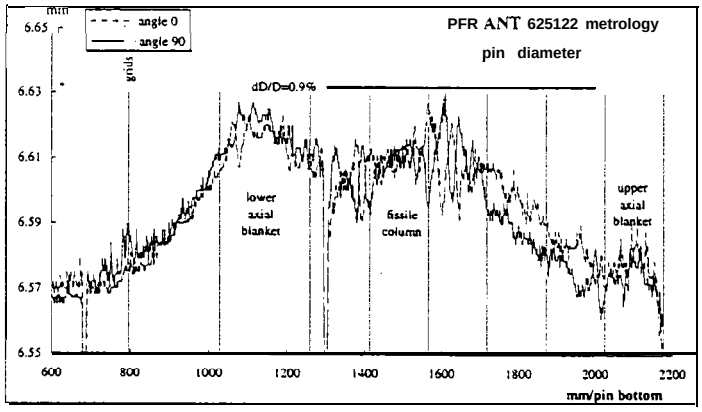


Fig.5 Profilometry of ANT 625122 pin

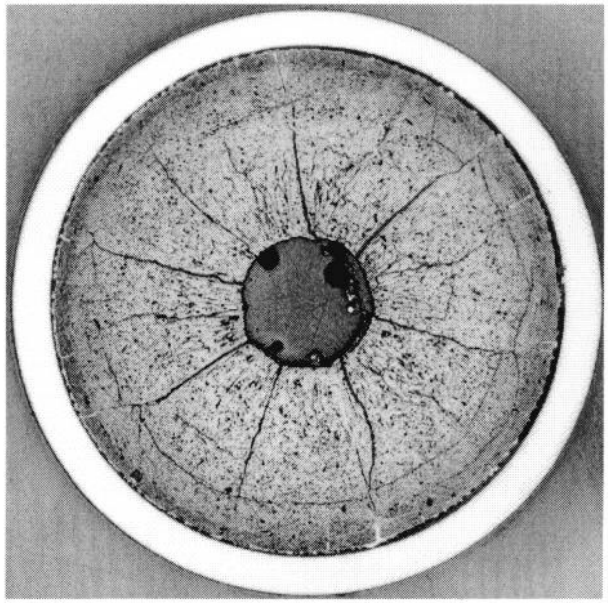
lower axial blanket. Some weak ovalization is observed along the fissile column but not along the blankets. Some external clad erosion was measured above the fuel. The eddy current measurements did not indicate any major local corrosion.

3.2. Destructive examinations

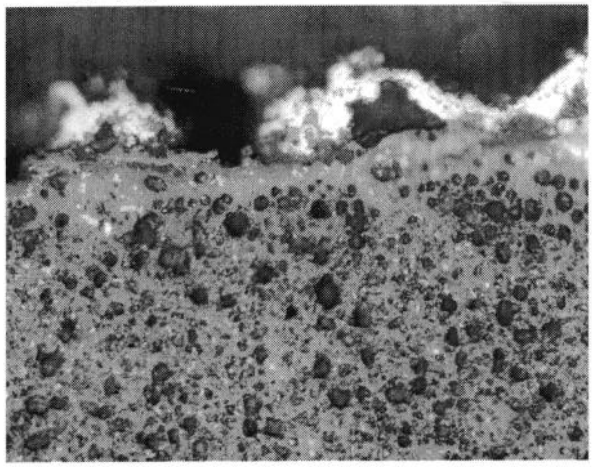
771 cc NTP of gas were collected by pin puncture from the 623388 LVD pin and 583 cc NTP from the 625122 ANT pin. The compositions of the gas have been determined by mass spectrometry. The released gas accounts for approximately 82 % of the gas created in the LVD fissile column and approximately 86 % of the gas created in the ANT fissile column. Code calculation on the pins showed that for ANT pin part of the released gas might come from the blankets. The actual release rate for the fissile column is then evaluated at 82 %. These release rates are consistent with JNC and CEA experiences at lower burn-ups.

Ceramographic examinations have been performed at five levels on each fissile column. Figures 6 and 7 allow an outline of the examinations at peak power node (PPN). At this level, in both cases, a high level of porosity is found at the pellet periphery, with larger pores in the ANT case. Around the central hole, though microprobe examinations show that plutonium redistribution greatly occurred (fig.13 and fig.14) the columnar grain structure has been modified by end of life (EOL) gas precipitation and grain fracturation at lower temperature.

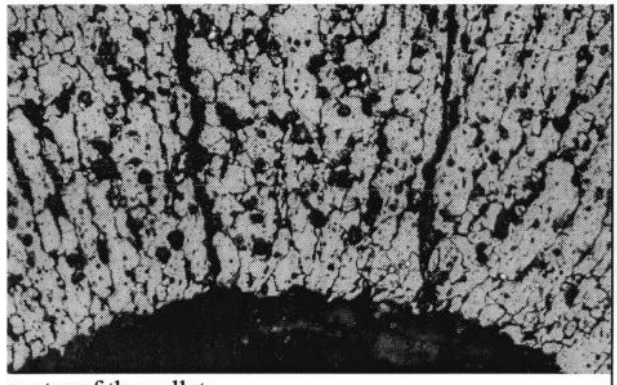
Figures 8 and 9 show the measurements of the central void diameter and pellet outer diameter at all the levels examined. The comparison with the fabricated nominal values shows that, at these points of irradiation, the pellet and clad dimensions remain very close to the fabrication values.



ceramography at PPN

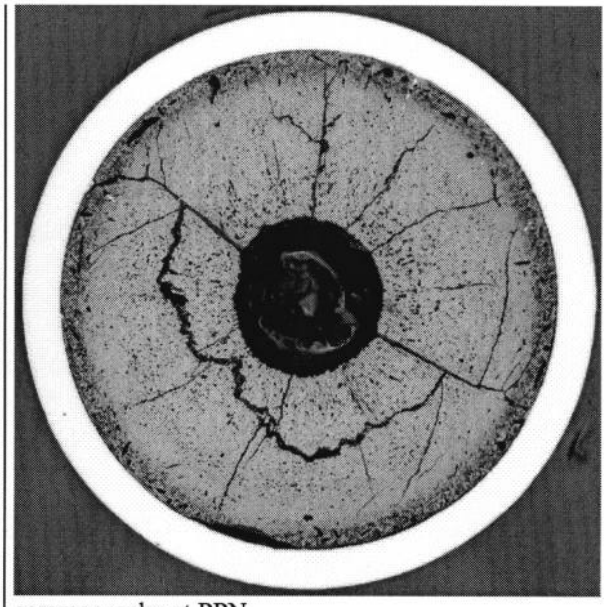


edge of the pellet

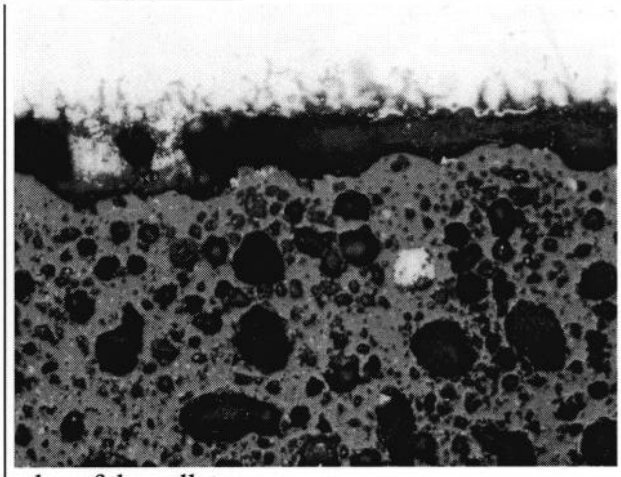


center of the pellet

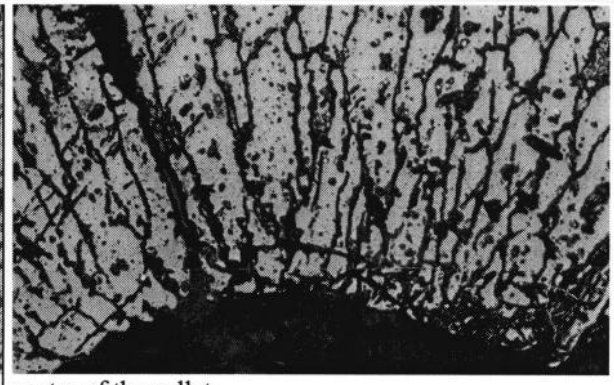
Fig.6 LVD PPN ceramography



ceramography at PPN



edge of the pellet



center of the pellet

Fig.7 ANT PPN ceramography

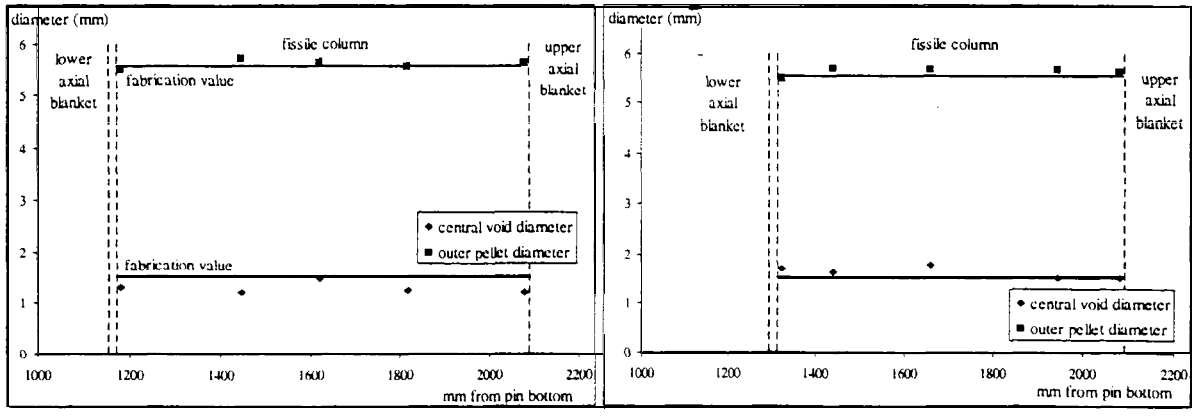


Fig.8 LVD 623388pin measurements

Fig.9 ANT 625122 pin measurements

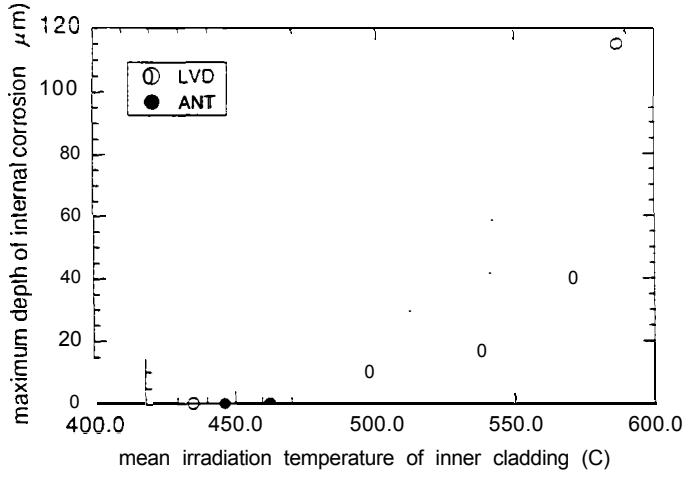


Fig. IO Clad internal corrosion for the **two** *pins*

Clad internal corrosion aue to fuel cladding chemical interaction (FCCI) was measured at five levels for the two pins. Figure 10 plots the relationship between maximum depth of internal corrosion and clad inner temperature in comparison of the two pins. It reveals that FCCI commence above the cladding inner temperature of -500°C and that the depth of ANT pin with higher O/M ratio is over twice larger than that of LVD pin at same temperature position. However, the amount is not considered to be serious at this LHR level.

The EPMA examinations confirm that the residual fuel to clad gap is filled with fission products compounds such as cesium, molybdenum, palladium, barium and tellurium associated with oxygen (Fuel to Clad Joint (JOG)) and cladding products like chromium (fig. 11). This JOG material is similar to that observed previously at lower burn-up . In spite of the high activity, the compounds found in the platform central void first detected by gammascanning were also examined. The X-ray mapping and quantitative analyses showed that in this area, large parts of the JOG material are probably cesium molybdate (fig.I2).

For the two examined pins, the cesium measurements show that at the PPN, about half of the cesium present at one level is still in the outer part of the pellet, the other half being in the JOG or in cracks (fig.2 and fig.3). The xenon profiles show that most of the remaining gas is in big pores. Only 20% of the retained gas is measured in the outer part of the pellets. The plutonium relocalisation presented fig.13 and fig. 14 highlights the columnar grain creation though the EOL low temperatures have concealed this structure. The fabricated densities were 10.6 g/cm' and 10.9 g/cm'. The measurements at three levels on the LVD pin and at PPN on the ANT pin extend from 9.68 to 9.85 g/cm' for local burn-ups from 12 to 23 %ha. This evolution is consistent with CEA and JNC experience.

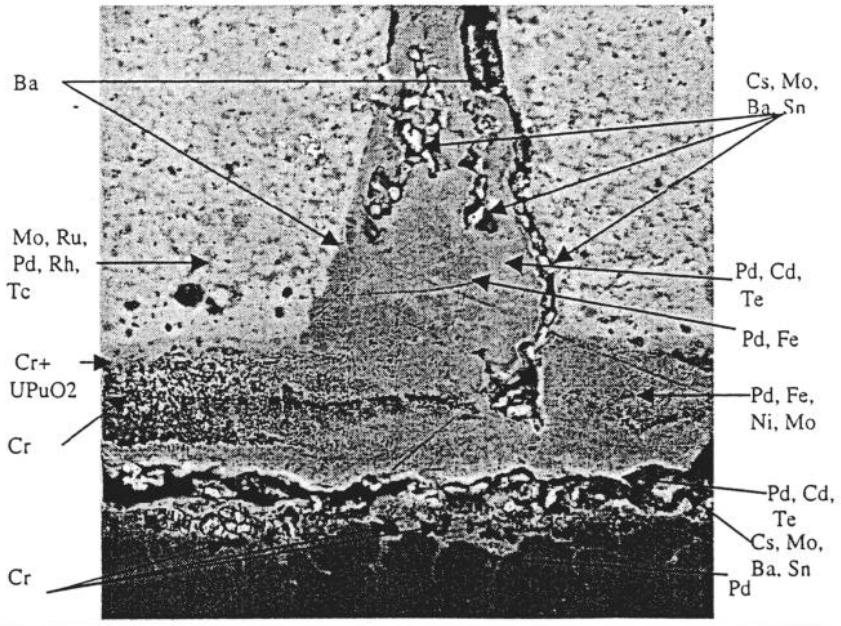
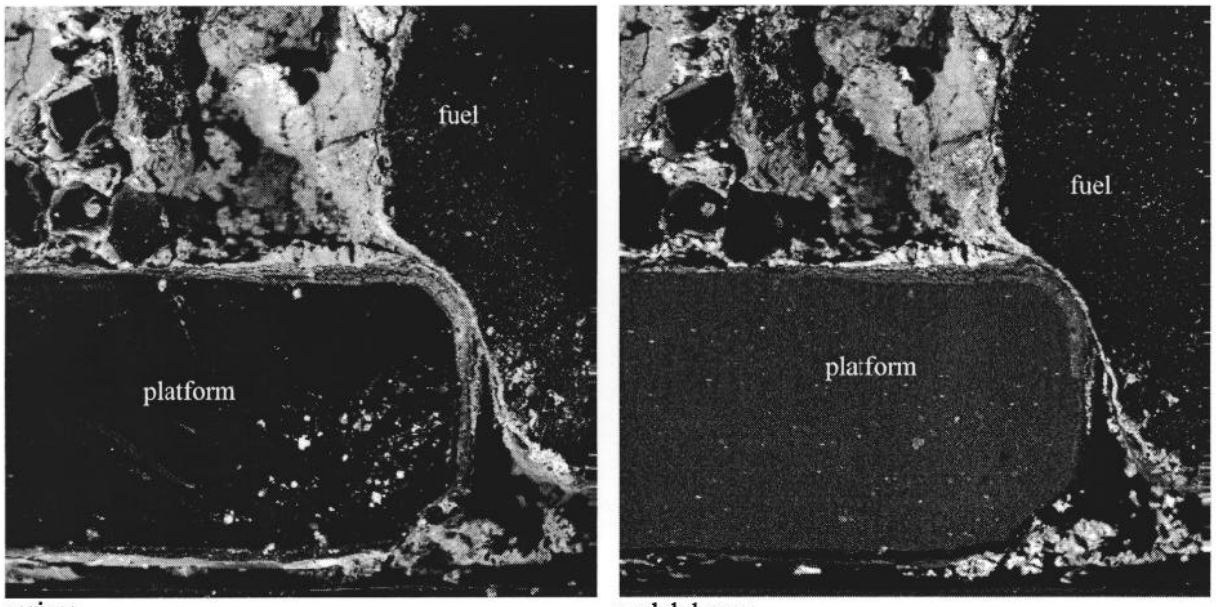


Fig.11 JOG material in LVD 623388 pins at PPN



cesium molybdenum

Fig.12 JOG material X-ray mapping at the platform level

4. EVALUATION OF IRRADIATION BEHAVIOR

It is generally a well known characteristic of fuel pins at high burn-up that fuel cladding mechanical interaction (FCMI) becomes significant in fuel pins with small cladding deformation and that accumulation of FP gas lowers gap conductance. PIE results described in Section 3 show that the fuel pin deformations are small and the fuel temperatures are low at high burn-up. Thus, the fuel irradiation behaviors are evaluated from the viewpoint of FCMI behavior and fuel temperature. At first, the annular pellet behaviors (pellet central void and outer diameter) are evaluated, then numerical analyses are performed on the cladding deformation and the fuel temperature in ANT and LVD test pins.

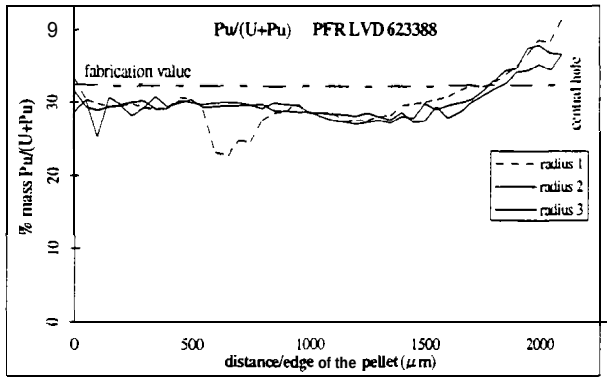


Fig. 13 Pu/(U+ Pu) along 3 L VD radii at PPN

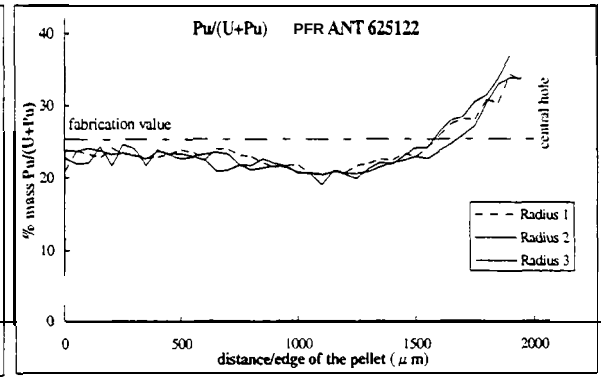


Fig. 14 Pu/(U+ Pu) along 3 ANT radii at PPN

4.1. Annular pellet behavior

X-ray radiography demonstrated that the condition of the central void within the fissile column was stable in the LVD and ANT pins. Ceramographic results (fig.6, fig.7) showed that central void geometry was kept without any significant reduction in diameter after the irradiation and that central void surface was smooth. Thus, it is confirmed that annular pellet retains the initial annular geometry up to high burn-up, which gives desirable heat transfer characteristic at high burn-up.

The increase of pellet outer diameter was small in LVD and ANT pins, as shown in fig.8 and fig.9, in spite of the irradiation up to high burn-up. In addition, EPMA measurements revealed that the residual fuel to cladding gap was filled with FP compounds (JOG). JOG is formed between 7 and 9 %ha burn-up[3], and it is known that fuel swelling is limited by the release of some FP from the fuel pellet to the residual gap with the increase of the oxygen potential of the fuel. Figure 15 plots the evolution with burn-up of pellet outer diameter based on the PIE data of ANT, LVD and JNC Monju type pins, where the data are separated into two O/M ratio groups in order to investigate the effect of O/M ratio. It reveals that a difference exists in the swelling behavior of LVD and ANT pins, and in the comparison of results from both pins the increase of pellet outer diameter in ANT pin with higher O/M ratio is relatively larger. This suggests the possibility that O/M ratio is reiated to sweiiing behavior at high burn-up.

4.2. Mechanical behavior

In order to evaluate the contribution of FCMI, fuel pin diameter changes due to cladding swelling and internal gas creep deformation were calculated. The correlations of PE16 (swelling, irradiation creep and thermal creep) and the irradiation conditions (dose, cladding temperature and internal gas pressure) were taken into account in this calculation. The analyses were performed in fissile columns of LVD 623387 pin and ANT 625198, 625122 pins, the contribution of FCMI induced diameter change was evaluated by comparing the above calculated value with the measured value. Figures 16, 17, 18 plot the results of analyses, respectively.

On fig.16, the calculated value agrees well with the measured value around the upper and central part of the fissile column, whereas the calculated values are lower than the measured value around the lower part. Therefore, it is considered that the contribution of FCMI has occurred around the lower part. However, the amount of deformation due to FCMI is small, FCMI seems to have been benign. On fig.17, the calculated value agrees well with measured value, and on fig.1 8, calculated value is a slight larger. These results indicate that the larger increase of pellet outer diameter in ANT pins has not affected the pin outer diameter change due to FCMI. In both measured results, significant decrease of outer diameter occurred in the upper part. This is considered to be related to clad erosion in flowing sodium coolant.

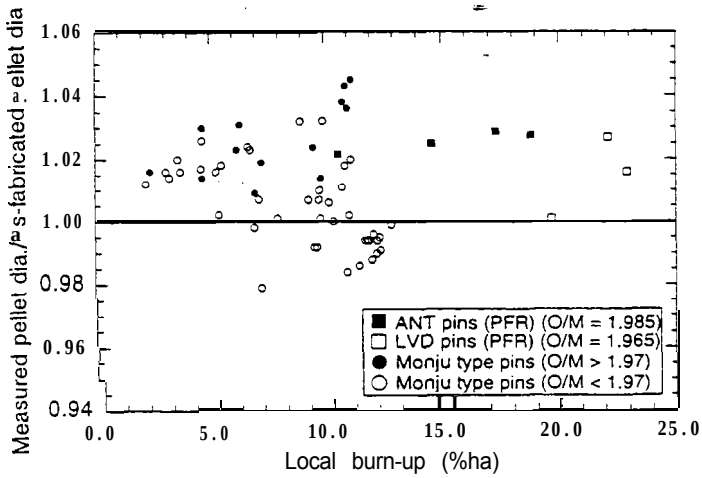


Fig.13 Evolution with burn-up of pellet outer diameter (ANT, L VD and Monju type pins in JNC)

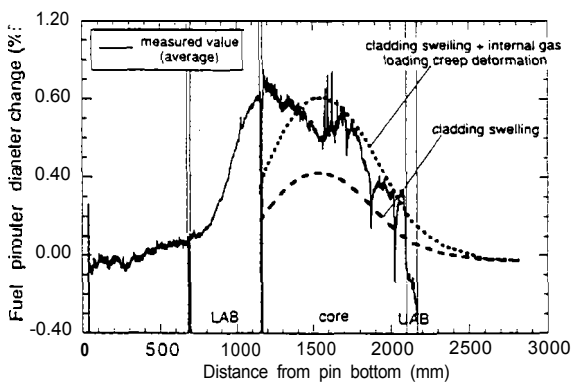


Fig. 16 Analyrical results of pin diameterchange (LVD 623387 pin)

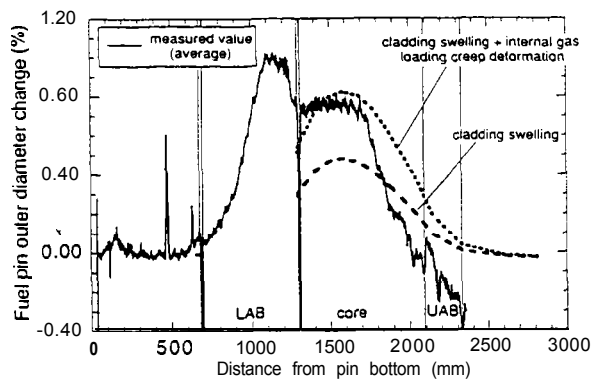


Fig. I 7 Analytical results of pin diameter change (ANT 625198 pin)

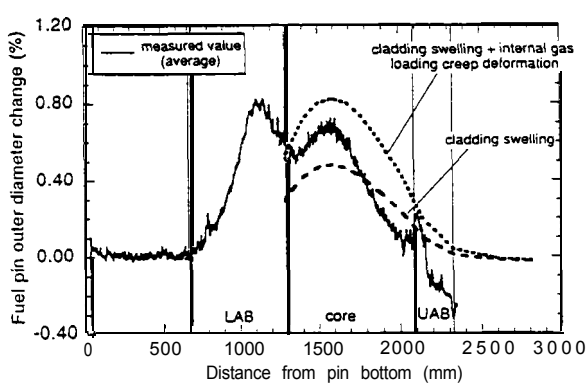


Fig. 18 Anabtical results of pin diameter change (ANT 625122 pin)

It is concluded that the outer diameter change in the LVD and ANT pins were induced by mainly cladding swelling and internal gas loading creep, and the contribution of FCMI to pin deformation appeared to be small. The benign FCMI behavior is related to the limitation of fuel pellet swelling due to JOG formation.

4.3. Thermal performance

The fuel radial temperature profiles were analyzed- in order to evaluate the thermal performance in high burn-up fuel pins. The calculations were performed on PPN ceramographic specimens of LVD and ANT pins by a one-dimensional heat transfer calculation code. The geometric conditions (fabrication parameters at BOL, ceramographic results at EOL) and local irradiation conditions (LHR and cladding mid-wall temperature) were taken into account in this calculation.

The calculations on LVD pin were conducted at BOL and EOL, and those on ANT pin were conducted at BOL, EOL and peak LHR cycle. In BOL cases, it was assumed that residual fuel to cladding gap was filled with He gas, where the gap conductance from the Ross & Stoute model[3] was used. In EOL cases, it was assumed that the residual gap was filled with the dense FP compounds (JOG) as shown in EPMA measurements, where Cs-MO-O compounds were considered to be thermodynamically stable . Thus the gap conductance was evaluated from the conductivity of Cesium molybdate (Cs_2MoO_4) At peak LHR cycle of ANT pin, it was assumed that the residuai gap was filled with mixed FP gas, whose composition was the same as other pin irradiated to an equivalent burn-up in PFR. Figures 19, 20 plot the results of analyses for PPN ceramographies of LVD and ANT pins, respectively.

On fig.19 (LVD), the centerline temperature at BOL is about 2400°C, which obviously exceeds the threshold temperature of columnar grain formation (about 1 800°C[4]). Whereas, the centerline temperature at EOL is as low as about 1800°C, because of the decrease in LHR with burn-up and FP compounds (JOG) in the residual gap compensate the decrease of gap conductance due to accumulation of FP gas. The PPN ceramograph of LVD pin showed that the fuel pellet central temperature has not entirely reached the threshold temperature'around EOL. Thus, the results of analysis are consistent well with the ceramographic evidence. On fig.20 (ANT), the

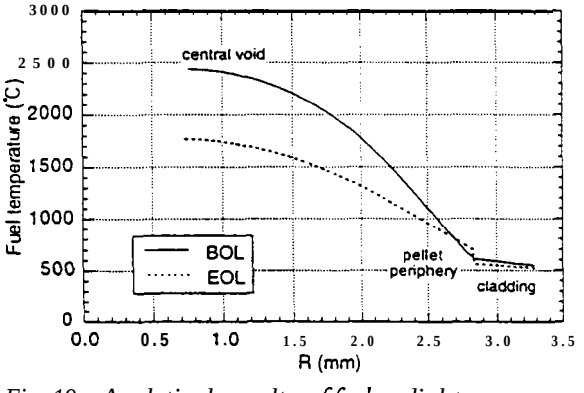


Fig. 19 Analytical results of fuel radial temperature profiles (LVD PPN ceramography)

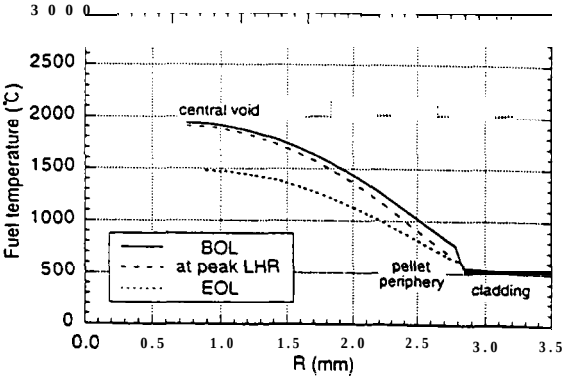


Fig.20 Analytical results of fuel radial temperature profiles (ANT PPN ceramography)

temperature at BOL and peak LHR cycle are slightly higher than the threshold temperature. The centerline temperature at EOL is reduced to about 1500°C partly due to the effect of JOG as in the LVD pin.

Therefore, the existence of JOG has a favorable influence in thermal performance, and it is considered that the fuel temperatures in LVD and ANT test pins have been low enough to give a large margin to fuel melting at high burn-up.

5. CONCLUSION

The newly obtained results of ANT test pins with higher O/M ratio indicate that FCC1 becomes larger and the fuel swelling behavior is different around high burn-up. However, the effects are evaluated not to be severe for the capability of high burn-up (-20 %ha). Therefore, we conclude the followings in O/M ratio ranging from 1.965 to 1.985,

(1) Fuel pin outer diameter changes was smaller than l%AD/D because of the high swelling resistance of PE16, and it was evaluated to be mainly due to cladding swelling and internal gas loading creep deformation, suggesting benign FCMI loading. The benign FCMI behavior is attributed to fuel pellet swelling being limited due to JOG formation and displacement toward the fissile column extremities at high burn-up.

(2) The fuel central void maintained the initial annular geometry without significant reduction in diameter up to high burn-up. The FP compounds (JOG) in the residual fuel to cladding gap was demonstrated to produce good heat conduction characteristic. These factors give a desirable heat transfer characteristic to the annular fuel, and the fuel temperature is considered to be low enough to give a large margin to fuel melting at high burn-up.

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MODELLING OF THE THERMOMECHANICAL AND PHYSICAL PROCESSES IN FR FUEL PINS USING THE GERMINAL CODE

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Abstract

In the frame of the R&D on Fast Reactor mixed oxide fuels, CEA/DEC has developed the computer code GERMINAL for studying fuel pin thermal and mechanical behaviour, both during steady-state and incidental conditions, up to high burn-up (25 at%). The first part of this paper is devoted to the description of the main models : fuel evolution (central hole and porosity evolution, Plutonium redistribution, O/M radial profile, transient gas swelling, melting fuel behaviour, minor actinides production), high burn-up models (fission gas, volatile fission products and JOG formation), fuel-cladding heat transfer, fuel-cladding mechanical interaction. The second part gives some examples of calculation results taken from the GERMINAL validation data base (more than 40 experiments from PHENIX, PFR, CABRI reactors) , with special emphasis on : local fission gas retention and global release, fuel geometry evolution, radial redistribution of plutonium for high burn-up fuels, solid and annular fuel behaviour during power ramps including fuel melting, helium formation from MA (Am and Np) doped homogeneous fuels.

1. INTRODUCTION

GERMINAL code is able to calculate mixed oxide fuel with the main following characteristics :

- As fabricated O/M : [1.90 to 1.999]
- Pu content (Pu/M) : [0 to 45%]
- Fuel relative density : [85 to 99]
- Solid or hollow pellets.
- peak linear rating : [steady state : up to 550 W/cm, slow transient : up to 1300 W/cm]
- Peak burn-up : [0 to 25 at%]
- Max. Cladding temp. [steady state : up to 650 °C, slow transient : up to 920 °C]

The main originality of the GERMINAL code is the modelling of high burn-up volatile fission products behaviour in accordance with the experimental observation of an increase of fission gas release rate and migration of volatile fission products out of the fuel (JOG formation) [1].

2. MODELS NOT SPECIFIC TO HIGH BURNUP FUEL

2.1. Central hole evolution

Central hole formation in solid pellets or central hole evolution in hollow pellets is due to both radial porosity migration and cracks healing under thermal gradient. Porosity migration is described by vapour transport of the species UO, UO_2, UO_3, PuO and PuO_2.The partial pressures in equilibrium are determined as fonction of the local temperature, uranium and plutonium content and oxygen pressure.

The oxide molar flux due to vapour transport is given by :

$$FLUX = - \frac{D}{RT} \frac{\delta P}{\delta r}$$

where **P** is the total pressure of gaseous species, **T** is the fuel temperature, **R** the gas constant and **D** the diffusion coefficient of the oxide molecules through the inert gas in porosity and cracks.

$$D = \frac{\sqrt{\frac{8RT}{\pi M}}\sqrt{\frac{M+Mg}{Mg}}}{3\pi\sigma^2 ng}$$

with **M** being the molecular weight of oxides, **Mg** the molecular weight of inert gas, **ng** the atomic density of the inert gas and σ the collision diameter.

2.2 **Plutonium radial redistribution**

The oxide molar flux due to vapour transport and the solid state diffusion of U and Pu atoms produce radial plutonium redistribution according to the following equations :

Pu migration by vapour transport :

$$\text{FLUX Pu} = \text{FLUX}\frac{\text{grad } P_{Pu}}{\text{grad P}}$$

with :

$$\text{grad } P_{Pu} = \frac{\delta P_{Pu}}{\delta r} \text{ and } \text{grad P} = \frac{\delta P}{\delta r}$$

where :

$\mathbf{P_{Pu}}$ = Partial pressure of Pu species (PuO and PuO_2), **P** = Total pressure

Pu migration in solid phase :

$$\text{FLUX Pu} = \frac{D_{Pu}}{V_{molar}}\left[\frac{\delta\left(\frac{Pu}{M}\right)}{\delta r} + \left(\frac{Pu}{M}\right)\left(1-\frac{Pu}{M}\right)\frac{Q_1}{RT^2}\frac{\delta T}{\delta r}\right] \quad \text{(moles/(cm}^2\text{.s)}$$

with :

$$D_{Pu} = DI + A\exp\left(-\frac{Q}{RT}\right) \quad \text{(cm}^2\text{/s)}$$

A = 0.34	(cm²/s)
Q = 110	(kcal/mole)
DI = 1. 10^{-16}	(cm²/s)
Q_1 = 35	(kcal/mole)

2.3. Oxygen radial profile

The oxygen radial redistribution model used in GERMINAL is issued from the RAND-MARKIN and ROBERTS model [2] based on the CO/CO_2 equilibrium.

This model takes into account the following hypothesis :

(1) Equilibrium between the oxide and the oxygen partial pressure

$$\Delta GO_2 = RTLnP(O_2) \qquad (1)$$

(2) Equilibrium between O_2 and CO/CO_2

$$CO + \frac{1}{2}O_2 \Leftrightarrow CO_2 \qquad \Delta G_R^0$$

$$\frac{P(CO_2)}{P(CO)P(O_2)^{\frac{1}{2}}} = \exp\left[-\frac{\Delta G_R^0}{RT}\right] \qquad (2)$$

(3) The ratio $\frac{P(CO_2)}{P(CO)}$ is radialy constant

By elimination of $P(O_2)$ in equations (1) and (2) we obtain :

$$AGO_2 = 2\Delta G_R^0 + 2RTLn\frac{P(CO_2)}{P(CO)}$$

Using the reiation between AGO, and the deviation from stoechiometry for a mixed-oxide $(U_{(1-y)}, Pu_y)O_{(2-x)}$ [3] :

$$\log_{10}\left(\frac{x}{y-2x}\right) = 1.796 - \frac{11315}{RT} - \frac{0.43\alpha}{RT}\Delta GO_2$$

with :

$$\frac{1}{\alpha} = \frac{A}{T^2} + \frac{B}{T}$$

A = - 482 10^4 and **B** = 8459

We determine by iterative procedure the value of the ratio $\frac{P(CO_2)}{P(CO)}$ which allows conservation of the O/M mean value in the slice.

2.4. **Transient gas swelling**

Experimental observations of transient gas swelling (International CABRI program) have been used to establish the GERMINAL transient fission gas swelling model. This gas swelling occurs during power ramps leading to nonequilibrium between fission gas retention and the thermal level reached by the fuel during power increase.

The basic equation is :

$$\frac{dg}{dt} = \frac{g\,max - g}{tog}$$

where : g is the fractional fuel swelling, **gmax** is the maximum fractional swelling, tog (s) is the time constant and **t** (s) is the time.

The time constant tog has been determined using several power ramping tests of the CABRI program :

$$tog = \frac{\frac{dP}{dt} + b}{c\frac{dP}{dt} + d} \times \frac{k}{D}$$

where : **P** (W/cm) is the linear power, **D** (cm^2/s) is the diffusion coefficient of the vacancies at grain boundary, k, **b, c** and **d** are constants depending of the range of the power transient.

$$D = 1321 \times 10^{-2} \exp(-\frac{Q}{RT})$$

where : Q is the activation energy for the vacancies migration toward grain boundary, R is the gas constant and **T (K)** is the local temperature.

$$\frac{Q}{R} = 28760$$

The maximum fractional swelling, gmax, is determined by equilibrium between intergranular gas pressure and inner pin pressure or fuel to cladding contact pressure in case of fuel-cladding mechanical interaction.

2.5. **Melting fuel behaviour**

In case of fuel melting, an optional axial molten fuel transfert model can be used. This model is based on the following assumptions :

(1) Beyond a given melt fraction (mobility threshold) the melting fuel is able to move axially in all the free volumes of the fissile column.

(2) Axial motion is governed by pressure equilibrium between the gas pressure inside the molten cavity (gas in porosity and cracks, retained fission gas) and the plenum pressure.

(3) Molten fuel moves upwards and downwards. A coefficient allows to promote the downwards direction in order to simulate the gravity contribution.

(4) Displacements are assumed instantaneous (pressure equilibrium faster than thermal transient). Therefore this model is only relevant for slow power transients such as control rod withdrawal.

2.6. **Minor actinides**

Usually, for classical MOX fuel, only three isotopes are considered in fuel calculation (^{235}U, ^{238}U and $^{239eq}Pu$).

In the frame of actinides burning studies, an optional model taking into account all Uranium and Plutonium isotopes as well as the main Americium, Neptunium and Curium isotopes has been introduced in GERMINAL code.

For thermochemical and thermomechanical modelling, it is assumed that the presence of minor actinides does not influence the fuel behaviour. As a consequence the use of that option is restricted to low minor actinides content (less than 5%).

With that restriction the code is able to correctly describe the evolution of the isotopic composition and the helium production versus time.

3. **MODELS SPECIFIC TO HIGH BURNUP FUEL**

The following models are able to calculate both low and high burn-up fuel

3.1. Fission gas model

Fission gas production is calculated using the following fission gas production rate :

- Fast neutron flux : 0.25 (Xe+Kr) atom per fission
  0.01 He atom per fission
- Thermal neutron flux : 0.3 1 (Xe+Kr) atom per fission

Fission gas release model takes into account the experimental observations showing at high burnup an increase of the fission gas release rate associated with a decrease of the grain size and the appearance of a large gap filled with fission products.

The increase in the fission release rate occurs when the local fission gas retention N exceeded a saturation threshold **GASMAX**.

GASMAX is determined by the equality of the fission gas atom flux leaving the fuel matrix and the resolution flux.

$$GASMAX = \frac{2.2414 \times 10^{4} \times 5.\times 10^{-24} \times DFISS \times QNSAT}{2(DTH + DATH)} \qquad (cm^3\ STP/g)$$

where : **QNSAT** is the cover ratio of the matrix by the gas :

$$QNSAT = \frac{2.224; 10^{18}}{} \quad (at/cm^2)$$

DTH and **DATH** are respectively the thermal and athermal diffusion coefficients :

$$DTH = 7.6 \times 10^{-6} \exp(\frac{-35225}{T}) \quad (cm^2/s) \qquad DATH = 1.2 \times 10^{-29} DFISS \quad (cm^2/s)$$

in which **DFISS** is the fission rate (fission/(cm^3.s))

The fission gas release equation is as follow :

- Before reaching the saturation threshold ($N < GASMAX$)

$$\frac{dN}{dt} = \beta - hN$$

where N (cm^3/g) is the gas retention, **t** (s) the time and β the fission gas creation rate.

- After the saturation threshold ($N > GASMAX$)

$$\frac{dN}{dt} = \beta - hN - k(N - N\,lim)$$

where **k** (s^{-1}) is a time constant and **Nlim** (cm^3/g) the limit of retention.

3.2.Volatile fission products and JOG formation

Among the volatile fission products, Caesium is the more interesting as it is the major constituent of the JOG (Joint Oxyde Gaine, as fuel to clad joint).

The JOG model is based on the following hypothesis [4] :

- Volatile fission products (Cs) behave as the fission gas (FG).

$$Cs(released) = FG(released) \times \frac{Cs(created)}{FG(\quad created)} \qquad z$$

9 Radial migration of Cs towards the periphery of the fuel pellet before reaching the saturation threshold.

9 After reaching the saturation threshold, Cs moves outwards the fuel to create the JOG

9 The radial migration of Cs induces a radial fuel swelling gradient :

$$\frac{dSW}{dt} = A + B\frac{Cs(\ retained)}{Cs(\ created)}$$

where : **SW** = fuel swelling, **A** = solid fission products swelling rate (0.244/at% burn-up)

and **B** = volatile fission products swelling rate (0.864/at% burn-up)

9 JOG formation occurs after reaching saturation threshold and the quantity of Cs transferred from a mesh of the fuel to the JOG (QCsJOG) is given by :

QCsJOG = FGRF x QCs(retained)

where : **FGRF** = fission gas release fraction in the mesh and **QCs(retained)** = quantity of Cs retained in the mesh

9 The JOG width is calculated assuming that the specific volume of volatile fission products in the JOG is the same as in the fuel (106.3 cm3/mol).

Notice that this model is a threshold one. When the condition GASRET > GASMAX is reached once a time in a fuel mesh, the high burn-up modelling is irreversibly apply to that mesh. This is not very easy to manage this in term of code results stability, particularly when the same calculation case is performed using different radial meshing.

3.3. **Fuel-cladding heat transfer coefficient**

Before JOG formation, fuel-clad heat transfer results from both conduction in a gas gap (H,) and radiative transfer (H,). The calculation of H_g takes into account the gas composition as well as its temperature and pressure, the gap size, the jump distance and the surface roughness of the fuel pellet and of cladding. Calculation of H_r uses the STEPHAN law.

After JOG formation two periods are considered according to the quantity of created JOG :

9 When JOG width becomes higher than 50 micrometers, the heat transfer calculation only results from conduction in the JOG material taken as Cs_2MoO_4 compound :

The thermal conductivity of Cs_2MoO_4 with a relative density of 0.943 is given by [5]:

$$\lambda(Cs_2MoO_4) = \frac{78.242}{T} + 0.15 + 1.7 \times 10^{-10} T^3 \qquad \text{if } T < 845 \text{ K}$$

$$\lambda(Cs_2MoO_4) = \frac{132.56}{T} + 0.03 + 3.2 \times 10^{-10} T^3 \qquad \text{if } T > 845 \text{ K}$$

with $\lambda(Cs_2MoO_4)$ in W/(m.K)

$$\lambda(\text{jog}) = \lambda(Cs_2MoO_4)\frac{(1-2\times \text{PORJOG})}{(1\text{- }2\text{ x }0.057)}$$

where : **PORJOG** is the JOG porosity.

- Between 0 and 50 micrometers of JOG thickness the thermal heat transfer calculation results both from the combination in parallel of transfer in a gas gap and transfer in the JOG.

3.4. **Fuel cladding mechanical interaction**

When the fuel-clad gap is closed, mechanical interaction between fuel and cladding is calculated by iterations between the contact pressure and the resulting plasticity temperature which determines the fuel zone where fuel cracks closure occurs.

The plasticity temperature is determined by solving the equation :

$$\dot{\varepsilon} = 1.5\times 10^{-13}\times\sigma\times \mathbf{Q} + \exp[18.3(1-D)]\text{ x }920(1+0.01Q)\exp[\frac{-46600}{Tpl}]\times\sigma$$

$$+\exp[24.1(1-D)]\text{ x }410\exp[\frac{-68900}{Tpl}]\times\sigma^{4.4}$$

where : $\dot{\varepsilon}\,(s^{-1})$ is the creep rate, $\sigma\,(MPa)$ is the stress induce by the contact pressure, Q (W/cm^3) is the volumique power, **D** is the relative fuel density and **Tpl** (K) is the plasticity temperature.

In case of central hole closure, the model calculates equilibrium between the pressure inside the porosity of the fuel plastic zone and the fuel-clad contact pressure.

After JOG formation, if the distance between fuel and clad becomes lower than the JOG width created, the JOG is axially moved as long as there is free volume inside the fuel to clad gap, after what, the JOG stays in place and participates to clad straining.

Normally mechanical calculation of the clad is done with a fine cylinder hypothesis but GERMINAL includes an option allowing thick tube mechanical calculation which is associated with fuel pin design criteria.

4. **COMPARISON CALCULATION/MEASUREMENT**

Data base of GERMINAL code contains a large number of fuel pins irradiated at various burn-up levels up to 25 at% in steady state and off-normal conditions. Many of them come from PHENIX standard subassemblies (6.5 mm OD cladding and solid pellets) but also from experimental ones (annular pellets, SPXl design, minor actinides doped fuels...). Elsewhere, experiments coming from other reactors (PFR, CABRI, HFR etc,) have been included these last years in the data base.

Except for the fission gas release comparison which takes into account all the data base, we give, for other parameters, comparisons between measurements and calculations chosen among the more representative and significant experiments beside the code modelling.

Table 1 summarises the main fabrication and irradiation characteristics of selected irradiations of which the results are subsequently commented.

TABLE I. FABRICATION AND IRRADIATION CHARACTERISTICS OF FUEL PINS USED IN THE COMPARISON CALCULATION/MEASUREMENTS

Pin reference	Main objectives	Fabrication characteristics	EFPD	Burn-up at PPN	Maximum cladding deformation
RIG 2 (PFR exp.) AGO test in CABRI	low burn-up at low linear rating gas swelling during transient	solid pellets 16% Pu content clad : CW 316	180	2.9 at%	< 0.5%
CPd 6004 (Phenix stand.) JOG 1 test in CABRI	intermediate burn-up without cladding deformation - behaviour of JOG layer	solid pellets 28% Pu content clad : CW 15-15 Ti	750	11.5 at%	< 0.5%
Boitix 1 (Phenix exp.)	intermediate burn-up with low cladding deformation	solid pellets 20% Pu content clad : CW 316 Ti	400	7.0 at%	< 1%
Boitix 3 (Phenix exp.)	high burn-up with strong cladding deformation	solid pellets 20% Pu content clad : CW 316 Ti	820	13.5 at%	< 7%
Viggen 6 (Phenix exp.)	high burn-up with medium cladding deformation	solid pellets 28% Pu content clad : CW 15-15 Ti	980	15.5 at%	< 3.5 %
Papyrus (Phenix exp.)	high burn-up without cladding deformation	solid pellets 20% Pu content clad : INC 706	850	14.5 at%	< 0.25 %
ANT (PFR exp.)	high burn-up with low cladding deformation	annular pellets 25% Pu content clad : STA PE16	990	18.9 at%	< 0.6 %
LVD (PFR exp.)	high burn-up with low cladding deformation	annular pellets 33% Pu content clad : STA PE 16	1095	23.1 at%	< 0.6 %
Superfact 1 (Phenix exp.)	burning of minor actinides (Np and Am) according to homogeneous way	solid pellets 24% Pu, 2% AM clad : CW 15-15 Ti	380	6.6 at%	< 0.1 %

4.1. Fission gas behaviour

Figure 1 shows comparison between calculated fission gas release at the end of irradiation and experimental pin puncture analysis for four ranges of burn-up. Results from GERMINAL code are generally in fair agreement with measured values whatever reached burn-up.

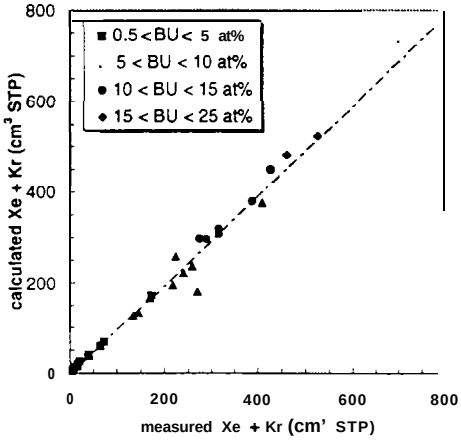

Fig. 1 Comparison between calculted an measuredfission gas release

This fair global agreement is confirmed by comparison on local retained fission gas. Figure 2 shows prediction and measurements for axial distribution-of retained fission gas for the so called Boitix 1 and Boitix 3 PHENIX fuel pins.

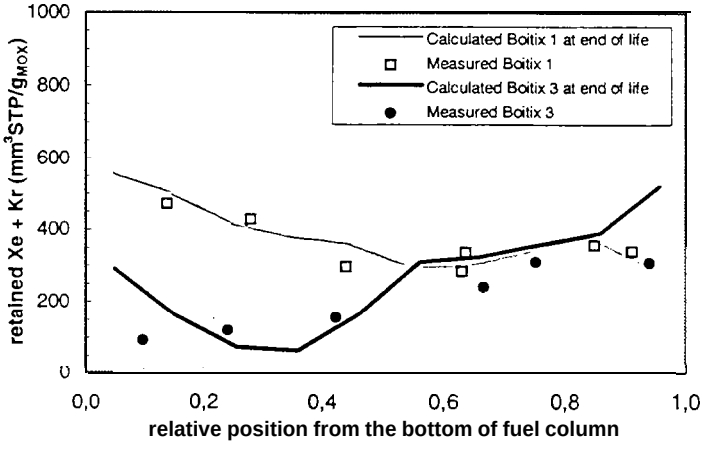

Fig 2 : comparison calculation/measurements for axial evolution of retained fission gas for two PHENIX fuel elements

Boitix 1 represents a fuel element at medium bum-up level with low cladding deformation. In such case, the axial profile of retained FG is classical with a minimum around the peak power node. Boitix 3 is characteristic of the behaviour of fuel elements irradiated at high bum-up with a strong cladding swelling located in the lower half of the fissile column. The overheating of the fuel in this area, leads to a local significative decrease of retained FG to very low values (< 150 mm^3/g_{MOX}) in spite of the high local production (2800 mm^3/g_{MOX}). The answer of GERMINAL fission gas release model is in rather agreement with measurements.

4.2. Pellet geometry evolution

Figure 3 shows the comparison of the central void diameter measured at different levels of the fuel column for two irradiations so called Viggen 6 and Papyrus with respective GERMINAL calculations.

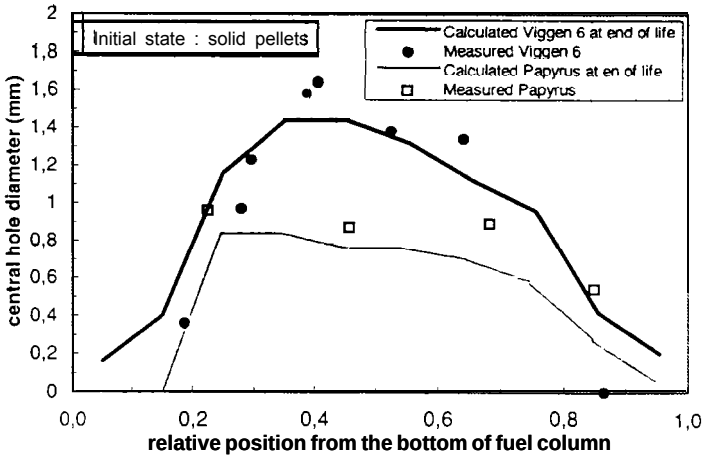

Fig 3 comparison calculation/measurement for axial evolution of central hole diameter for two PHENIX high B U fuel elements

Both Viggen 6 and Papyrus are typical of fuel elements at high bum-up level. Their difference lies in the range of clad swelling which remains low for Papyrus pin (cladding in Inconel 706) whereas for Viggen 6 one (cladding in CW 15-15 Ti) it locally reaches a relative deformation of 3.5%. Consequently, thermal fuel behaviour is greatly influenced by cladding straining or not, Fuel restructuring may restart for Viggen 6 pin when cladding swells, leading to an increase of the central void size and this phenomenon is well described by GERMINAL. Nevertheless, some discrepancies may be noted at the end regions in which the linear rating is generally low (< 200 W/cm). A possible cause of disagreement is an extrapolation error of restructuring model from short time and high temperature to long time and low temperature parameters.

Figure 4 shows for these two same fuel pins, the comparison between the fuel to cladding apparent gap width, measured from optical ceramographies and the calculated one.

From medium burn-up level (- 6 at%), a lot of SEM examinations has shown a progressive evolution of the fuel cladding gap with the opening of so-called JOG (Joint Oxyde Gaine) filled with fission product compounds such the cesium molybdate which is majority represented [l]. Properties and behaviour of JOG were studied for unstrained and strained pins : The JOG thickness depends on bum-up level but particularly to clad straining.

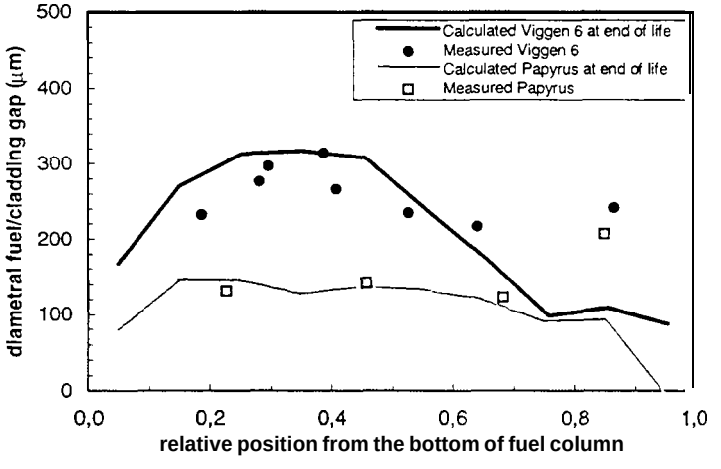

Fig 4 : comparison calculationfmeasurementfor axial evolution of fuel to clad gap for two PHENIX fuel elements

For Viggen 6 and Papyrus pins which have reached about the same bum-up, the disparity in JOG thickness in the lower part of the fuel column is correlated to the absence or the presence of clad straining. This fact is well taken into account by GERMINAL, the agreement between measurements and calculated gap size are quite acceptable.

In both cases, discrepancies exist in the upper region of the fuel column which can be correlated to the presence of inner corrosion of the cladding. Produces of corrosion form a layer between the fuel and the cladding which increases the fuel to clad gap. Otherwise, this effect of inner corrosion of the cladding is not modelled in GERMINAL yet.

At very high burn-up levels, absence of cladding strain by swelling may have influence on the growth of the JOG layer. Variations of power during irradiation, but also shutdowns and restarts, induce stresses by fuel cladding differential expansion on the JOG layer. Good plastic properties of the JOG may lead to axial motions at levels along the fuel column where it is less stressed. Figure 5 shows this phenomenon which is particularly important for the LVD pin irradiated in PFR up to 23.1 at% with low clad straining (0.6%).

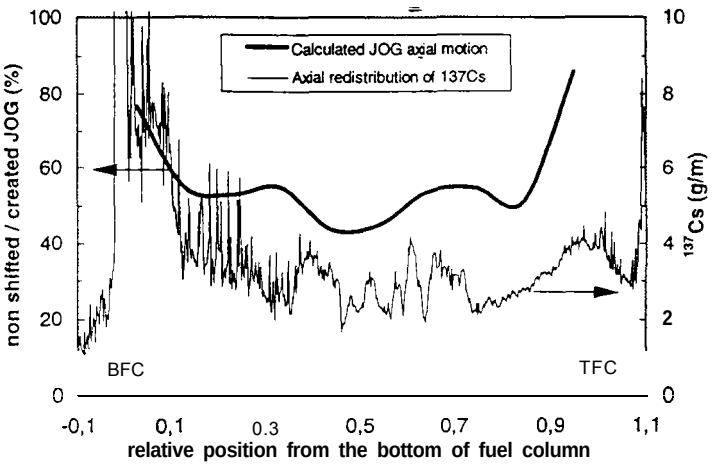

Fig 5 : correlation between the axial redistribution of ^{137}Cs and the calculated ratio of non displaced to created JOG for PFR LVD fuel pin irradiated up to 23. I at%

Axial migration of ^{137}Cs is very extensive with strong accumulations beyond both ends of the fuel column. As caesium is among the major constituents of the JOG, we can accept that a great part of the axial displacement of caesium is imputable to axial motion of JOG. The ratio of non displaced to created JOG calculated by GERMINAL follows the same feature.

4.3. **Plutonium redistribution**

Figure 6 shows for Viggen 6 (solid pellets) and ANT (annular pellets), the comparison between plutonium measurements by microprobe analysis (average on 3 radius) and the calculated radial plutonium redistribution.

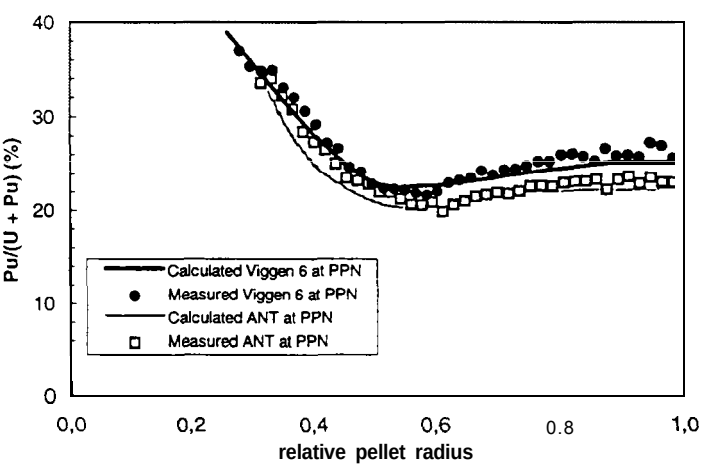

Fig 6 : comparison calculation/measurements for radial plutonium redistribution-for solid and annular pellet fuel pins

In both cases, calculations are in fair agreement with measurements.

4.4. Fuel behaviour during slow transients

For studying the behaviour of fuel pins during a control rod withdrawal accident, in which the power increase rate is typical 1%PN/s, a certain number of slow ramp tests were performed last years

in the CABRI reactor. Among others, these tests have allowed to enhance modelling of fuel swelling gas.

For example, figure 7 gives the evolution of the central hole size of a RIG 2 pin, irradiated at low rating (140 W/cm) during its end of life, before and after a slow ramp test in CABRI reactor (AGO test) up to 565 W/cm.

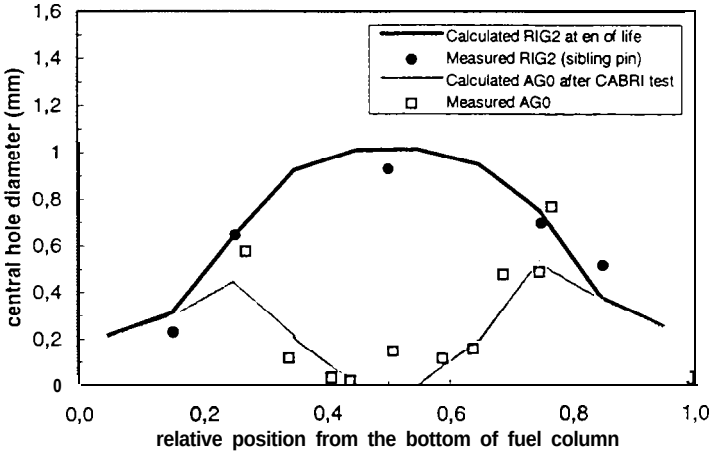

Fig 7 : comparison calculation/measurements for axial evolution of central hole diameter before and after slow transient test

The fission gas swelling is, in that case, particularly sizeable because of the strong ratio of retained gas in the fuel. This large swelling leads to the closure of the central hole in the mid zone of the fissile column, but without fuel melting. Calculations give a suitable description of this phenomenon.

The JOG (so called JOG1 and JOG2) tests have been performed in CABRI reactor with the goal to evaluate the thermal conductivity of JOG compounds [6]. JOG1 test was realised from a standard PHENM fuel pin irradiated up to 11.5 at% (CPd 6004). Figure 8 shows the comparison, after the JOG1 test, of fuel melting radius measured on optical ceramographies at several levels of the fuel column and the calculated solidus and liquidus radius from GERMINAL code.

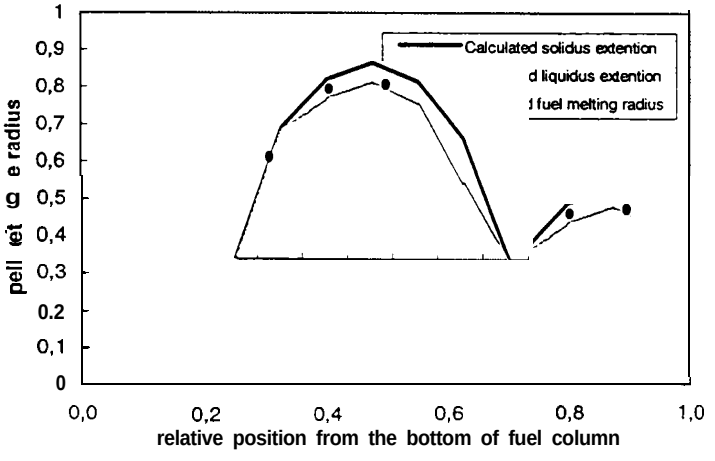

Fig 8 : comparison calculation/measurements for axial evolution of fuel melting radius after the CABRI JOG1 test

The good agreement between calculation and measurements allows to predict a power to melt with a suitable accuracy.

Recalculations of other similar tests have been performed with solid or annular fuels irradiated between 6 and 15 at% have also given satisfactory agreement (see table 2).

TABLE II. COMPARISON OF CALCULATED AND MEASURED MELTING RADIUS AFTER RAMP TESTS IN CABRI REACTOR

CABRI test I (fuel burn-up)	type of transient	maximum LR (W/cm)	Calculated relative solidus	Calculated relative liquidus	relative melting radius from ceramography
JOG1 (11.5)	low	773	0.48	0.43	0.46
JOG2 (15.5)	low	741	0.54	0.49	0.47
PFX (6.4) [a]	low	783	0.38	0.31	0.34
PF1 (6.4) [a]	low	885	0.63	0.58	0.64
E9 (5)"	low	1347	0.85	0.84	0.86
E5 (5)"	high		0.87	0.79	0.83

[a] Ramp tests from SPX1 type fuel pins

4.5. Evolution of MA doped fuel

Transmutation of minor actinides (MA) in FR may be obtained according to a homogeneous way which consists to add a low proportion of MA to a standard MOX FR fuel. Superfact experiment, irradiated in PHENIX reactor during 380 EFPD, was made of several MA fuel pins with 2% ^{241}Am or 2% ^{237}Np content. Table 3 summarises results concerning both the transmutation of these MA and the helium production.

TABLE III. MINOR ACTINIDES (Am AND Np) CONSUMPTION AND HELIUM PRODUCTION IN SUPERFACT HOMOGENEOUS FUEL PINS

	Superfact $(U, Pu, Am)O_2$ fuel pin ^{241}Am initial mass : 3.98 g	Superfact $(U, Pu, Np)O_2$ fuel pin ^{237}Np initial mass : 3.54 g
	^{241}Am final mass (g)	^{237}Np final mass (g)
Chemical analysis :	2.87±0.05	2.47±0.12
GERMINAL calculation :	2.98	2.41

	Helium (cm^3 STP)	Helium (cm^3 STP)
Pin puncture :	40	14.2
GERMINAL calculation :	60	15.1

The comparison between chemical analysis for MA and pin puncture measurements for helium with GERMINAL calculations is in satisfactory agreement. Discrepancy on helium evaluation for Am doped fuel pin may result to a proportion of retained He in the fuel during the 20 months separating the end of irradiation to the pin puncturing.

5 CONCLUSION

GERMINAL fast reactor fuel code builds up the synthesis of CEA knowledge on the fast reactor oxide fuel pin concerning the thermochemical and thermomecanical behaviour in steady state and incidental conditions. Since more 20 years, modelling improvements of the

code have been obtained as one goes along that us understanding of the physical phenomena and the experimental characterisation progressed.

At present, GERMINAL is able to predict, with a satisfactory confidence, the behaviour of FR oxide fuel element in a large range of composition and for various irradiation conditions up to strong burn-up levels.

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IN-CORE FUEL MANAGEMENT AND ADVANCED FUEL CYCLE OPTIONS

(Session IV)

Invited Paper

OVERVIEW OF NEUTRONIC FUEL ASSEMBLY DESIGN AND IN-CORE FUEL MANAGEMENT

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Abstract

The civil and military utilization of nuclear power results in stockpiles of spent fuel and separated plutonium. Recycling of the recovered plutonium in Light Water Reactors (LWR) is currently practiced in Belgium, France, Germany, and Switzerland, in Japan it is in preparation. Modern MOX fuel, with its optimized irradiation and reprocessing behavior, was introduced in 198 1. Since then, about 1700 MOX fuel assemblies of different mechanical and neutronic design were irradiated in commercial LWRs and reached fuel assembly averaged exposures of up to 51 .OOO MWd/t HM. MOX fuel assemblies reloaded in PWR have an average fissile plutonium content of up to 4.8 w/o. For BWR, the average fissile plutonium content in actual reloads is 3.0 w/o. Targets for the MOX fuel assembly design are the compatibility to uranium fuel assemblies with respect to their mechanical fuel rod and fuel assembly design, they should have no impact on the flexibility of the reactor operation, and its reload should be economically feasible. In either cycle independent safety analyses or individually for each designed core it has to be demonstrated that recycling cores meet the same safety criteria as uranium cores. The safety criteria are determined for normal operation and for operational as well as design basis transients. Experience with realized MOX core loadings confirms the reliability of the applied modern design codes. Studies for reloads of advanced MOX assemblies in LWRs demonstrate the feasibility of a future development of the thermal plutonium recycling. New concepts for the utilization of plutonium are under consideration and reveal an attractive potential for further developments on the plutonium exploitation sector.

1. INTRODUCTION

In the early years of the civil utilization of nuclear power, plans for optimized fuel cycle scenarios using a combination of breeder and fission reactors were developed. Essential for that scenario was a commercial reprocessing capacity for spent fuel large enough to establish a closed fuel cycle. Several countries developed the reprocessing technology. After most countries decided to delay

their fast breeder reactor program, only a few countries further developed the reprocessing technology. Today, France and Great Britain operate commercial reprocessing facilities, in Japan construction of a plant is in progress.

The civil and military utilization of nuclear power results in stockpiles of spent fuel and separated plutonium. Since commercial breeder reactors are not available, either the final disposal in geological formations or reprocessing and recycling of its valuable components are possible choices. The decision of how to dispose of the spent fuel depends on the local political situation in each country. In Belgium, France, Germany, Switzerland, and Japan plutonium recycling in thermal reactors is reality or is in preparation.

The status of the plutonium fuel assembly development and its operational qualification is presented in two other sessions of this conference (Refs [1,2]). This paper gives a survey of neutronic fuel assembly and core design with MOX fuel for LWR. It describes the experience gained in almost 30 years and the status reached today.

2. STATUS OF PLUTONIUM RECYCLING

The experience with modem MOX fuel assembly designs in PWR and BWR is depicted in Table I.

2.1. Belgium

The first MOX rods to be irradiated in a power reactor were loaded in 1963 in the BR3 reactor, an 1 1MW_e PWR. The number of MOX assemblies in that core was progressively increased and reached a ratio of 48 % in the last cycle in 1985. From the reprocessing of 530 t of spent fuel over a period of 10 years about 300 kg of fissile plutonium are recovered each year by the Belgian utility ELECTRABEL. Since 1995, MOX fuel is part of reloads for the PWRs Tihange 2 and Doe1 3. Typically, 8 or 12 MOX fuel assemblies are reloaded to the core each cycle [3]. Both units are licensed for an operation with up to 37 MOX fuel assemblies in the core.

2.2. France

In 1985 it was decided to recycle plutonium in French pressurized water reactors. FRAGEMA submitted a generic safety report for plutonium recycling for the 900 MW, plant type in 1986. In 1987 a license was granted for a maximum of 30 % MOX fuel assemblies per reload and a total of one-third of the core loading. In the same year the first 16 MOX assemblies were loaded to the St. Laurent Bl core. Currently, 20 plants out of twenty eight 900 MW, PWRs are licensed for plutonium recycling and the target is to reload about 100 t heavy metal per year of MOX fuel corresponding to the reprocessing of 850 t heavy metal of spent fuel.

2.3. Germany

Experience with the plutonium recycling technology in Germany dates back to the insertion of a lead assembly at the Kahl nuclear power plant (VAK, BWR) in 1966. First recycling programs on an industrial level were conducted at the Obrigheim nuclear power station (KWO, PWR) in 1972 and at the first Gundremmingen plant (KRB-A, BWR) in 1974. Since then 10 PWRs and 2 BWRs were licensed for the use of MOX fuel assemblies. Furthermore, for 3 PWRs and 2 BWRs licensing is in preparation. Cores with up to 50 % MOX fuel assemblies are licensed and core designs for PWR with ≃ 35 % MOX ratio have been realized.

2.4. Japan

In the Long-term Program for Research, Development and Utilization of Nuclear Energy by the Japanese Atomic Energy Commission it is stated that recycling of nuclear fuel including the use of MOX fuel in LWRs is national policy for the future energy security, economy and resource saving [4]. A commercial reprocessing plant with a capacity of 800 t of spent fuel per year is under

construction at Rokkasho-mura to be commissioned in 2005. The Japanese LWR MOX fuel demonstration was started in 1986 at Tsuruga Unit-l for BWR and in 1988 at Mihama Unit-l for PWR [5]. The first MOX fuel licensing was started in 1998 for both PWR and BWR plants.

2.5. Switzerland

The first MOX fuel assemblies were loaded to the Beznau Unit 1 in 1978 and in 1984 to Unit 2 (KKB, PWR). In 1996, a maximum of 26 % of the fuel assemblies in the core were MOX assemblies. The Goesgen nuclear power plant (KKG, PWR) started its recycling program in 1997. In addition to the commercial recycling programs, R&D work was performed at the Paul-Scherrer-Institut (PSI), Wtirenlingen. The international PROTEUS experiment investigated mainly the effect of tighter lattices and increased plutonium concentrations on the PWR design.

TABLE I. EXPERIENCE WITH MOX RELOADS IN BWR AND PWR FROM 1981 TO 1998

COUNTRY/ REACTOR -/ FA-TYPE	NO. OF REACTORS	NO. OF MOX FA RELOADED	MAX. AV. PU_{FISS} IN W/O / CARRIER MATERIAL	MAX. FA – EXPOSURE AT EOC IN MWD/THM
Belgium				
PWR (17x17-24)	2	56	4.9 / U_{tails}	43900
France				
PWR (17x17-24)	17	992	4.5 / U_{tails}	44250
Germanv				
PWR (18x18-24)	2	4	4.6 / U_{tails}	8000
PWR (16x16-20)	5	364	4.2 / U_{tails}	44900
PWR (15x15-20)	1	32	3 .o / U_{nat}	42000
PWR (14x14-16)	1	41	3.8 / U_{nat}	37000
BWR (9x9-l)	2	116	3.0 / U_{tails}	32000
Switzerland				
PWR (15x15-20)	1	28	4.8 / U_{tails}	23000
PWR (14x14-17)	2	152	4.1 / U_{tails}	51000

2.6. Others

In the US experience with MOX fuel was gained mainly throughout the 1960s and 1970s. During that time MOX assemblies were part of reloads for PWRs and a BWR. After the political decision was made not to pursue the recycling of plutonium, work on that topic was suspended. Today, the reduction of stockpiles of weapons grade plutonium from the dismantlement of nuclear warheads is part of the strategic arms reduction treaty. Recycling of that material in LWRs is considered as one way to assure its peaceful use.

Investigations on MOX fuel were performed in the United Kingdom and countries of the former Soviet Union. It was concluded that AGRs and VVERs are suited for being loaded with a certain quantity of MOX fuel assemblies, but no commercial recycle activity in these reactors was reported. In the United Kingdom a commercial reprocessing facility and a MOX fuel fabrication plant are in operation.

3.1. Early Qualification Steps

Neutronic codes are qualified by comparison of the derived results with measurements and by theoretical benchmarks. For first approaches to the MOX technology, the experience gained with the neutronic properties of depleted uranium fuel was extrapolated. This was a reasonable approach, since more than 50 % of the energy in high exposure uranium fuel is produced by the plutonium isotopes ^{239}Pu and ^{241}Pu. The first explicit qualification steps for MOX fuel were performed in the 1960s. Safety and feasibility of reactor quantities of MOX fuel was demonstrated in the late 1960s and early 1970s in zero power critical facilities, like the Plutonium Recycle Critical Facility (PRCF) in Hanford, USA, the KRITZ facility in Studsvik, Sweden or the VENUS critical facility at CEN/SCK in Mol, Belgium.

These experiments were especially planned for the basic qualification of cross sections and codes. For handling and safeguards reasons, the measurements were performed on fresh fuel. Typical reload enrichments at that time were 3.0 to 3.2 w/o ^{235}U and consequently the matching average concentrations of fissile plutonium were in the range of 2 to 3 w/o, equivalent to plutonium concentrations of 3 to 4 w/o. The experiments were designed to represent the physical and neutronic conditions of PWRs and BWRs as closely as possible. Investigated were effects of varying plutonium compositions and the temperature dependence of the reactivity of the designed cores. The measurement of pinwise fission rate distributions in configurations with central MOX fuel assemblies surrounded by uranium fuel assemblies was another important aspect of the experiments. Confidence in the calculational methods was gained by the recalculation of those measurements and the demonstration of an adequate accuracy.

3.2. Actual Status of Design Method Qualification

Since then, measurements from operating power reactors contributed to the verification of the design codes and were the basis for required updates of the cross section data base. These data comprise results from routinely performed startup and core follow measurements as well as from especially defined programs for the verification of specific aspects, e.g. the comparison of the control rod worth in uranium and MOX fuel assemblies. Reference [6] gives a comprehensive overview on existing experimental data. For the determination of the accuracy of predicted local power density distributions, aside from critical experiments, γ-scan measurements have been conducted on irradiated PWR and BWR MOX fuel assemblies at the reactor site. More recently, additional

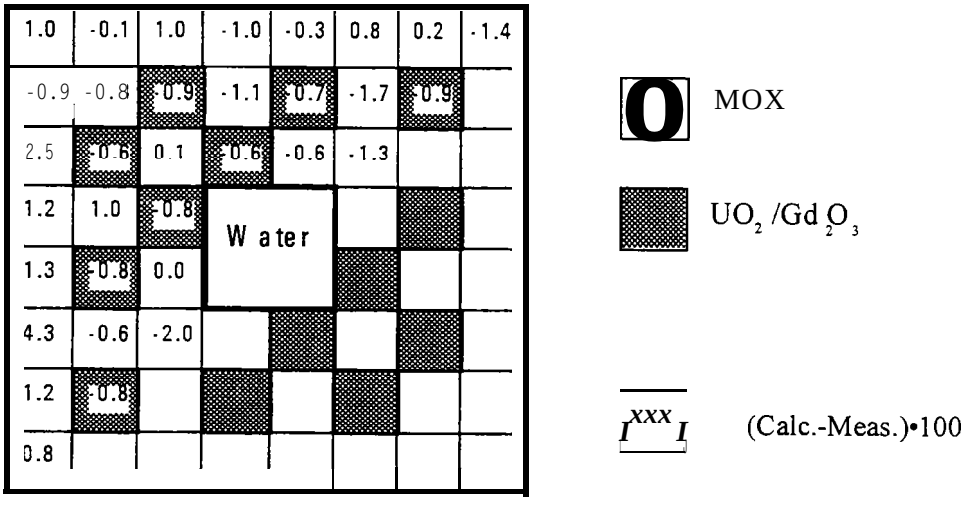

FIG. I VENUS BWR UO_2/MOX Mockup Core Comparison of Measured and Calculated Fission Rates

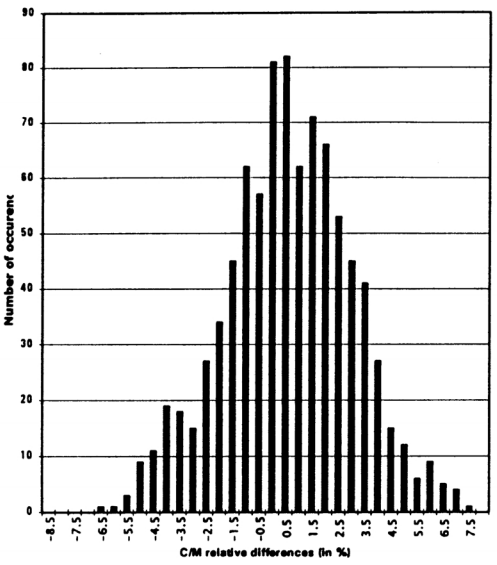

FIG. 2 PWR - Comparison of Calculated and Measured Detector Signals

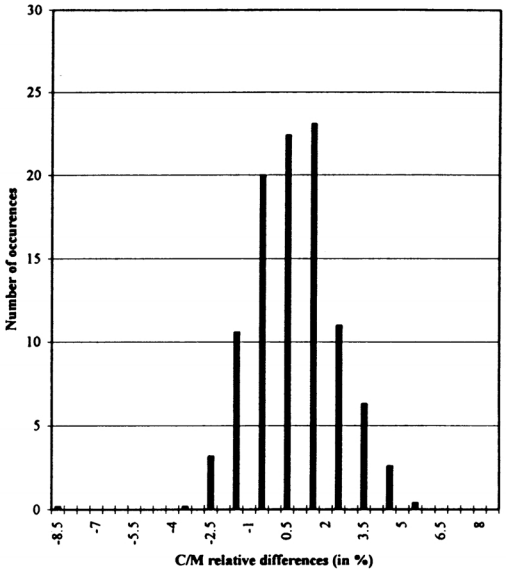

FIG. 3 BWR - Comparison of Calculated and Measured γ- Tip Signals

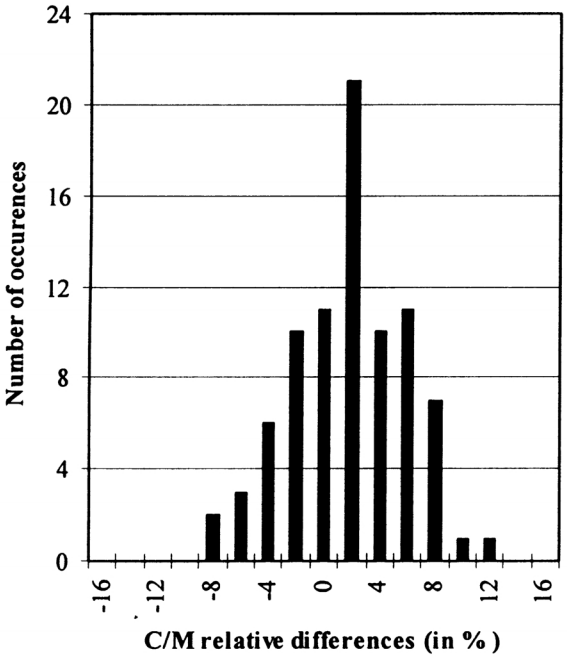

FIG. 4 PWR - Comparison of Calculated and Measured Control Rod Worths

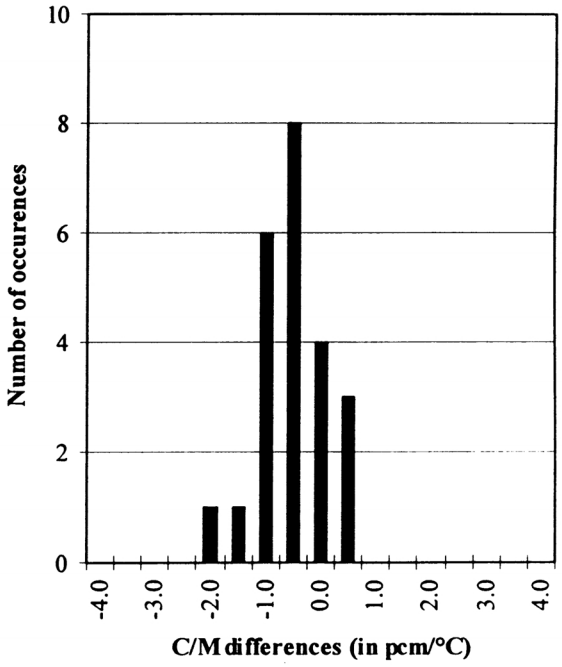

FIG. 5 PWR - Comparison of Calculated and Measured Moderator Temperature Coefficients

experiments and measurements (e.g. PROTEUS, VENUS, EPICURE, Refs 7 - 93) were performed to address the effect of design parameter changes like changed moderation ratio, increased exposures or higher plutonium contents. Some of those activities were conducted in international cooperation.

Modem core design with its higher enrichments of uranium and MOX fuel assemblies has become more demanding with respect to accuracy and reliability of the design codes. Qualified codes prove their capability of predicting safety parameters for a large variety of core designs every year. Current nuclear design methods for both PWR and BWR MOX fuel employ the standard calculational methods also applied for UO_2 fuel. Advanced features for MOX fuel description were introduced by some vendors, e.g. updated nuclear cross section data, corrections derived from colorset calculations, or extensions to the nodal core calculation methods.

The international standard for neutronic fuel assembly design is the use of 2-dimensional multigroup transport codes. These codes determine the flux and power density in either the homogeneously or heterogeneously represented pins and provide condensed and homogenized few group (typically 2 energy groups) cross sections for the reactor codes. The multigroup cross section data used in today's production codes for uranium and MOX design are generally based on data files, like ENDF/B [10], JENDL [11], or JEF [12]. The derived cross section libraries utilized in the spectral codes are qualified by the recalculation of a large variety of measurements and suited for the description of actual core designs. Three dimensional coarse mesh methods with pin power reconstruction techniques are applied to steady state and transient reactor calculations for PWRs and BWRs. These methods have proved to be adequate, accurate and efficient.

Fig. 1 demonstrates the level of accuracy reached with the recalculation of pinwise fission rate distributions measured in critical experiments. The VENUS BWR UO_2/MOX mock-up, selected as an example out of a series of similar experiments (Ref [13]), used an 8x8 fuel bundle geometry with a central water rod. The maximum plutonium concentration in the MOX rods in this example was 9.9 w/o Pu_{fiss}. The calculated data was derived from a 2-dimensional transport calculation. Benchmarking of the PWR design codes on UO_2/MOX mock-ups with high plutonium concentrations confirms the same level of accuracy (e.g. Ref [14]). Fig. 2 shows an example of the quality of prediction of axially integrated detector signals in MOX fuel assemblies for PWR, Fig. 3 an example for γ-tip-measurements in BWR. A comparison of measured and calculated control rod worths and moderator temperature coefficients in PWR, including cores with MOX assemblies, is depicted in Fig. 4 and 5.

The conclusion from those comparisons is that the prediction of key safety parameters for actual cores with MOX fuel assemblies is of the same quality as for uranium cores. However, taking into account a smaller data base with measurements for cores with MOX fuel and specified manufacturing tolerances for plutonium concentration and composition, specific tolerance factors may be applied to certain predicted safety parameters in MOX fuel assemblies (e.g. linear heat generation rate, DNBR).

Special consideration should be given to minor isotopes. These isotopes can be described as of minor important for today's neutronic MOX fuel assembly and core design but may have impact on future developments or on radioactivity aspects. One representative of that group of isotopes is ^{242}Pu, which will become more important with higher plutonium concentrations and the degradation of the plutonium quality. Differences in cross section data for curium and some americium isotopes are found by comparisons of different data files. An improved representation in future cross section data files will become important for high burnup MOX designs.

4. NEUTRONIC FUEL ASSEMBLY DESIGN

4.1. General Aspects and Design Targets

The nuclear characteristic of MOX fuel is a result of the neutronic properties of the different plutonium isotopes. The n-values for thermal neutrons of the fissile plutonium isotopes are higher

than that of ^{235}U, where both fission and capture cross sections are about a factor of 2 larger. The absorption cross sections of the plutonium isotopes ^{240}Pu and ^{242}Pu show strong resonance peaks in the near thermal region. As a result, the neutron spectrum in MOX fuel is hardened, i.e. at the same power level the thermal neutron flux is much lower than in uranium fuel (see Figure 6).

In the past, studies were performed to find technically and economically optimized MOX fuel assemblies. The issue of those studies was that despite of its different neutronic characteristics no changes in the mechanical and thermal hydraulic designs are required in comparison to the uranium assemblies loaded to the core at the same time. Only slightly different properties of the (U, Pu)O_2 fuel, e.g. melting temperature and thermal conductivity, do not require changes of the fuel rod design. With an unmodified mechanical fuel rod and assembly design the following targets were addressed in neutronic design studies:

(1) The discharge burnup of the MOX fuel assemblies should match closely that of the uranium fuel assemblies; or

(2) The average plutonium content was chosen as high as reasonable to reduce the number of required MOX assemblies.

These design criteria were complemented by more general demands. MOX fuel assemblies should be compatible to uranium assemblies with respect to loading strategies and the number of possible in-core cycles without additional restraints for their operation.

Neutronic fuel assembly and core design cannot be seen as independent processes. Core design calculations are used to determine the appropriate average plutonium concentration for the MOX fuel assemblies required to meet the design goals. The neutronic MOX fuel assembly design procedure starts after the average plutonium content is defined. Then again, the result is confirmed with core design calculations.

The neutronic MOX fuel assembly design for thermal reactors has to face large gradients of the thermal flux at the interface of MOX and uranium fuel assemblies. The fact of an increasing thermal flux in the direction of the adjacent uranium fuel assembly is addressed by a gradation of the

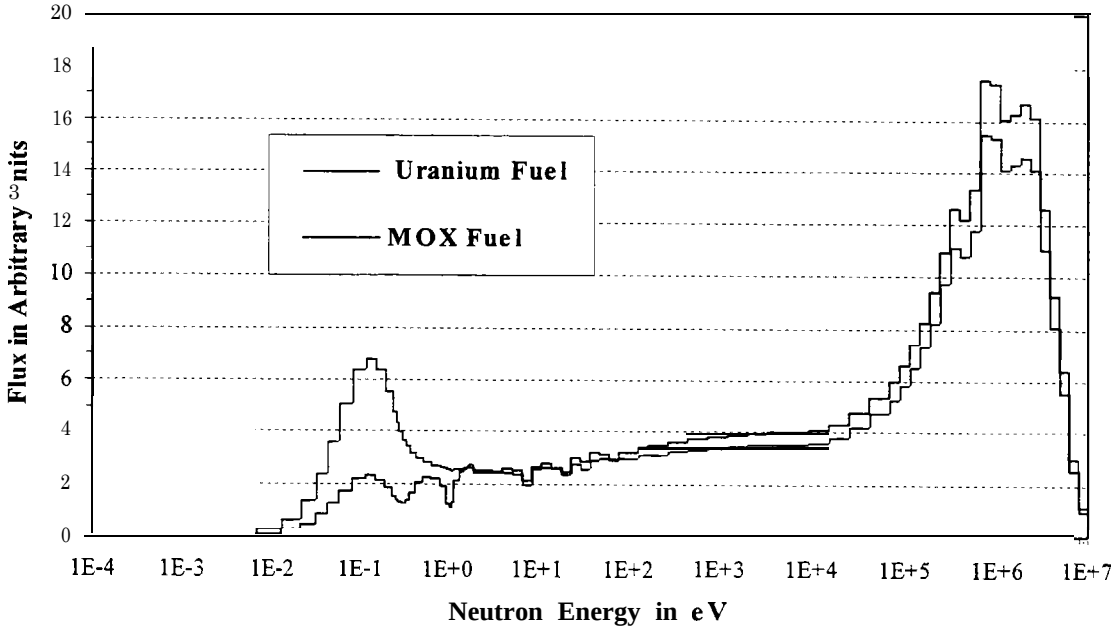

FIG. 6 Neutron Flux vs. Fission Neutron Energy in Uranium and MOX Fuel (PWR)

plutonium content in the fuel rods at the fuel assembly edge. Two to 3 rod types are typical for MOX fuel assemblies used in the last 25 years of plutonium recycling in PWR. Optimized BWR fuel assemblies are more heterogeneous. Larger water gaps and water structures within the fuel assembly result in MOX fuel assembly designs with 4 to 7 different rod types. The 'all plutonium' assembly, comprising of MOX fuel rods only, is appropriate for PWRs and BWRs. In comparison to the earlier 'island' type MOX fuel assembly design, with MOX rods in the center and uranium rods at the periphery of the assembly, it has clear economic advantages. The additional costs associated with fabrication and transport are then restricted to a minimized number of fuel assemblies.

For PWRs, MOX fuel assemblies are currently designed without burnable absorbers, whereas for BWRs neutron poisons are normally required in all fuel bundles. The exposure dependent reactivity behavior of the BWR fuel bundles is optimized by using UO_2/Gd_2O_3 rods. MOX fuel rods are generally unpoisoned. Optimized BWR MOX bundles are designed with a limited number of gadolinium rods and a minimum of different rod types to gain the maximum economic advantages. Those designs are still considered as of the 'all plutonium' type.

4.2. MOX Carrier Material and Plutonium Composition

The carrier material for the plutonium and the plutonium composition are important parameters for the MOX fuel assembly design. Both have impact on the reactivity characteristic of the MOX fuel.

4.2.1. The carrier material for the plutonium

Three options for carrier material were investigated for the commercial MOX recycling strategies - natural uranium (U_{nat}), tails uranium (U_{tails}) from the enrichment process and depleted uranium separated in the reprocessing process. First designs were based on natural uranium. Actual MOX fuel is mainly fabricated with tails uranium as carrier. For economic reasons, typical ^{235}U enrichments in the tails assay are 0.2 - 0.3 w/o. The somewhat lower ^{235}U enrichment compared to natural uranium is compensated for by an increase of the plutonium content of 0.3 - 0.4 w/o Pu_{fiss}. Depleted uranium was used in test assemblies only.

4.2.2. The plutonium composition

The isotopic composition of the plutonium is determined by the origin, the initial enrichment, the discharge exposure and the intermediate storage time of the reprocessed fuel. So far, the commercial plutonium used for the manufacturing of MOX fuel was mainly generated in uranium fuel assemblies. Piutonium from spent MOX assemblies was used for demonstration purposes only. The plutonium quality can be defined as ratio of the fissile isotopes in the isotopic composition to the total plutonium content. Table II shows examples for plutonium qualities employed in MOX fuel

TABLE II. EXAMPLES OF PLUTONIUM QUALITIES USED FOR MOX FUEL FABRICATION

Plutonium/ Pu-Quality	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
			in w/o		
Magnox-Pu/ *78.9*	0.3	74.3	19.9	4.6	0.9
LWR-Pu/ *68.9*	1.5	60.1	24.5	8.8	5.0
2nd Generation-Pu/ 58.0	1.3	43.8	34.3	14.2	6.4
Weapons grade Pu/ 95*)	≈ 0	95	5	≈ 0	≈ 0

*) weapon grade plutonium, so far not used for the fabrication of commercial MOX fuel
Pu-Quality = $(^{239}Pu + ^{241}Pu)*100/Pu_{tot}$

fabrication. Variations of the plutonium quality affect the exposure dependent reactivity of the MOX fuel. The higher the plutonium quality the higher is the beginning of life reactivity and the larger is the slope of the reactivity vs. exposure. This has impact on the average plutonium concentration required for meeting the basic design goals. Fuel assembly design is not sensitive to reasonable changes of the plutonium quality. New optimization of the enrichment zones is only required if the recalculation of the fuel assembly design results in unacceptable power density distributions or if a change in the average plutonium concentration is required due to safety evaluations. Independent of neutronic aspects related to the core, variations in the plutonium quality have to be considered for handling and storage of the MOX fuel assemblies.

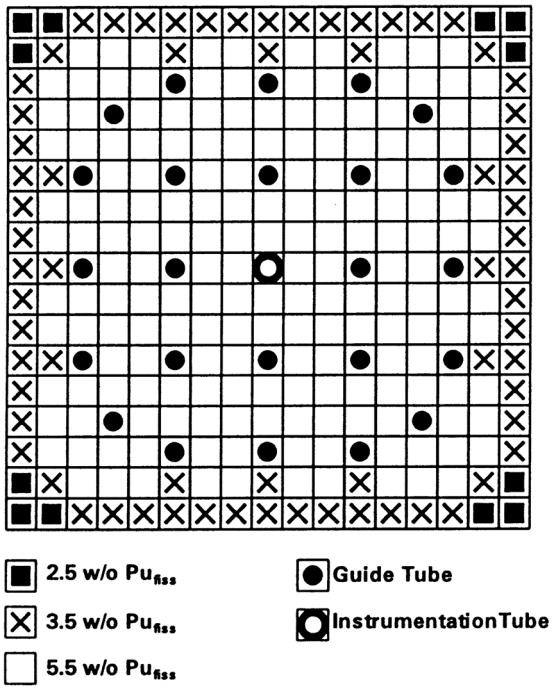

FIG. 7 MOX Fuel Assembly for the Reload in Belgian 900MW$_e$ PWRs

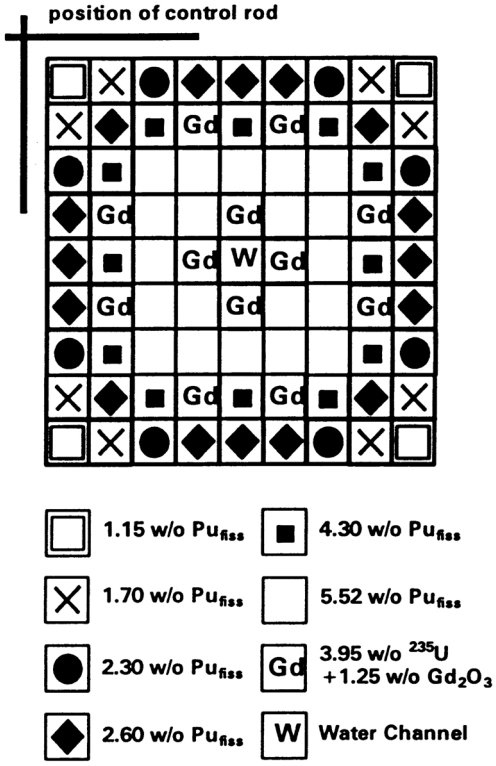

FIG. 8 MOX Fuel Assembly for the Reload in German 1300MW$_e$ BWRs

4.3. Examples of MOX Fuel Assembly Designs for PWR and BWR

Figures 7 and 8 show examples of MOX fuel assemblies designed for actual reloads for PWR and BWR in Belgium and Germany.

Modern MOX fuel assembly designs utilize central water structures like water channels for BWR or additional 'water rods' for PWR. This additional moderator increases the reactivity of the undermoderated MOX rods and flattens the power density distribution in the fuel assembly without introducing new rod types with increased plutonium concentrations. This design feature became particularly important with increasing plutonium concentrations and exposures. A reasonably flat power density distribution in a uranium fuel assembly environment limits the maximum exposure values and is a prerequisite for meeting the mechanical design criteria and for enough operational flexibility.

In BWR bundles the reduced reactivity worth of the gadolinia rods is to a certain extent offset by placing them near regions with high moderation, i.e. adjacent to the central water structures [15].

Under actual licensing conditions MOX fuel assemblies are loaded into the core together with uranium fuel assemblies. Design criteria for these MOX assemblies have to be defined in harmony with those for the uranium assemblies. For safe and economic reactor operation it is essential to meet those criteria and to provide enough operational margins.

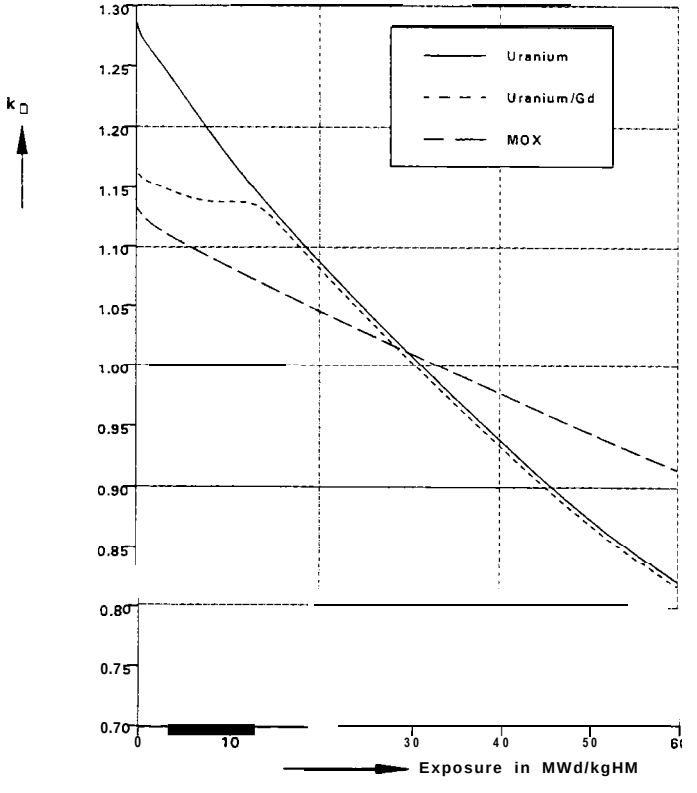

FIG. 9 k-infinite vs. Exposure for U, U/Gd and MOX Fuel (PWR)

The historical development of neutronic core design is characterized by an increase of the reload enrichments and, consequently, an increase of the discharge burnup. In the 1970s typical reload enrichments were 3.1 - 3.5 w/o ^{235}U. In the meantime many reactors worldwide are licensed for enrichments of 4.0 w/o ^{235}U and higher. In conjunction with low leakage loadings utilizing burnable absorbers, batch averaged exposures of 46 MWd/kg in PWRs and 43 MWd/kg in BWRs have been realized. MOX fuel assemblies had to follow that trend. An increasing content of fissile plutonium had to be used in order to match the increased uranium enrichments.

Today, MOX fuel assemblies with an average fissile plutonium content of up to 4.8 w/o (7 w/o Pu) are part of reloads for PWRs in Belgium and Switzerland. For German BWR, the average fissile plutonium content in actual reloads is 3.0 w/o (4.3 w/o Pu) with a maximum of 5.5 w/o in the highest enriched rods. BWR MOX bundles with an average fissile plutonium content of 3.6 w/o to be reloaded in 1999 are in fabrication.

Currently, for unchanged lattices the plutonium content in a fuel assembly should not exceed 13 w/o. Benchmarking calculations for infinite MOX lattices indicated that for plutonium concentrations greater than 13 w/o, k-infinite for the voided case may become higher than for moderated conditions, corresponding to a positive void reactivity coefficient. Further investigations are needed for qualification of nuclear data and codes in that point [6].

5.1. Reload Strategies

Parallel to the increase of the reload enrichments, out-in core loading strategies, where fresh fuel assemblies were positioned at the core periphery, were replaced by low-leakage loadings with its significantly increased fuel economy. The low-leakage ratio can be defined as number of reload assemblies not facing the core periphery divided by the total number of reload fuel assemblies. The higher the low-leakage ratio the more fuel assemblies need to be limited in its initial reactivity. This can be achieved either by a split of the reload enrichment, or far more common, by the use of burnable absorbers. For PWR the exposure dependent reactivity characteristic of MOX fuel (see Figure 9) allows to substitute fresh uranium fuel assemblies with burnable absorbers by MOX assemblies. Higher reactivity values and therefore higher power densities at higher exposures in comparison to uranium assemblies are unfavorable for waterside cladding corrosion and fission gas pressure buildup. These phenomena have to be considered for optimized core loadings. In BWR, with an adequate fuel bundle design, recycling cores don't require significant adjustments of the reload strategy in comparison to all uranium core designs. [16, 17]

Figure 10 shows an example of a core design realized in a Belgian 900 MW, PWR. The prediction standard for local power is demonstrated by the comparison of axially integrated measured and calculated detector signals, depicted in the same figure. The MOX fuel assemblies of that core were designed in compliance with the safety considerations of the licensing authority and the economics considerations of the utility. The content of fissile plutonium in those MOX fuel assemblies is 4.8 w/o with U_{tails} as carrier material.

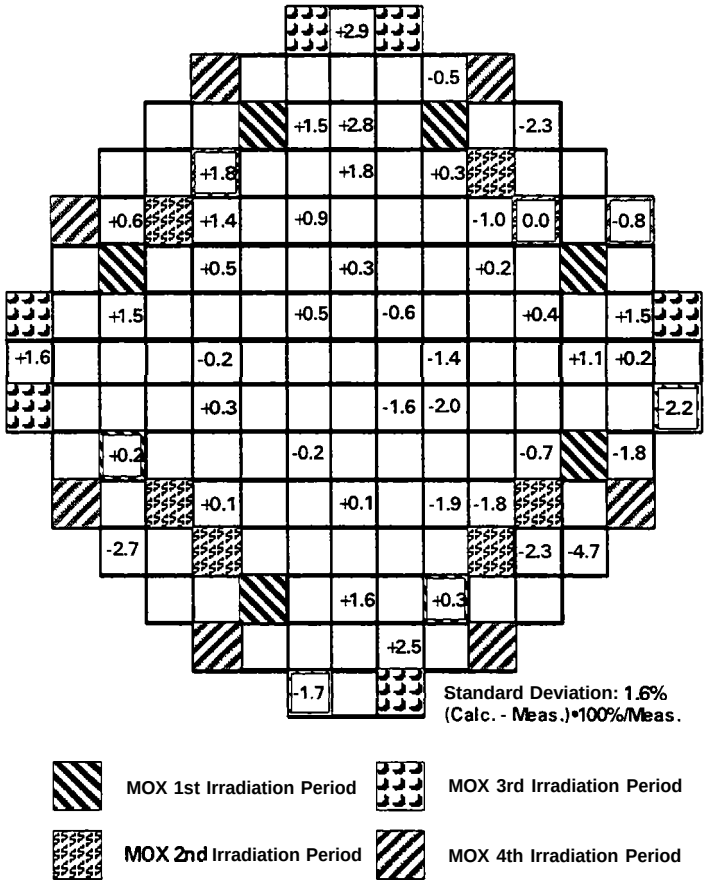

FIG. IO Comparison of Calculated and Measured Detector Signals in a MOX Loaded 900 MW_e PWR

Those recycling cores have to meet the same safetqfcriteria as uranium cores. This has to be demonstrated either for each designed core or in cycle independent safety analyses [18, 19]. The safety criteria are determined for normal operation and operational or design basis transients (e.g. main steam line break accident, LOCA, ATWS or external events). Some of the neutronic aspects affected by plutonium recycling are discussed in the following sections.

5.2. MOX Impact on Normal Operation

5.2.1. PWR

The control rod worth in MOX fuel assemblies is smaller than in uranium fuel. This effect is most pronounced in fresh fuel. It declines with decreasing moderator temperature and increasing exposure. Core design has to account for that. For the actually licensed ratio of MOX assemblies in the core, results from studies indicated that the influence on the net control rod worth (stuck rod configuration) caused by changes in the core loading pattern is larger than the MOX influence. A tentatively smaller bank worth is often accompanied by a reduced stuck rod worth and results then in an almost unchanged net control rod worth.

The boron system is not only used for continuous reactivity control under normal operating conditions but is also utilized for the long term shut down of the plants. The reduced boron reactivity worth can be compensated for, depending on the number and design of the loaded MOX fuel assemblies, by an increase of the boron concentration in the storage tanks or by an increase of the ^{10}B-enrichment in the borated water bearing systems.

The limits for local power, $F\Delta H$, or DNBR remain unchanged. Incore or excore systems are set to the same limiting values as for uranium cores.

5.2.2. BWR

The impact of MOX fuel assemblies on the control rod system is small. The thermal flux recovers in the large water gaps between the bundles and results in almost unchanged control rod worths. Hence, cores are normally designed with a scattered distribution of the MOX assemblies with only one or two MOX assemblies assigned to each control rod.

5.3. MOX Impact on Transients

5.3.1. Reactor Kinetics

The effect of a reduced delayed neutron fraction by introducing MOX fuel impacts fast power transients and reactivity initiated accidents (RIA). The core power responds more rapidly to perturbations of the reactivity. Part of that effect is compensated for by a slightly more negative Doppler coefficient. Safety analyses are generally performed by assuming a conservatively low fraction of delayed neutrons.

5.3.2. Decay Heat Power

Decay heat has impact on large break loss of coolant accidents. Directly after shut down, decay heat power is smaller for MOX fuel than for uranium fuel. With time, the decay heat is more and more determined by the actinides and, after the decay of the short living fission products, becomes therefore higher in MOX fuel. The differences in the decay heat are of only little importance for the maximum cladding temperature reached during large break LOCA accidents.

5.3.3. PWR

More negative moderator temperature coefficients have to be considered in cold shutdown reactivity balances and transients with moderator cool down in conjunction with the tendency to a reduced bank worth and a smaller boron worth.

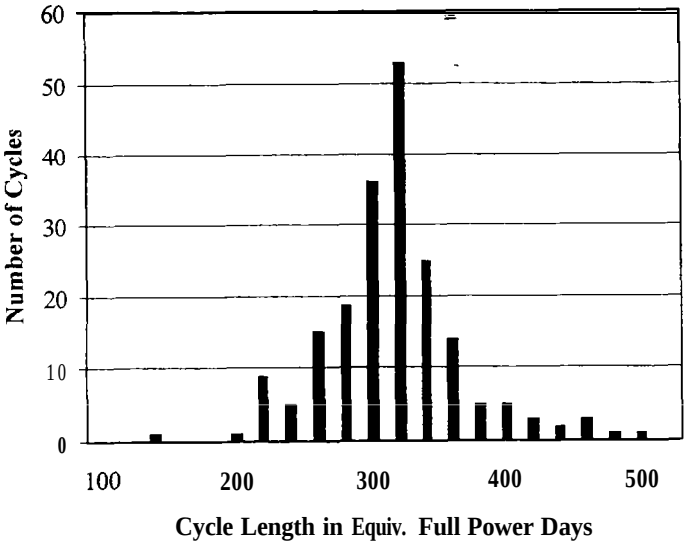

FIG. 11 Cycle Length Variation in German PWRs (Status: 1998)

5.3.4. *BWR*

The more negative moderator temperature and void coefficients affect the cold shutdown reactivity balances, transients with moderator cool down and transients with coolant pressure increase. MOX fuel with very high concentrations of plutonium may cause a shift to less negative values of the void coefficient [6].

5.4. **Operational Flexibility**

The operation of nuclear power plants is determined by the demands of the electricity suppliers. Load follow operation, variations in cycle length and reload batch size, but also unplanned outages are challenges for the flexibility of fuel assembly and core design. MOX fuel assemblies are providing the required flexibility under those boundary conditions. Fig. 11 gives an overview of cycle lengths realized with German PWRs [20].

6. SAFEGUARDS ASPECTS

The IAEA safeguards criteria for fresh MOX fuel assemblies are more restrictive than for normal uranium fuel assemblies. MOX fuel assemblies are transported in specially developed, sealed containers with safety trucks. After arriving at the plant site the seal is removed by an IAEA representative. Radioactivity and heat production make it advisable to store the MOX assemblies directly in the storage pool. Continuous video surveillance and a monthly inspection by an IAEA inspector make sure that no unauthorized manipulations occur. At some plants fresh MOX assemblies are stored in the dry storage facility. Then, each assembly is separately sealed and regularly inspected. Core loading is performed under surveillance of an IAEA inspector. No additional measures are required for the transportation of discharged MOX fuel assemblies relative to discharged uranium fuel assemblies.

7. CONCLUSION

In-core fuel management with MOX assemblies is common practice in a large number of nuclear power plants in several countries. A ratio of about 30 % (50 % in some plants) of MOX fuel assemblies in the core is possible while meeting applicable safety criteria. Experience with such core

loadings confirms the reliability of the applied modem design codes. The applicability of methods used for neutronic fuel assembly and core design has been proven for actual plutonium concentrations and compositions used.

Plans for a further increase of plutonium concentrations and the use of degraded or weapons grade plutonium qualities should be accompanied by

- a continuous verification of the utilized nuclear data and design methods.

- post irradiation examinations, i.e. isotopic measurements on highly depleted MOX fuel and, if possible, critical experiments with depleted fuel.

Conceptual studies for reloads of advanced MOX assemblies in LWRs demonstrate the feasibility of a future development of the thermal plutonium recycling. Hence, new concepts for the utilization of plutonium are under consideration or investigation and reveal an attractive potential for further developments on the plutonium exploitation sector.

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EXPERIENCE OF MOX-FUEL OPERATION IN THE GRUNDREMMINGEN BWR PLANT: NUCLEAR CHARACTERISTICS AND IN-CORE FUEL MANAGEMENT

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Abstract

After 4 years of good experience with MOX-fuel operation in the BWR plants Gundremmingen unit B and C the number of inserted MOX-FAs will be increased in the future continuously. Until now all MOX-FAs are in good condition. Furthermore calculations and measurements concerning zero power tests and tip measurements are in good agreement as expected: All results lead to the conclusion that MOX-FAs can be calculated with the same precision as uranium-FAs.

Today the NPP Gundremmingen with two 1344 MW_{el} units is the only commercial BWR plant in the world operating with MOX-fuel – after a five years lasting licensing procedure. In Gundremmingen recycling of plutonium started in 1995 with 16 9x9-MOX-fuel assemblies (FAs) in unit C. One year later unit B followed with the insertion of 32 9x9-MOX-FAs designed by Siemens, 16 of them fabricated by BN at Dessel in Belgium and 16 by Siemens at Hanau in Germany. In unit B the number of MOX-assemblies was increased continuously in the following cycles (see Fig. 1): presently (spring 1999) 124 MOX-FAs are in the core of unit B. The maximum number of MOX-FAs in one core is limited by license to 300 corresponding to 38% of the whole core. Up to now the leading MOX-FAs have reached burn-ups of up to 42 MWd/kg.

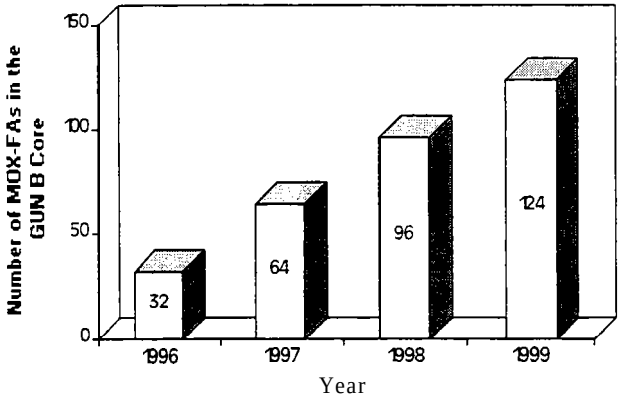

Fig. 1: Development of the number of MOX-FAs in the BWR unit Gundremmingen B

All MOX-FAs contain 1 water rod, 12 U/Gd-rods and 68 MOX-rods with 6 different enrichment levels. The 16 first fabricated MOX-FAs have an initial average amount of fissionable plutonium of 2.2 w/o in natural uranium which corresponds to about 4 kg Pu_{fiss}/FA. For all subsequent MOX-FAs the average Pu_{fiss}-content is 3.0 w/o in tails-uranium corresponding to about 5 kg Pu_{fiss}/FA; their radial rod type distribution is shown in Fig. 2. The initial Pu_{fiss}-content of the MOX-rods is in the range of 1 to 6 w/o. The isotopic plutonium composition corresponds to Pu which has been recovered from LWR uranium fuel with a burn-up in the range of 35 MWd/kg U.

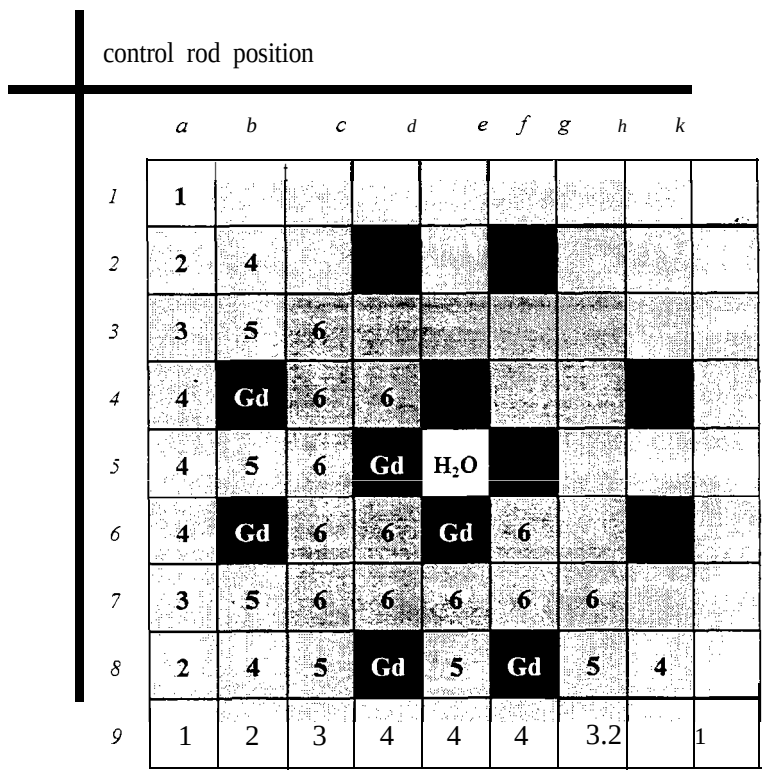

Fig. 2: 9x9 MOX-FA of Gundremmingen with 3 .O w/o Pu_{fiss} in U_{tails}

Compared to U235 and U238 the absorption cross sections of the main Pu isotopes show pronounced resonances in the thermal energy range. Therefore the thermal neutron flux in the interior of the MOX-FA is significantly lower than in the burn-up equivalent 9x9 uranium FA (Fig. 3). Due to the water gap between the FAs the thermal neutron flux increases at the periphery. For the MOX-FA this increase is enhanced if uranium-FAs are positioned next to a MOX-FA. This is shown in Fig. 3. where for the MOX-FA the results of CASMO single-bundle and four-bundle calculations are given. The four-bundle cell (with periodic boundary conditions) is composed of 1 MOX-FA and 3 uranium-FAs.

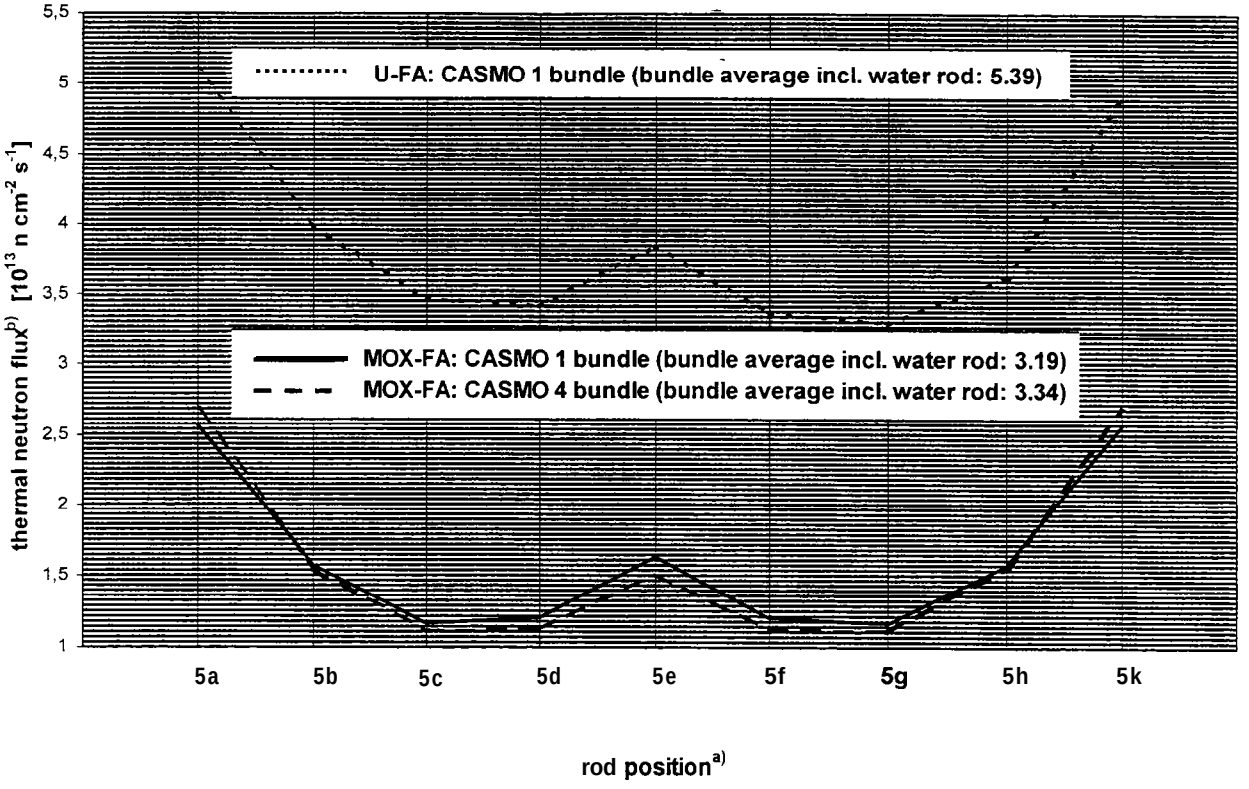

Fig. 3: Thermal neutron flux along row No. 5 for the unit-radiated MOX- and uranium-FA, each at full power conditions with 45 % void

[a] see Fig. 2

[b] $E_n \leq 1.85$ eV

Well known effects, which result from the harder neutron energy spectrum in the MOX-FA (in comparison to the uranium-FA) are

- the flatter k_{oo}-curve (Fig. 4)
- the reduced gadolinium worth (compare k_{oo} in Fig. 4 for zero burn-up)
- the reduced control rod worth (Fig. 5).

A further feature of MOX-FAs is that - in **contrast** to fissioning of U235 and U238 - fissioning of Pu239 and Pu241 yields significantly smaller amounts of so-called precursor fission products which are responsible for the production of the delayed fission neutrons. Hence the delayed fission neutron fraction of MOX-fuel is lower than in uranium-fuel. This is of importance for transients and accident analysis.

Allthough both FA-types have the same number of enrichment levels the local power peaking as well as the resulting R-factors (for dry-out calculations) are higher in the MOX- FA than in the uranium-FA. Therefore most of these MOX-FAs have to be placed in the outer region of the reactor core to reduce radial peaking factors.

Up to now the incore fuel management for Gundremmingen is performed by NIS with the 1.5 group core simulator COSIMA *[1]*. The nuclear data bases used in COSIMA for the various FA-types are generated with CASMO *[2]*, i.e. CASMO one-bundle calculations for the uranium-FAs and CASMO four-bundle calculations for the MOX-FAs (1 MOX-FA surrounded by typical uranium neighbours). The decision for four-bundle calculations in case of MOX-FAs is based on a simplified core benchmark which was considered both by CASMO and by COSIMA (Fig.5).

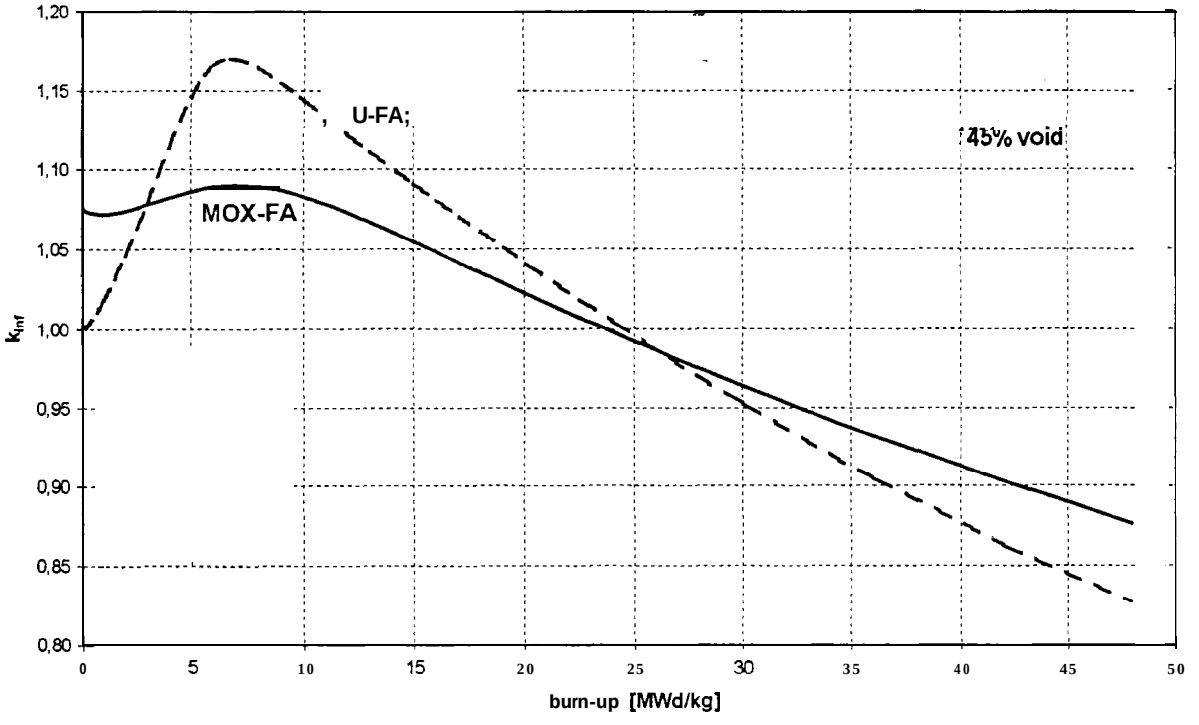

Fig. 4: Comparison of k_{inf} vs. burn-up of 9x9-MOX and 9x9-U FAs, each at full power conditions with 45% void

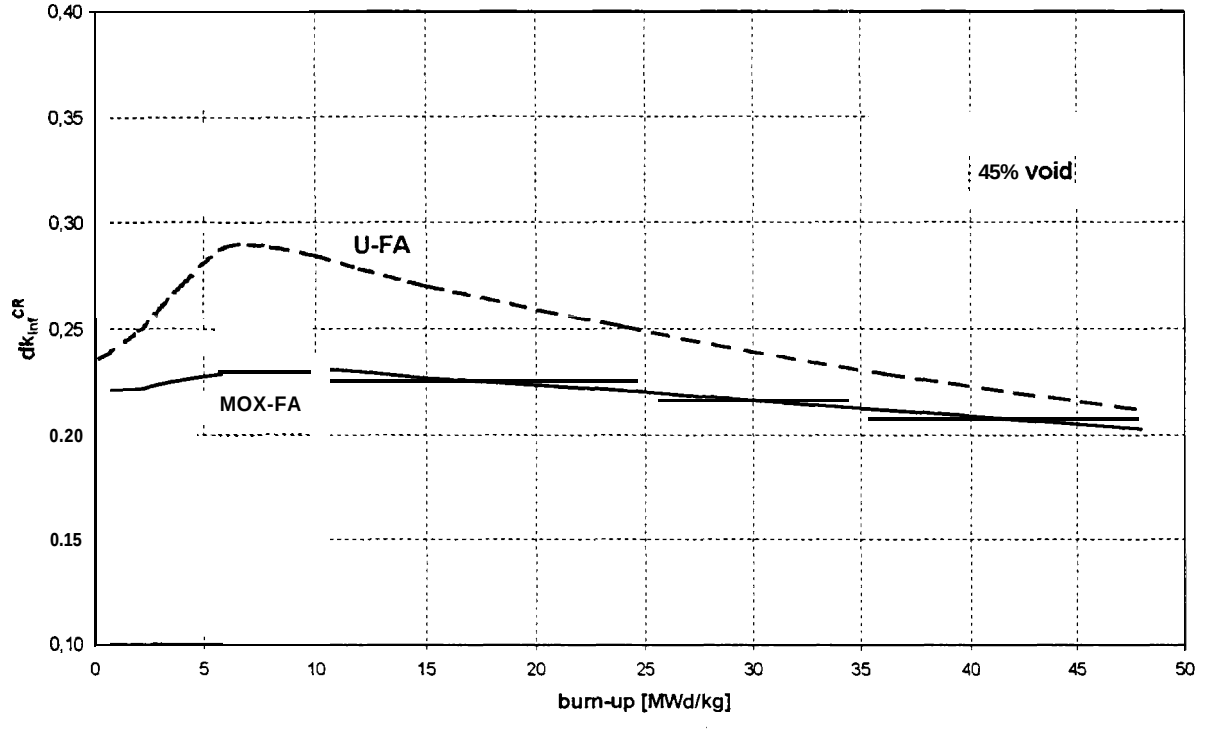

Fig. 5: Comparison of the control rod worth dk_{inf}^{CR} at full power conditions with 45% void for the 9x9 MOX- and U-FA

The upper part of Fig. 6 shows the four-bundle cell which is typical for the conventional scatter loading scheme and which contains 1 MOX-FA (of highest reactivity corresponding to about 8 MWd/kg burn-up) and 3 uranium-FAs with burn-ups of 0, 16 and 24 MWd/kg respectively. In the lower part of Fig. 6 the results for the FA-power distribution are given as calculated by CASMO and COSIMA.

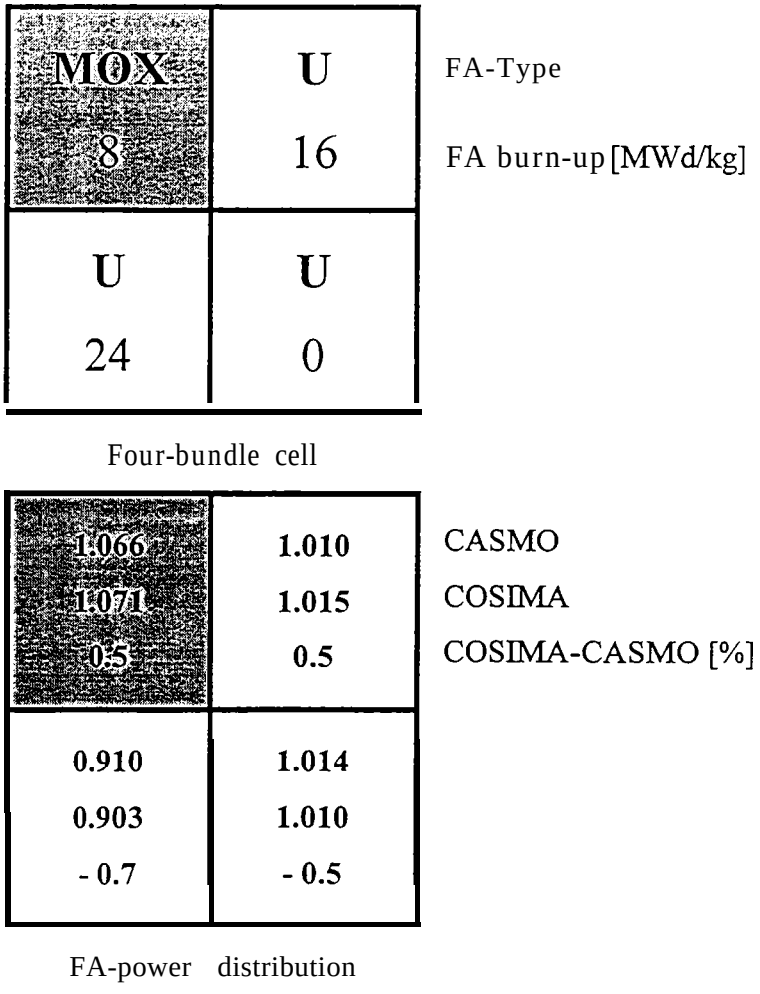

Fig. 6 Four-bundle cell with periodic boundary conditions (100 % reactor power, 45 % void) and corresponding FA-power distribution as calculated by CASMO and COSIMA

From these results it is concluded, that the FA-power distribution as calculated by COSIMA is in good agreement with the CASMO results if COSIMA uses for the MOX-FA the four-bundle data base. On the other hand, COSIMA underestimates the MOX-FA power by about 1 to 2 percent when the data base from CASMO one-bundle calculations is used.

But of more interest than theoretical studies is our incore-experience with MOX-fuel in the high power density BWR cores:
The differences in reactivity between measurement and precalculations in zero-power-tests regarding cores containing MOX-FAs and pure uranium cores are investigated. The analysis according to Table 1 shows that the shutdown margin in cores containing MOX-FAs is precalculated with the same precision as in pure uranium cores.

Table 1: Comparison of precalculated and measured stuck rod shut-down reactivities for beginning of cycle

Reactor cycle No.	No. of MOX-FAs in the reactor core	Stuck rod shut-down reactivity [% Δk_{eff}]	
		Precalculated	Measured
6	-	-2.6	-2.5
7	-	-1.8	-1.7
8	-	-2.1	-2.2
9	-	-2.1	-2.2
10	-	-2.4	-2.4
11	-	-2.4	-2.4
12	32	-1.9	-2.2
13	64	-3.0	-2.9
14	96	-2.3	-2.3
15	124	-1.9	-2.0

In addition to the stuck rod shut-down reactivities the control rod worths have to be investigated in the zero-power-tests. Table 2 shows a good agreement of precalculation and measurement for control rod four-bundle cells without and with MOX-FAs.

Table 2: Comparison of precalculated and measured control rod worths for the cold, zero power and Xe-free reactor core

Number of MOX-FAs in the control rod four-bundle cell	Differential control rod worth [% Δk_{eff}/dm]	
	Precalculated	Measured
0	0.10	0.13
	0.36	0.30
	0.43	0.44
	0.44	0.38
1	0.35	0.42
	0.40	0.36
	0.45	0.40
2	0.14	0.14
	0.43	0.46

The differences between results from TLP measurements and precalculations in mixed MOX-uranium cells and in pure uranium cells are investigated as well. The bandwidths of differences between measurements and calculations are almost congruent (see Fig. 7).

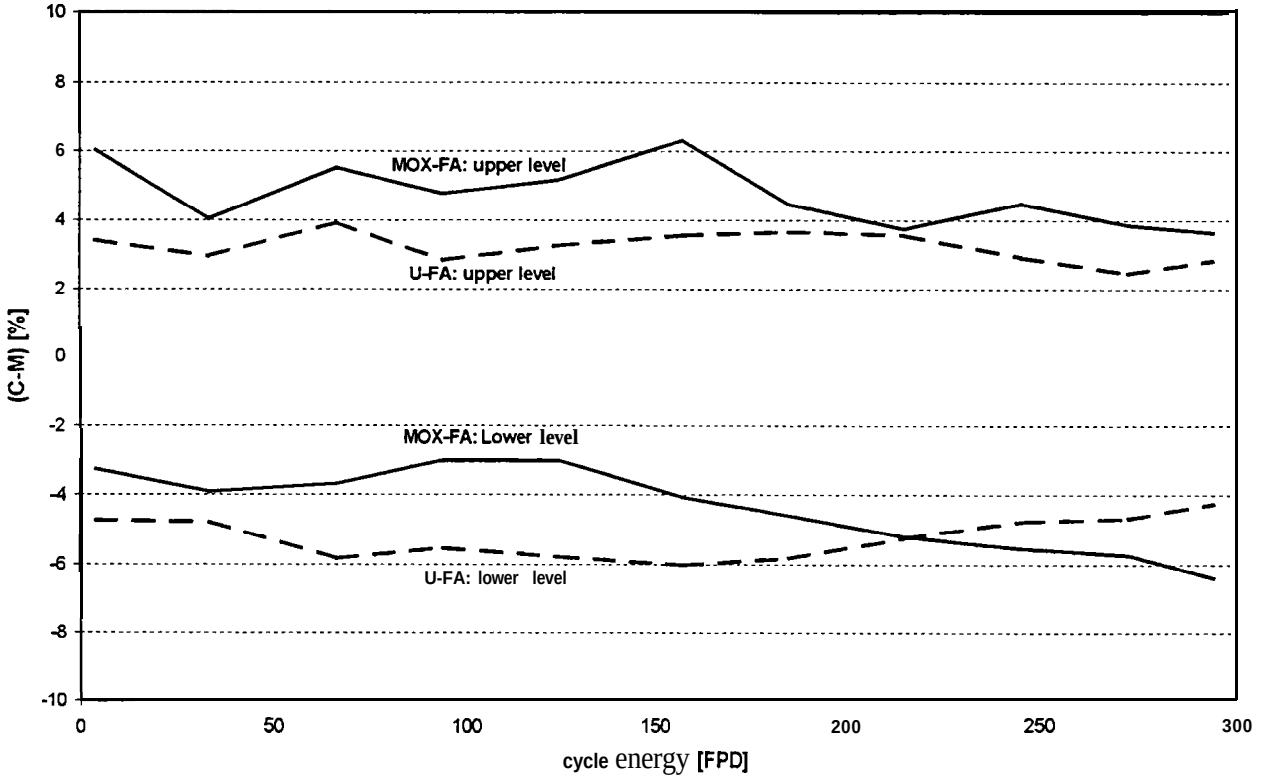

Fig. 7: Bandwidths of differences between TIP-measurements and calculations in Gundremmingen, unit B, cycle 14

All results lead to the conclusion that MOX-FAs can be calculated with the same precision as uranium-FAs. Because of the good prediction of the neutronic **behaviour** of the MOX-FAs no additional core monitoring is required.

Concerning expected radiation dose rates and man-doses during the handling of fresh MOX-FAs a detailed study including a handling procedure to minimize man-doses was performed by NIS (see Table 3).

Table 3: Comparison of measured and precalculated radiation exposure during the handling of MOX-FAs

		Measurement	Precalculation
Contamination	a β/γ	<0.037 Bq/cm² <0.37 Bq/cm²	
Aerosol activity	*a* β	1 Bq/m³ 3.5 Bq/m³	
Dose rate at MOX-FA – Surface	γ n	380 µSv/h 190 µSv/h	530 µSv/h 333 µSv/h
Total man-dose for 8 FAs	γ n	520 µSv 250 µSv estimated*)	
Total man-dose for 16 FAs	**γ + n**	**1540 µSv**	**2800 µSv**

*) based on calculated neutron dose rates and handling time. The official albedo measurements came to 0 µSv (lower detection limit: 200 µSv).

Due to the conservative assumptions with respect to the isotopic plutonium composition and the handling time the man-dose turned out to be only 55 % of the precalculated man-dose. If the official neutron values are taken into account, the total man-dose has been less than 50 % of the precalculated man-dose.

As could be demonstrated, the overall performance of MOX-FAs in the BWRs Gundremmingen is successful until now. The fact, that there is not any defective MOX-FA so far, is of importance. The behaviour of all 9x9 MOX-FAs is according to design The precalculations are in good agreement with results of in-pile measurements.

After the good experience with 140 9x9-MOX-FAs we expect the first 10x10-MOX-FAs to be inserted in Gundremmingen in the year 2000.

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PROGRESS OF FULL MOX CORE DESIGN IN ABWR

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Abstract

Full MOX ABWR core design has been made, based on the MOX design concept of 8x8 bundle configuration with a large central water rod, 40GWd/t maximum bundle exposure, and the compatibility with 9x9 high-burnup UO_2 bundles. Core performance on shutdown margin and thermal margin of the MOX-loaded core is similar to that of UO_2 cores for the range from full UO_2 core to full MOX core. Safety analyses based on its safety parameters and MOX property have shown its conformity to the design criteria in Japan. In order to confirm the applicability of the nuclear design method to full MOX cores, Tank-type Critical Assembly (TCA) experiment data have been analized on criticality, power distribution and β_{eff}/l mesurements.

1. INTRODUCTION

Full MOX ABWR core concept with favorable features for MOX utilization is developed from the perspectives of core performance and safety evaluation.([1],[2]) Its nuclear design method is validated with the criticality analyses of JAERI's TCA experiment by comparing the errors for full MOX cores with those for full UO_2 cores.

2. FULL MOX CORE SCHEME

2.1. Utilization in ABWR

Full MOX ABWR core design has been made with a lattice pitch .wider compared to a conventional BWR (larger H/HM ratio), meaning the mitigation of void coefficient change effect with MOX loading. Its reload core has flexibility to the MOX loading fraction from full UO_2 core to full MOX core, based on the increased performance margin. The main specifications are not changed from those of the standard ABWR, such as 3926MWt thermal power, 872 fuel bundles and 205 control rods.

2.2. Design Concept

The main design concepts of full MOX core are the following, and the basic specifications of core and fuel are shown in Table 1.([4])

(1) The MOX bundle configuration is selected to be the same design as STEP-2 UO_2 bundle (39.5GWd/t discharge exposure) having plenty of operational experience. This bundle has a large water rod at the center of 8x8 fuel rod configuration (as shown in Fig. 1).
- -60 fuel rods consist of 48 of MOX rods and 12 of Gd-containing UO_2 rods.
- -In order to suppress the fuel rod internal pressure, the MOX fuel rod is designed to make the fuel stack length shorter and the plenum volume larger than those of the UO_2 fuel rod.

(2) The MOX bundle exposures are selected to be the same exposures as those of STEP-l UO_2 bundle (33GWd/t discharge exposure and 40GWd/t maximum bundle exposure), which are conservative figures based on the MOX irradiation experience.([5])
- -The maximum pellet exposure is selected to be 58 GWd/t correspondingly.

(3) The bundle average fissile material content is selectedto be about 3wt% of Puf content and about lwt% of U-235 enrichment based on depleted-uranium matrix and Gd containing rods with enriched uranium for the conditions of 13-month cycle length and standard Pu quality of 67wt% (fissile Pu fraction to total Pu).

-Fissile Pu content is restricted to be less than 8wt% (total Pu content (13wt%).

-Pu quality is to be that of reactor grade (Pu quality less than 80wt%).

(4) Full MOX core is planned by EPDC (Electric Power Development Co.Ltd.) to reach gradually from the initial core, loaded with MOX bundles up to about one third of full core, where UO_2 bundles loaded with MOX bundles are 9x9 high-burnup bundles (STEP-3: 45GWd/t discharge exposure).

-The initial MOX fuel has the same Pu fissile content as the reload MOX fuel.

-The design concept of initial core and transition core is basically similar to that of UO_2 multi-enrichment core, where at first the initial lower enrichment UO_2 bundles are replaced with reload ones.

TABLE I. BASIC SPECIFICATION OF CORE AND FUEL DESIGN

Items	Specification		
Core			
Type	Advanced Boiling Water Reactor (ABWR)		
Thermal power (MW)	3,926		
Core flow (t/h)	52.2×10^3		
Core pressure (MPa[abs])	7.17 (73.1kg/cm^{2}a)		
Number of fuel bundles	872		
Number of control rods	205		
Fuel Assembly	MOX Fuel	Reference UO_2 Fuel (STEP-2 8x8)	UO_2 Fuel (STEP-3 9x9)
Lattice Confuguration	8x8	8x8	9x9
Average ^{235}U content *'(w-t%)	1.2	3.5	3.8
Average Puf content *[1](wt%)	2.9		
Maximum burnup (MWd/t)	40,000	50,000	55,000
Number of fuel rods	60	60	74 *[3]
Pellet material	UO_2-PuO_2(MOXrods) UO_2-Gd_2O_3(UO_2rods)	UO_2 UO_2-Gd_2O_3	UO_2 UO_2-Gd_2O_3
Cladding outside diameter (mm)	12.3	12.3	11.2
Cladding wall thickness *[2](mm)	0.86	0.86	0.71
Cladding Material	Zirc-2 (with Zr liner)	Zirc-2 (with Zr liner)	Zirc-2 (with Zr liner)
Number of water rods	1	1	2
Water rod outside diameter (mm)	34.0	34.0	24.9

*[1] For reload fuel

Average ^{235}U content : weight of ^{235}U/(weight of total Pu+weight of total U)

Average Puf content : weight of fissile Pu/(weight of total Pu+weight of total U)

*[2] Including Zirconium liner thickness of about 0.1 mm

*[3] Including 8 partial length rods

3. CORE PERFORMANCE AND SAFETY EVALUATION

3.1. Fuel and Core Design

Rod layout of the MOX fuel bundle is shown in Fig. 1, where four kinds of Pu contents are applied for Pu content of each MOX fuel rod. The neutron multiplication factor (k-infinity) is shown in Fig.2 in comparison with 9x9 UO_2 fuel. MOX fuel has a remarkable feature that k-infinity variation

with exposure is small compared to UO_2 fuel. This trend makes a favorable property of MOX fuel to reduce the bundle power peaking.

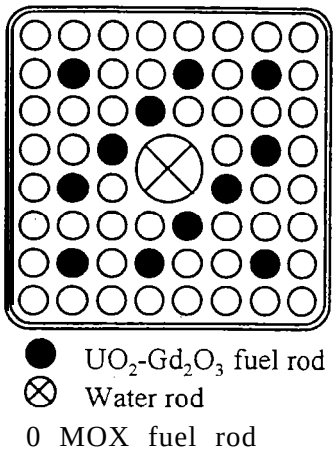

FIG. 1. Rod layout of MOX fuel bundle.

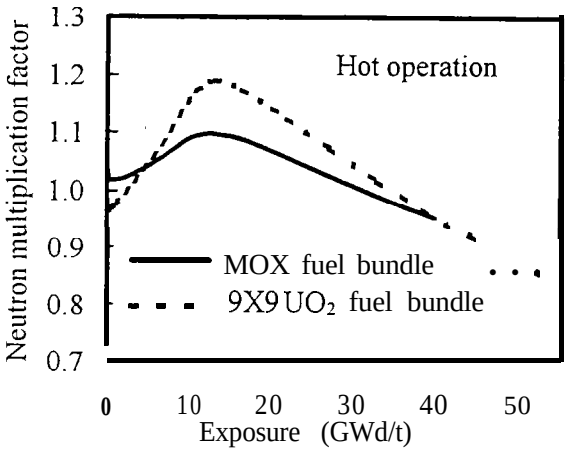

FIG.2. Neutron multiplication factor vs. exposure.

Fission power fractions of main isotopes are shown for the 9x9 UO_2 and MOX fuel bundles in Fig.3. In the 9x9 UO_2 fue bundle, fission power of Pu isotopes accounts for about 60% of the total power at the stage of discharge. Fission power fractions of main isotopes are also shown for the equilibrium cycle from full UO_2 core to full MOX core in Fig.4. .The significant contribution of Pu isotopes to fission power for UO_2 fuel bundle and for UO_2 core means that with excellent operating experiences for UO_2 core, the treatments of Pu in the nuclear calculation are adequate.

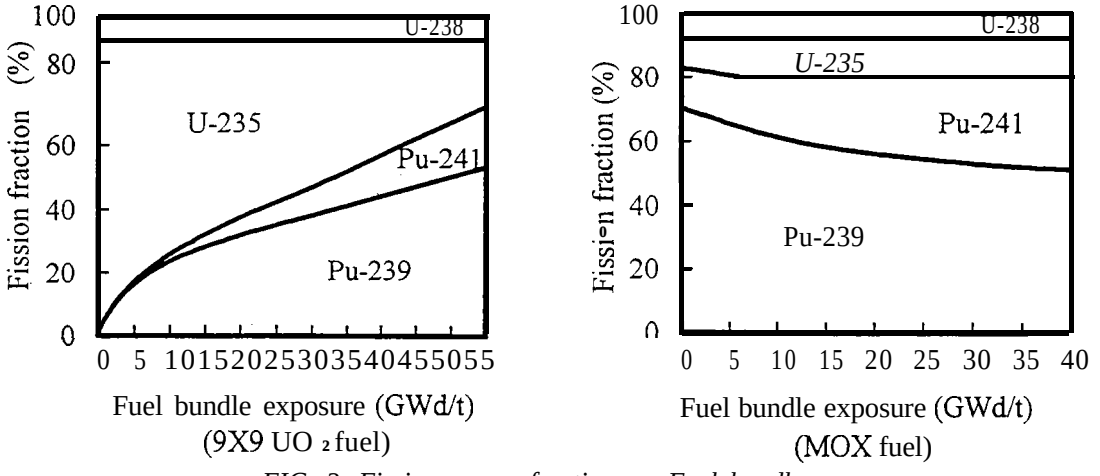

FIG. 3. Fission power fraction vs. Fuel bundle exposure.

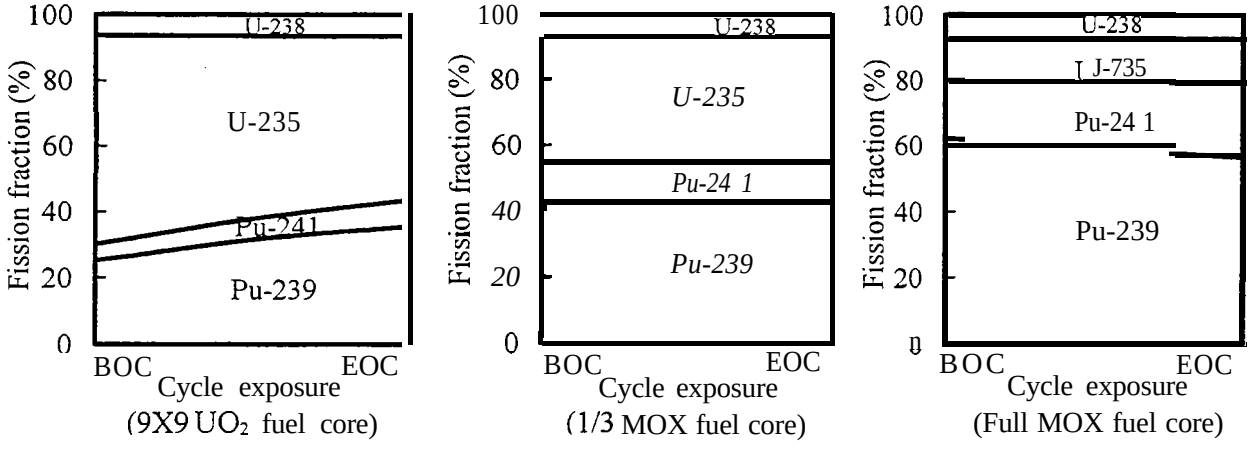

FIG.4. Fission power-fraction vs.Cycle exposure.

Although MOX fuel tends to become higher pellet temperature owing to lower thermal conductivity and increased FP gas release compared with- UO_2 fuel, the pellet center temperature remains sufficiently lower against fuel melting temperature through the lifetime. Also, though MOX fuel tends to become higher rod internal pressure owing to increased FP gas and He gas release, the rod internal pressure remains still eqivalent to UO_2 fuel rod at the end of lifetime due to increased gas plenum for MOX rod. The rod internal pressure of the MOX fuel is shown in comparison with 9x9 UO_2 fuel in Fig.5.

Core performance on shutdown margin and thermal margin of the MOX-loaded core is similar to that of UO_2 cores for the range from full UO_2 core to full MOX core. Shutdown margin, Maximum Linear Heat Generation Rate (MLHGR), and Minimum Critical Power Ratio (MCPR) for the equilibrium cycle are shown in the comparison among full UO_2 core, 1/3 MOX core and full MOX core in Fig.6. Noticeably, MCPR is higher in the full MOX core owing to its lower radial power peaking, compensating for the ascending of MCPR operating limit (OLMCPR) due to void coefficient increase.

Safety parameters,i.e. reactivity coefficients (void coefficient, Doppler coefficient) and dynamic parameters (delayed neutron fraction, prompt neutron lifetime) change continuously depending on the MOX loading fraction. Those parameters are shown to be dependent on MOX loading fraction in Fig.7. While the void coefficient of the full MOX core is about 20% larger in absolute value than the full UO_2 core, the delayed neutron fraction of the full MOX core is about 20% smaller compared to the full UO_2 core.

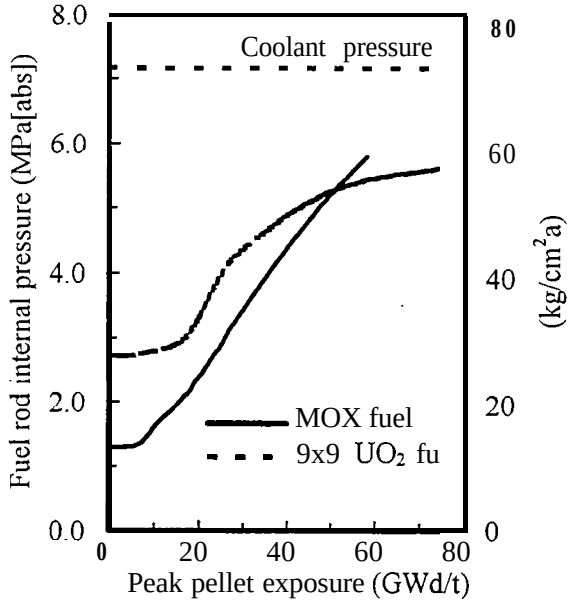

FIG. 5. Fuel rod internal pressure vs. pellet exposure

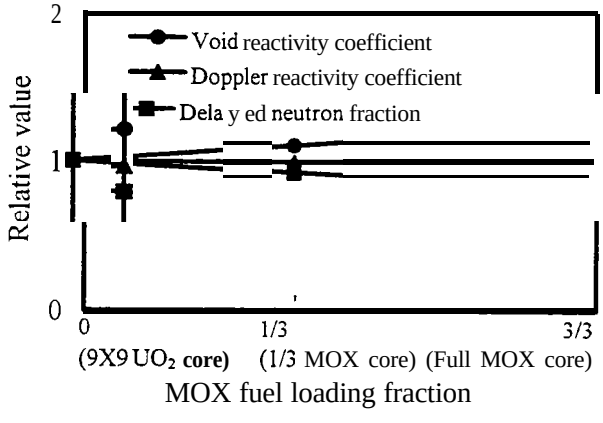

FIG. 7. Safety *parameters vs. MOX ioading fraction*

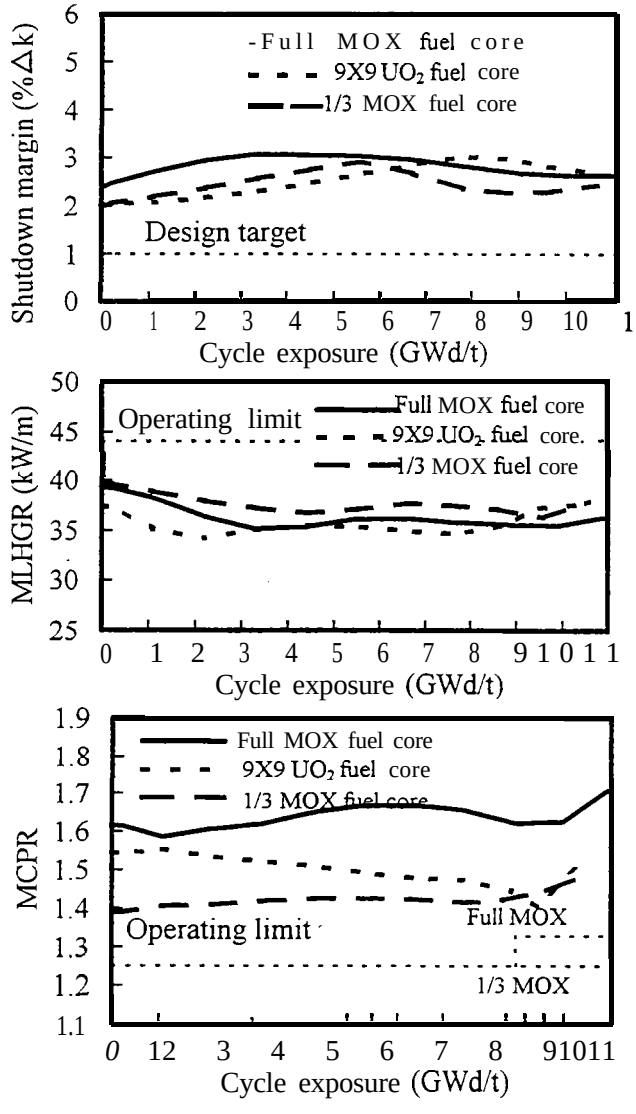

FIG. 6. Comparison of equilibrium core performance

3.2. Safety Evaluation

Based on the above safety parameters and MOX property, safety analyses have been performed on stability, abnormal transient during plant operation, transient and accident for control rod system, and accident, showing the conformity to the design criteria in Japan.

(l)Stability

Although "core stability" and "regional stability" are affected by the void coefficient increase in absolute value, their decay ratios are about 0.7 and about 0.6 respectively, at the minimum pump speed-maximum reactor power condition of the full MOX core, satisfying the decay ratio criterion of 1.0.

(2)Abnormal Transient during Plant Operation

The increase of void coefficient in absolute value has a suppressing effect on void increasing events like "partial loss of coolant flow". However, the effect increases the power response to void decreasing events like "generator load rejection without turbine bypass", resulting in the increase of AMCPR (index of thermal margin change). AMCPR is shown as a function of MOX loading fraction in Fig.8, where the limiting event changes from "loss of feed water heating" to "generator load rejection without turbine bypass" as MOX loading fraction increases. As this result depends on the control rod scram reactivity curve, the relevant design curves are to be selected for the definite duration of cycle period. AMCPR and reactor pressure responses in "generator load rejection without turbine bypass" are shown in Fig.9 as transient analysis result.

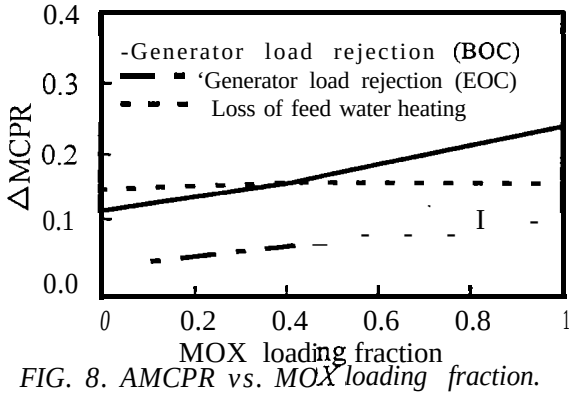

FIG. 8. AMCPR vs. MOX loading fraction.

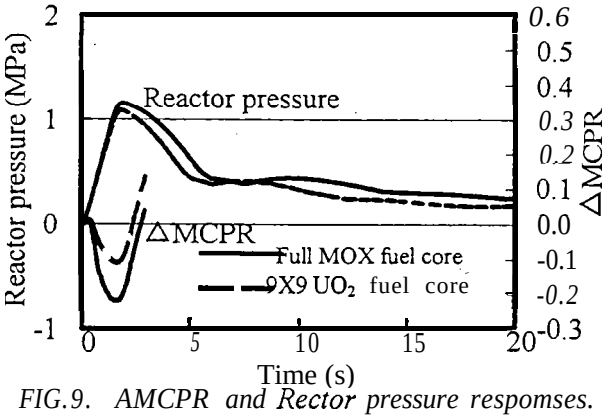

FIG.9. AMCPR and Rector pressure respomses.

(3)Transient and Accident for Control Rod System

The fuel enthalpy response of "Control Rod Drop Accident (CRDA)" is shown in Fig. 10 as a representative event of transient and accident for control rod system, where the dropping control rod worth is assumed to be 1.3%Δk. The maximum fuel enthalpy in the full MOX core is almost equivalent to that of the UO_2 core, although the response depends on the detailed design such as power distribution in the core.

(4)Accident

"Loss of Coolant Accident (LOCA)" being an event where the void fraction increases, the MOX core with negative void

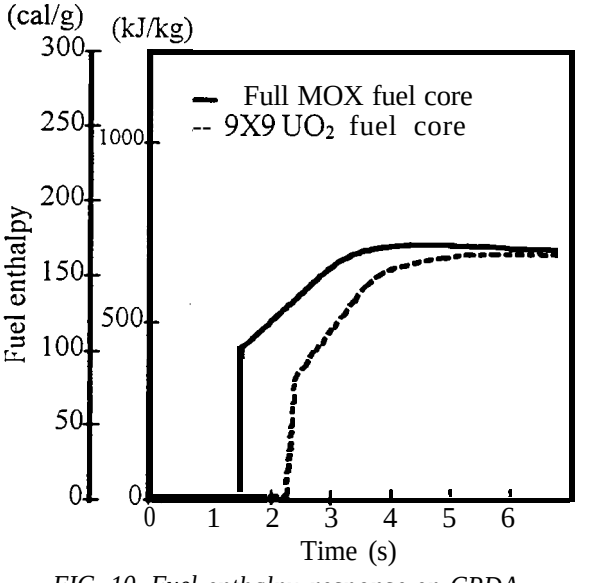

FIG. 10. Fuel enthalpy response on CRDA.

coefficient larger than UO_2 core in absolute value tends to, reduce the reactor power further at the LOCA. Therefore, it is conservative for the MOX core evaluation to adopt time dependent reactor power identical to the UO_2 core, while using material properties of the MOX. The fuel cladding temperature of LOCA is evaluated for High Pressure Core Flooder system (I-IPCF) pipe break" as the severest case for the drop of reactor water level. The cladding surface temperature rises due to boiling transition, but the core is always covered with two phase coolant mixture over the whole transient period. The peak cladding temperature (PCT) is about 600°C, satisfying the criterion of 1200°C, although PCT of MOX core becomes a little higher than UO_2 core.

4. VALIDATION OF FULL MOX NUCLEAR DESIGN

Besides ample operational experience where Pu builds up to about 60% fission fraction at the time of discharge, the BWR nuclear design method has been verified for MOX configuration through MOX mock-up criticality experiment (partially loading of MOX bundles at VENUS facility)[3] and lead-use basis utilization (irradiation of two MOX bundles at Tsuruga Unit-1)([3],[6]). In order to confirm the applicability of the nuclear design method. to full MOX cores, Tank-type Critical Assembly (TCA) experiment data is employed and analyzed on criticality and power distribution mesurements from the uniform MOX rod arrays (48 cases) and the uniform UO_2 rod arrays (40 cases), including different H/HM ratios. And also the β_{eff}/l (effective delayed neutron fraction to prompt neutron lifetime) ratios measured with pulse neutron technique are compared for the uniform MOX rod arrays (4 cases) and the uniform UO_2 rod arrays (4 cases).

4.1. TCA experiments

Critical experiments for full MOX and full UO_2 cores were conducted on TCA of the Japan Atomic Energy Research Institute (JAERI), where Pu content in the MOX fuel is 3.0wt% and ^{235}U enrichment in the UO_2 fuel is 2.6wt%. The moderator is light water and moderator-to-fuel volume ratios range from 1.50 to 3.00 for UO_2 cores and from 2.42 to 5.55 for MOX cores.([7],[8])

The vertical cross-sectional view of TCA is shown in Fig. 11. Criticality is attained by adjusting the water level. The core configuration of TCA experiment is shown in Fig. 12, where MOX and UO_2 fuel rods are arranged in a rectangular shape. The critical sizes of MOX and UO_2 cores are measured by changing the moderator-to-fuel ratios and the number of fuel rods in a column and row. Table II shows the specifications of MOX fuel rod and UO_2 fuel rod.

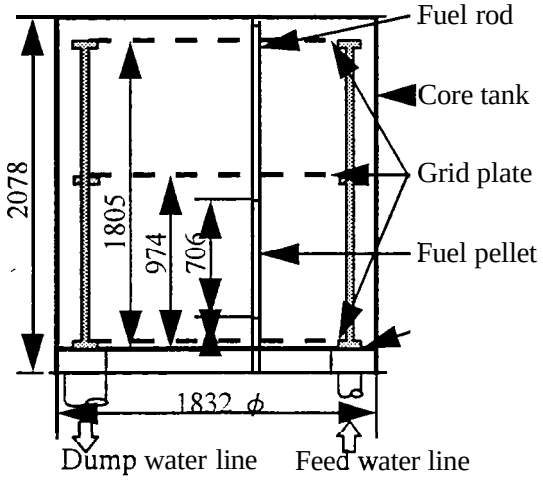

FIG. 11. Vertical cross-sectional view of TCA.

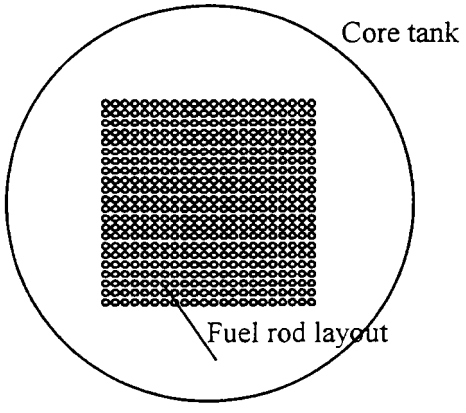

FIG. 12. Core configuration of TCA experiment.

TABLE II. FUEL SPECIFICATIONS

Items				MOX	uoz
Fuel	Enrichment (wt%)			3.01 *1	2.596
	Isotope ratio (wt%)	Uranium	U-235	0.72	2.596
			U-238	99.27	97.404
		Plutonium	Pu-238	0.494 *2	–
			Pu-239	6X.18 *2	–
			Pu-240	22.02 • ?	–
			Pu-241	7.26 *2	–
			Pu-242	2.04 *2	–
		Americium	Am-24 1	530ppm *3	–
	Pellet	Diameter(mm)		10.65	12.50
		Density(g/cm^3)		6.056	10.40
		Effective fuel length(mm)		706	1441.5
Cladding	Material			Zircaloy-2	Al
	Inner diameter			10.83	12.65
	Thickness (mm)			0.70	0.76

*1 : $PuO_2/(PuO_2+UO_2)$, Date of assaying : *2 1971/8/19, *3 1971/8/16 (The variation of composition with time was taken into account in the analysis.)

4.2. **Analysis and Evaluation**

BWR design-purpose codes, HINES (two-dimensional fuel assembly nuclear calculation code) and CERES (two-dimensional core calculation code) are employed. The former code is made up of a fast energy part (10MeV-0.625eV) of the neutron slowing down equation with a GAM-type 68-energy group neutron cross section library and a thermal energy part (below 0.625eV) of the transport equation with a THERMOS-type 30-energy group neutron cross section library mainly from ENDF/B files, to obtain three group macroscopic cross sections for the subsequent use in the diffusion-basis core calculation. In the TCA experiment analysis, the group constants are produced from unit fuel cell calculations. The axial bucklings required in two-dimensional diffusion calculation are obtained from the critical core heights and the measured reflector savings.

The evaluated results show the eqivalent accuracy between full MOX cores and full UO_2 cores as follows.

(1) For criticality, the histgrams of neutron multiplication factor are shown for full MOX cores (48 cases) and full UO_2 cores (40 cases) in Fig. 13. Full MOX analyses have the almost identical evaluation results as UO_2 analyses within the same standard deviation (0.2%Δk) in both cases.

(2) For power distribution, the comparisons between calculation and measurement of relative fuel rod power are shown for full MOX cores and full UO_2 cores in Fig.14, and the power distributions in the horizontal direction are shown in Fig. 15. Full MOX analyses have the almost same deviation (1% $\sim$ 2%: root mean square of the difference between calculation and measurement) as UO_2 analyses.

(3) For β_{eff}/l ratio, the comparisons between calculation and measurement of effective delayed neutron fraction to prompt neutron lifetime are shown in Fig. 16. Full MOX analyses have the almost same deviation (below 4%: difference between calculation and measurement) as UO_2 analyses. In this evaluation, four group macroscopic cross sections are provided for the subsequent diffusion core calculation to take account of fission neutron spectrum difference between prompt neutron and,delayed neutron.

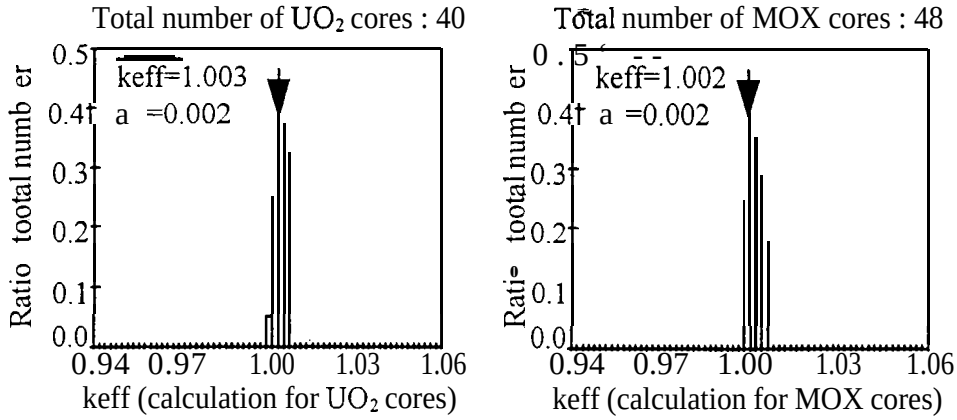

FIG. 13. Histgram of neutron multiplication factor.

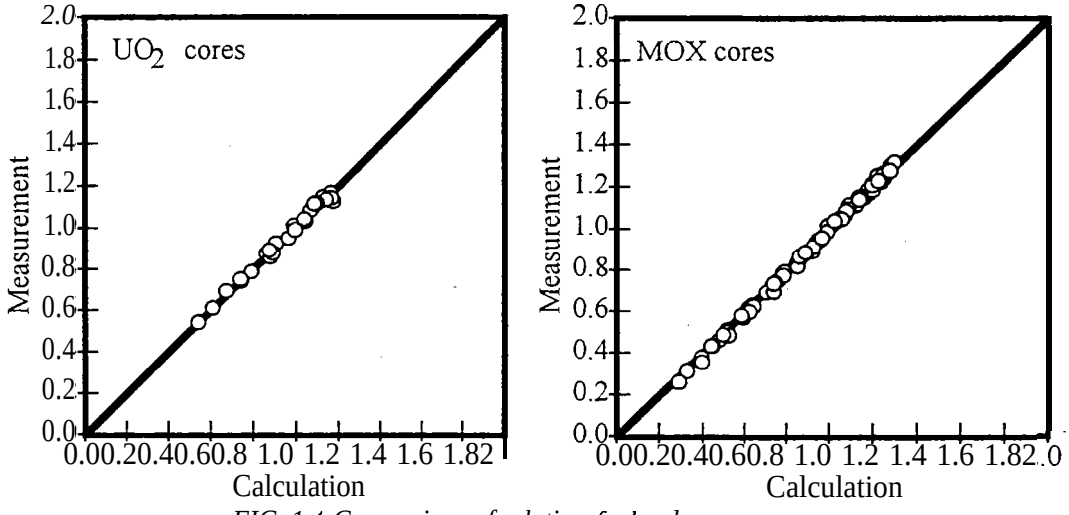

FIG. 1 4: Comparison of relative fuel rod power.

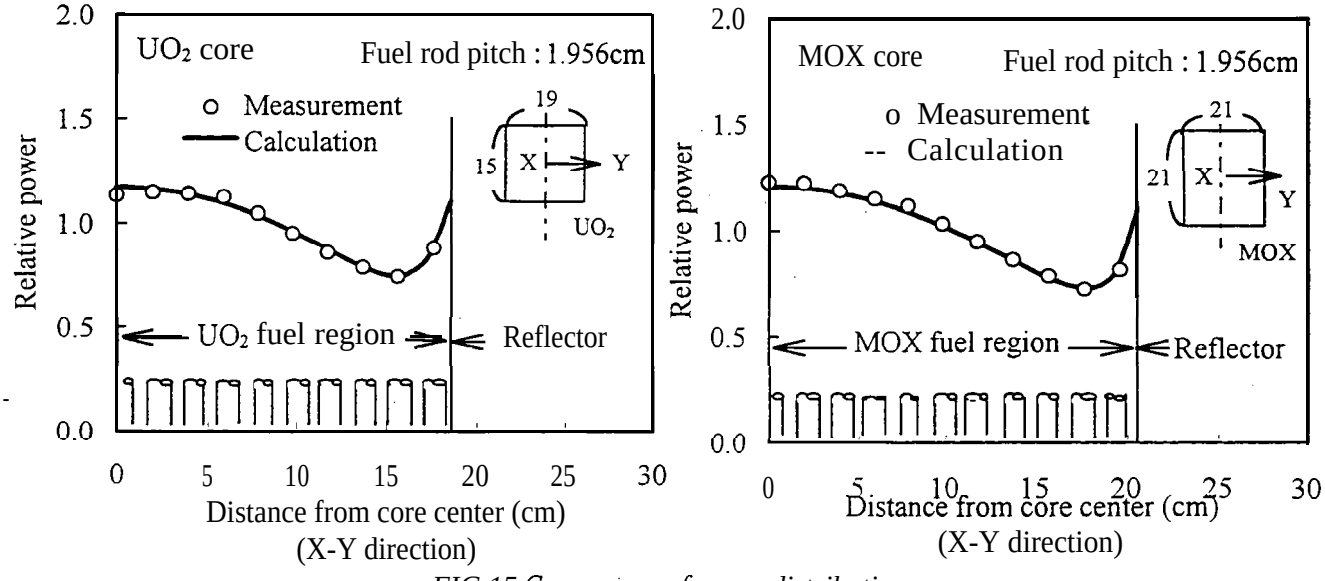

FIG.15 Comparison of power distribution

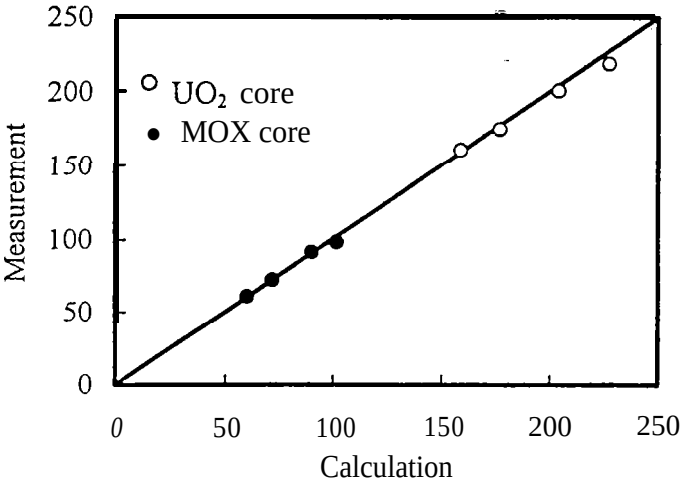

FIG. 16. Comparison of β_{eff}/l.

5. CONCLUSION

(1) Full MOX ABWR core design has been made, based on the MOX design concept of 8x8 bundle configuration with a large central water rod, 40GWd/t maximum bundle exposure, and the compatibility with 9x9 high-bumup UO_2 bundles. Utilization in ABWR with a lattice pitch wider compared to a conventional BWR (larger H/HM ratio) means the mitigation of void coefficient change effect with MOX loading.

(2) Besides fuel mechanical characteristics, core performance on shutdown margin and thermal margin of the MOX-loaded core is similar to that of UO_2 cores for the range from full UO_2 core to full MOX core. Safety analyses based on its safety parameters and MOX property have shown its conformity to the design criteria in Japan.

(3) In order to confirm the applicability of the nuclear design method to full MOX cores, Tank-type Critical Assembly (TCA) experiment data have been analized on criticality, power distribution and β_{eff}/l mesurements. The evaluated results show almost the same accuracy for full MOX and full UO_2 cores.

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EFFECT OF PLUTONIUM VECTOR ON CORE WIDE NUCLEAR DESIGN PARAMETERS

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Abstract

The technicalities of MOX usage from a nuclear design perspective depend to a considerable extent on the isotopic quality of the plutonium (Pu) used. This paper explores the effectiveness of the Lifetime Average Reactivity (LAR) approach to matching varying fissile qualities in order to meet given cycle energy requirements. The paper also investigates the impact of these different fissile qualities (and hence Pu sources) on the key core wide nuclear design parameters for an equilibrium PWR fuel cycle. The study concludes by comparing the effect of both 30% and 45% MOX core fraction on the key core wide nuclear design parameters.

1. INTRODUCTION

1.1 General

MOX fuel usage is well established in commercial reactors in several European countries and is now being introduced into Japanese reactors as well as being considered for irradiation in LWRs in the United States of America and Russia as part of the weapons disposition programme. These different markets mean that there is a variety of sources of Pu ranging from the gas cooled reactors in the United Kingdom (Magnox and Advanced Gas Cooled Reactors (AGRs)), through the more familiar LWR source of Pu from Europe and Asia, to the more recent investigation into the use of weapons grade Pu in US LWRs. These different sources of Pu mean a variety of fissile contents and hence a need for different Pu concentrations. In turn this results in different impacts on core wide nuclear design parameters.

In supplying MOX fuel assemblies for a reactor, it is necessary to specify the fissile content of the MOX assemblies. For UO_2 assemblies, this task is straightforward as the fissile content simply depends on the ^{235}U enrichment. However, for a MOX assembly the task is more complicated because the fissile content depends on both the concentration of the PuO_2 in the UO_2 matrix and on the Pu isotopic composition (Pu vector). As has been mentioned, the Pu vector can vary to a considerable extent depending on the type and burnup of the fuel used as the source of the Pu.

In order to match these varying Pu compositions to each other or to UO_2 fuel, one of the fundamental parameters of fuel for any reactor type is the reactivity averaged over its lifetime in the reactor, or Lifetime Average Reactivity (LAR). The most rigorous approach to ensure total matching would be to complete a whole core equivalence assessment, but this can prove time consuming, particularly when there are many Pu compositions to consider. The use of LARs using assembly (lattice) calculations rather than an explicit whole core assessment is often preferred as it simplifies this matching approach. This saves the designer time and effort and therefore proves an ideal scoping tool and first order approximation to the whole core situation.

This paper investigates the approach and effectiveness of matching LARs at the lattice level and the limitations of such a method. The paper then goes on to address the whole core nuclear design issues of different quality plutonium using the Studsvik Scandpower nuclear design suite, CASMO-SIMULATE [l]. The range of Pu isotopic compositions (Pu vectors) assessed can be seen in Table I.

Furthermore, with MOX fuel experience growing around the world, higher MOX contents and core fractions are likely in the near future. Therefore, the report also considers the whole core effects of using a MOX core fraction of 30% and 45%.

TABLE I. Plutonium Vectors from a Range of Pu Sources

	Pu238 w/o	Pu239 w/o	Pu240 w/o	Pu241 W/o	Pu242 w/o	Fissile w/o
Case A	2.46	54.69	26.16	9.51	7.18	64.20
Case B	1.08	63.24	25.30	6.30	4.09	69.54
Case C	0.60	61.64	30.31	4.53	2.92	66.17
Case D	0.10	70.00	24.90	3.90	1.10	73.90
Case E	0.00	95.00	4.00	1.00	0.00	96.00

NB. Each vector is shown 'as separated' – i.e. does not include the 2 years ageing assumed in the calculations

1.2 **Physics of MOX Fuel**

A plutonium-uranium mixed oxide (MOX) assembly for a thermal reactor generally looks exactly the same mechanically as a conventional uranium dioxide (UO_2) assembly. But whereas a UO_2 assembly obtains the bulk of its energy from fissions in ^{235}U, a MOX assembly obtains most of its energy from plutonium fissions.

In thermal MOX fuel, only the odd isotopes ^{239}Pu and ^{241}Pu are fissile; the even isotopes do not fission to a significant extent and behave as neutron absorbers. ^{240}Pu is different in that neutron captures lead to ^{241}Pu generation so that it can also be considered a fertile nuclide. Both ^{239}Pu and ^{241}Pu have considerably higher fission cross-sections than ^{235}U, but this does not necessarily translate into higher reactivities because they also absorb thermal neutrons more strongly.

The neutron spectrum in a MOX assembly is very different to that in a UO_2 assembly for a comparable reactivity. For a typical Pu isotopic composition (Pu vector), the total Pu enrichment needs to exceed the ^{235}U enrichment of the equivalent UO_2 assembly. The total thermal absorption cross section is therefore higher not just because there are more absorbing atoms, but also because the absorption cross sections of the odd Pu isotopes exceed that of ^{235}U. In the most important energy region for fissions (up to about 0.6 eV) the absorption cross section for a MOX assembly is approximately two or three times larger than that of the equivalent UO_2.

Since there is only a modest difference in the number of neutrons released per fission between U and Pu, the neutron source in the energy range typical of fission neutrons (0.1 to 10 MeV) just depends on the power density and is the same for MOX and UO_2 to a first approximation. The fission neutrons are moderated until they reach thermal energies in much the same way in both assembly types. When in the thermal range, the larger thermal absorption cross section in a MOX assembly causes the thermal flux to be much lower than in the equivalent UO_2 assembly. Therefore, whereas the fast flux (normally taken to be everything above 0.6 eV) is the same in the two assembly types, the thermal flux is much lower in MOX. The fast to thermal flux ratio is therefore much higher in MOX and the spectrum is therefore 'harder'.

Normally, the reactivity versus burnup characteristics of MOX and UO_2 assemblies are very different; the gradient of reactivity with burnup is considerably smaller in MOX than in UO_2, partly because of the different neutron spectra and partly because of the fertile captures in ^{240}Pu (this tends to hold down the initial reactivity through neutron captures but eventually contributes to the overall reactivity through the generation of fresh ^{241}Pu). A typical example of this difference can be seen in Figure 1. Consequently, it is not possible to match the reactivity of MOX and UO_2 assemblies at all points in time; it is only possible to match the reactivity over the lifetime in the reactor. In practice, this does not limit the performance of MOX assemblies since acceptable fuel loading patterns can be found by matching the Lifetime Average Reactivities (LARs).

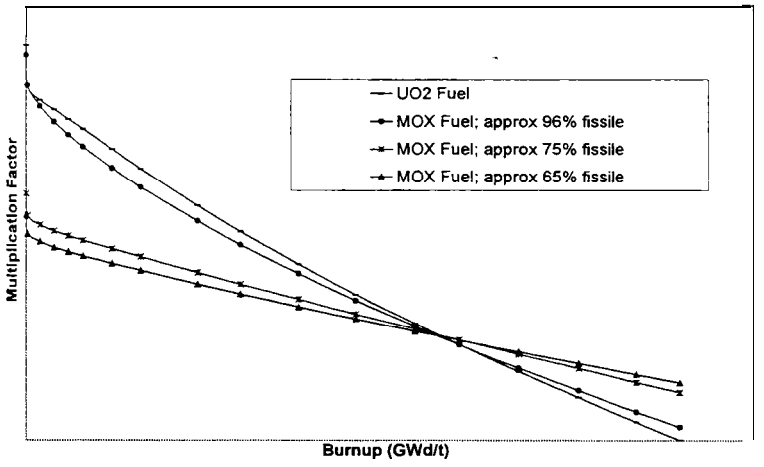

FIG. 1. Multiplication Factor Versus Burnup for Different Pu Qualities

2. METHODS USED

2.1 Matching Lifetime Average Reactivity

For a reactor in which the core is divided into n refuelling batches, one simple approach to determining the LAR is to average the multiplication factor k_∞ of the fuel at the end of its first, second, third and up to its nth dwell (where 'dwell' is the number of cycles of irradiation). Figure 2 illustrates this schematically for a typical unpoisoned UO_2 and MOX assembly in a reactor with a 3 batch loading scheme (i.e. $n=3$) and as used in this study. For such a loading scheme, the core will consist of 1/3 fresh assemblies, 1/3 once burnt and 1/3 twice burnt assemblies. Points (i), (ii) and (iii) on Figure 2 correspond to the condition of the fuel at the end of its first, second and third dwells respectively, and if the reactor is in equilibrium, all of the fuel in the core of this batch type will have the same k_∞ versus burnup curve. The LAR of a given batch is therefore the average of the k_∞'s at points (i), (ii) and (iii). The two fuels shown in Figure 2 have the same LAR.

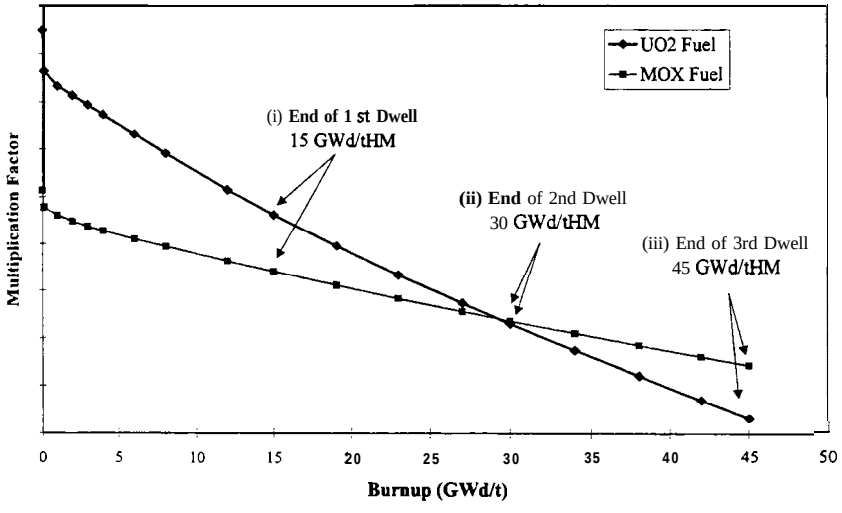

FIG. 2. Calculation of Lifetime Average Reactivity for MOX and UO_2 Fuels

For an idealised reactor with no neutron leakage, the end of the cycle coincides with an LAR of 1.0, meaning that at the end of the cycle, in the absence of control rods, burnable or soluble neutron absorbers, the core is just capable of sustaining the chain reaction. In practice, there is always some neutron leakage so the LAR must be designed to be greater than one, typically in the range 1.03 to 1.04. It is essentially this requirement that determines the enrichment of fuel for either the UO_2 or MOX fuel.

In thermal reactors it is usual to want to match the LAR of a MOX assembly to that of a UO_2 assembly co-resident in the core. However, several points -must be borne in mind when comparing LARs of UO_2 and MOX assemblies.

Firstly, as illustrated above, the burnup characteristics of a MOX assembly are very different from those of a UO_2 assembly with the same LAR. This is particularly important when MOX fuel is first introduced into the core because if the fuel is matched to the value of LAR for an equilibrium cycle, then there will be a likely shortfall in cycle length during the transition cycles due to the lower k_∞ at the beginning of the MOX irradiation (see Figure 2 above).

Secondly, the concept of LAR is only meaningful if the cycle length and number of batches in the loading pattern are also given. It is clear that UO_2 and MOX assemblies may have equivalent LARs for a particular combination of cycle length and batch number, but they will not generally be equivalent for any other combination. The same is true for two MOX assemblies with different Pu concentrations and Pu vectors, but crucially to a much lesser extent, primarily due to the similarity between k_∞ characteristics of MOX fuel.

Thirdly, it has been assumed in the preceding discussion that the LAR is obtained from a simple averaging of the k_∞ values at the end of each dwell. This is a reasonable approximation, but to be fully rigorous, the LAR should be determined from a weighted average of the k_∞'s, with weighting factors to account for the contribution of each assembly to the overall core power. This is more important for the UO_2 fuel than the MOX since the k_∞ value is much more dependent on burnup. In practice, uniform power weightings are a good approximation.

Finally, it should be remembered that this approach matches only the **reactivity** of the fuel; it does NOT make any attempt to match the power produced by each assembly as well. This becomes an important issue as far as peaking factors and overall assembly burnups are concerned.

2.2 **Determination of MOX Contents and Fuel Management**

In order to investigate the effectiveness of the LAR matching approach, the initial task was to match the LAR of each of the different MOX fuel types in Table I (cases A to E) to that of the UO_2 fuel (4.00 w/o ^{235}U) by adjusting the Pu content until the LAR was equivalent within ± 0.15% reactivity (150 pcm) which equates to approximately 3 to 5 effective full power days (EFPD). This limit would typically be set in conjunction with the utility. The Studsvik Scandpower lattice code CASMO-4 [2] was used to calculate the reactivities of each fuel type and hence determine the Pu concentrations.

Once the LARs had been matched and the Pu contents determined, the resulting compositions were then used in a single assembly model (again using CASMO-4) to generate the cross sections used in the whole core assessment using SIMULATE-3 [3]. The CASMO-4 models generated a full data library including all feedback effects (Doppler, moderator etc) for each lattice design. The MOX assemblies consisted of three enrichment zones (high, medium and low) to control within-assembly power peaking.

A standard 3-loop Westinghouse PWR was examined for this study. The general parameters were:-

Assemblies	157 assemblies ; 17x17 pins
Assembiy pitch	21.50 cm
Rod Diameter (Nominal)	0.475 cm
Required cycle length	15.50 +/- 0.10 GWd/tHM
Initial UO_2 Core Enrichments (cycle 1)	2.00, 2.50 and 3.20 weight percent ^{235}U [4]
Target $F_{\Delta H}$	I1.60
Loading strategy	3 batch; 52 feed assemblies (16 'LOW' & 36 'HIGH'); out-in-in

The Studsvik Scandpower graphics-based multi-cycle design tool, X-IMAGE[5] was used to generate the equilibrium cycles and subsequently improve the loading pattern. The initial scoping loading pattern can be seen in Figure 3.

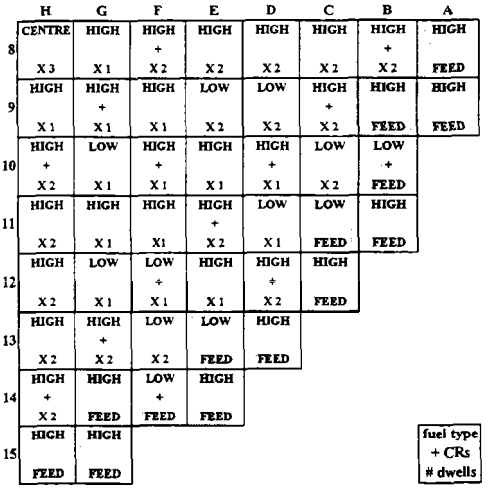

FIG. 3. Basic Equilibrium Loading Pattern

A UO_2 equilibrium cycle was examined initially to determine a base case for both cycle length and key nuclear design parameters. For cycle 2 onwards, a split batch of 4.00 ('LOW') and 4.50 ('HIGH') w/o ^{235}U was used to achieve the required cycle length and reduce within assembly peaking. Various key safety parameters were then calculated using the whole core code SIMULATE-3, including peak assembly integrated two dimensional pin power ($F_{\Delta H}$), boron coefficient, Moderator Temperature Coefficient (MTC), control rod worth, and delayed neutron fraction. Although no actual limits were placed on any of the safety parameters, a realistic loading pattern was determined to make the study as applicable as possible (e.g. target $F_{\Delta H}$, assembly burnups).

The initial all-UO_2 cycle 1 loading pattern was also used as the beginning of a transition to the MOX equilibrium cycle. The MOX assemblies were introduced in subsequent, transition cycles until one-third of the core consisted of MOX assemblies and the core had reached equilibrium; 16 MOX and 36 UO_2 feed assemblies were used i.e. equivalent loading to the split batch all-UO_2 core with the MOX assemblies replacing the 'LOW' UO_2 assemblies in *number, location and reactivity.* This MOX loading (approximately one-third MOX core fraction) was chosen as a typical loading for a current PWR design based on current safety limits. It should be noted that the all-UO_2 and the MOX-UO_2 cases used IDENTICAL loading patterns, with only a few minor alterations to reduce peaking i.e. all batch types (fresh, once burnt and twice burnt assemblies) were kept in the same locations (as shown in Figure 3) and only *within* batch shuffles were completed. This ensures that the loading pattern type is kept constant and there is little effect on neutron leakage.

The initial MOX loading pattern was produced using a Pu vector that was 'typical' in terms of the reactivity versus burnup characteristics; Case A was chosen arbitrarily. Once a reasonable, realistic loading pattern had been determined using the 'typical' Pu vector, this loading pattern was then maintained for all subsequent MOX assessments to eliminate any loading pattern specific issues (e.g. highly burnt MOX assemblies in control rod locations would erode the shutdown margin, change of location of feed assemblies would alter the core leakage). Furthermore, by keeping the loading pattern identical for each Pu vector, it aiiows an assessment of the extent to which the assembiy powers are affected, given that the LARs had been matched.

SIMULATE-3 was then used to determine the values for the key safety parameters ($F_{\Delta H}$, boron coefficient, MTC, control rod worth and delayed neutron fraction) for each of the equilibrium cycles with the varying plutonium qualities.

The approach was then repeated for a higher MOX core fraction; 24 MOX and 28 UO_2 assemblies in the feed fuel, resulting in a MOX core fraction of approximately 45%. Again, the all-UO_2 equilibrium cycle was used for comparison.

3. RESULTS

3.1 Investigation into Matching Lifetime Average Reactivity

The first stage of the investigation was to determine the effectiveness of matching the LARs of MOX and UO_2 assemblies at the lattice level. This was done by matching the LAR of the MOX fuel to that of the 4.00 ^{235}U ('LOW') by varying the Pu content. This was initially done for a PWR Pu vector Case A in Table I). In order to match the LAR of the 'LOW' UO_2 fuel, approximately 7.7 w/o Pu_{tot} was required. When this fuel was loaded into the whole core model and depleted to equilibrium, the resulting cycle length for the MOX-UO_2 core was only 0.06 GWd/tHM lower in cycle length than the all UO_2 core. This suggested that matching the LAR to that of the UO_2 at the lattice level had worked successfully. However, by closer examination of the loading pattern shown in Figure 3, it can be seen that the MOX feed assemblies (i.e. 'LOW' feed) and several burnt MOX assemblies are either on, or close to, the periphery of the core. Since the MOX fuel has a harder neutron spectrum, a lower k_∞ (for the feed fuel) and lower power (for the feed fuel), there is *less* neutron leakage and therefore the cycle length should, if anything, be longer, not shorter.

In order to investigate this effect, the loading pattern was changed such that the MOX feed assemblies (and the 4.00 ^{235}U in the all UO_2 case) were loaded more towards the centre of the core. Again, an all-UO_2 and a MOX-UO_2 loading pattern were developed in order to determine the effects. The new 'less out-in-in' loading pattern can be seen in Figure 4.

	H	G	F	E	D	C	B	A
8	CENTRE X3	HIGH X1	LOW + FEED	HIGH X2	HIGH X2	LOW X2	LOW + X1	HIGH FEED
9	HIGH X1	HIGH + X2	HIGH X1	LOW X2	LOW FEED	HIGH + X1	HIGH X1	HIGH FEED
10	LOW + FEED	LOW X1	HIGH + X2	LOW X2	HIGH + X1	HIGH X2	HIGH + FEED	
11	HIGH X2	LOW X2	LOW FEED	HIGH + X2	LOW X1	HIGH X1	HIGH FEED	
12	HIGH X2	LOW FEED	LOW + X1	HIGH X1	HIGH + X1	HIGH FEED		
13	LOW X2	HIGH + X2	HIGH X2	HIGH X1	HIGH FEED			
14	LOW + X1	HIGH FEED	HIGH + FEED	LOW FEED				
15	HIGH FEED	HIGH X2						

fuel type
+ CRs
# dwells

FIG. 4. Equilibrium Loading Pattern -Less 'OUT-IN-IN

In this case, when the MOX assemblies replaced the 'LOW' UO_2 assemblies, there was a noticeable difference in cycle length of approximately 0.20 GWd/tHM. One reason postulated for this difference is that the matching of the LARs is completed on a *single* assembly basis and at a constant boron concentration and so there is no allowance for the neutron spectrum differences between the MOX and UO_2. In the whole core situation, the neutron spectrum will be somewhere between that of the UO_2-only and MOX-only cases. Therefore, a 2x2 supercell ("quad" model) was set up with one MOX assembly surrounded by three UO_2 driver assemblies in order to represent the whole core situation more closely. Figure 5 shows the k_∞ versus burnup for the single assembly MOX case and the MOX assembly in the 2x2 supercell. The single assembly model has a lower value of k_∞ for all burnups compared with the supercell model. Therefore, if the LAR *was* matched on the basis of supercell calculations, a lower MOX content would be predicted compared with using the single assembly results. Hence the cycle length would be even shorter.

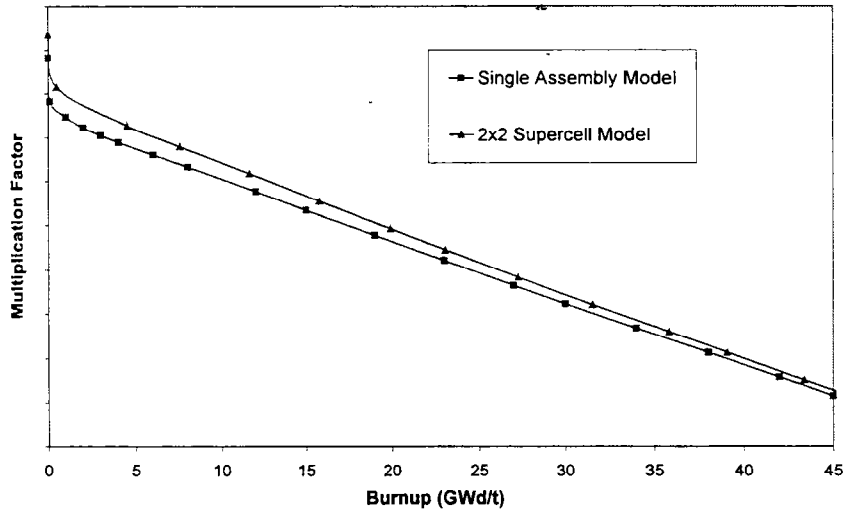

FIG. 5. Effect on Reactivity of Modelling Single Assembly or 2x2 Supercell

The actual reason for the difference in cycle length is believed to be due to the reactivity versus burnup nature of the MOX fuel compared with the UO_2. The beginning of life k_∞ for the MOX fuel is much lower than that of the equivalent UO_2, thereby altering the leakage of the core and hence the power share of the batches.

In summary, there are two key conclusions about the approach of matching the LAR of MOX to UO_2 at the lattice level; the effect of power sharing has a significant effect on the accuracy of LAR matching (it is possible to match reactivity but *not* power) and hence matching the LAR of a MOX assembly to that of a UO_2 assembly is not a viable approach.

Since matching the LAR of MOX to UO_2 failed because of the extreme differences between the reactivity versus burnup curves, it is reasonable to expect that matching the LARs for different Pu vectors will be more successful since the reactivity trends are much more similar. Therefore, the next stage in the investigation was to determine the effectiveness of matching MOX LARs.

In order to determine the MOX content required to match the all-UO_2 cycle using a PWR Pu vector (Case A in Table I) a *whole core* assessment was completed. By simply choosing 2 Pu contents that gave cycle lengths either side of the required value, linear interpolation was used to calculate the actual Pu content needed to match the required cycle length. In order to match the all-UO_2 cycle length of 15.45 GWd/tHM for the same core as before, a Pu content of 8.00 w/o Pu_{tot} was found to be required.

For each of the 5 Pu vectors in Table I, the LAR was matched (at the lattice level) to that of the 8.00 w/o Pu_{tot} to within ± 150 pcm. The calculated weight percentages can be seen at the top of Table II. It can be seen that as the quality of the Pu is reduced (i.e. the fissile isotopes reduce and the absorbing isotopes increase), the weight percent of MOX required to maintain cycle length increases. These MOX contents (based on LAR matching at the lattice level) were then loaded into the equilibrium whole core model, using *exactly* the same loading pattern as that used for the initial MOX-UO_2 core (see Figure 4). As can be seen in Table II, there was very good agreement between the cycle lengths for each of the Pu vectors investigated. It should also be noted that in all but one of the cases, the $F_{\Delta H}$ values are also in excellent agreement. Case E (high fissile Pu) is unique in that the reactivity versus burnup curve is very similar to the UO_2 curve, but the physics nature of the Pu results in a disparity as far as power peaking is concerned i.e. it is possible to match reactivity but not power. It should also be noted that the batch burnups for each of the Pu vectors are very similar to each other and more importantly the discharge burnup of the UO_2 fuel is not compromised by the presence of the MOX fuel. In summary, matching the LAR of MOX fuels works very well and is an effective and efficient way of predicting required Pu contents.

TABLE IL Whole Core Results for 30% MOX Core Fraction

PU CONTENT	TOTAL / FISSILE	UO2	Case A		Case B		Case C		Case D		Case E	
			8.00		6.70		7.40		5.80		3.95	
		4.5, 4.0	5.14		4.66		4.90		4.29		3.79	
CYCLE LENGTH (GWd/tHM)		15.45	15.45		15.45		15.45		15.47		15.40	
Critical Boron Conc.	BOC	1610	1610		1600		1550		1600		1860	
	EOC	10	10		10		10		10		10	
F-DELTA-H	BOC	1.467	1.519		1.522		1.564		1.517		1.821	
	EOC	1.329	1.331		1.334		1.330		1.336		1.317	
		HIGH LOW	*UO2*	*MOX*	*UO2*	*MOX*	*UO2*	*MOX*	*UO2*	*MOX*	*UO2*	*MOX*
BATCHBURNUP (GWd/tHM)	3rd dwell	45.5 48.4	44.9	49.7	45.1	49.3	45.1	49.3	45.2	49.2	45.0	49.0
	2nd dwell	31.5 35.5	31.3	34.8	31.4	34.7	31.5	34.4	31.5	34.7	31.1	35.6
	1st dwell	14.3 19.6	14.4	18.0	14.4	18.0	14.5	17.7	14.4	18.1	14.0	19.6
MTC (HFP) (pcm/F)	BOC	-8.24	-14.66		-14.70		-15.38		-14.47		-10.79	
	EOC	-33.30	-35.14		-35.32		-35.11		-35.31		-34.83	
Boron Coeff (pcm/ppm)	BOC	6.56	-5.21		-5.33		-5.33		-5.42		-5.39	
	EOC	-8.30	-6.47		-6.64		-6.51		-6.80		-7.39	
Control Rod Worth (ARO cf ARI) (pcm)	BOC	8720	7640		7750		7720		7840		7910	
	EOC	9542	8280		8410		8320		8520		8870	
Delayed n-fraction	BOC	0.0061	0.0053		0.0053		0.0053		0.0053		0.0049	
	EOC	0.0053	0.0048		0.0048		0.0048		0.0048		0.0048	

3.2 Whole Core Assessments

Using the MOX contents calculated above in Table II, the effect of the different Pu vectors and contents on the key core wide nuclear design parameters were calculated using SIMULATE-3. The results can also be seen in Table II.

One of the limiting factors for the use of MOX fuels in many commercial reactors is the delayed neutron fraction as this affects the transient response under fault conditions. As can be seen from the results, the delayed neutron fraction for any of the MOX contents is significantly lower (approximately 15 %) than that for the all UO_2 core. However, the delayed neutron fraction does remain almost constant for each of the cases, regardless of the Pu content or quality. This is an important conclusion as it indicates that MOX assemblies with a higher Pu content will not have a significant impact on this area of the transient behaviour of the core.

One of the key safety parameters with respect to MOX utilisation is control rod worth. This often contributes to a limitation on the use of MOX to only 30% loading in a standard PWR. From the results in Table II it can be seen how much the control-rod worth is eroded in going from an all UO_2 to a MOX-UO_2 core; a minimum of approximately 10% for Case E, and up to 13% for the higher Pu content, Case A. Again, the effect of the Pu content is not significant but more importantly the result indicates the extent to which the presence of any Pu affects the control rod worth. The reduced worth of the control rods re-emphasises the effect that MOX fuel has on neutron absorbers in the core e.g. burnable poisons and boron in the coolant.

This is illustrated by the significant effect on the boron coefficient. Table II shows that the boron coefficient is reduced by 20% in going from an all-UO_2 to a MOX-UO_2 core. There is a further reduction of approximately 5% in the boron coefficient as the Pu content increases from Case B to Case A. Again, this becomes an important consideration for either high Pu content MOX fuels (for longer cycle lengths) or for lower fissile Pu arising from high burnup UO_2 fuels.

The MTC for the MOX-UO_2 cores is more negative than for the all UO_2 core, particularly at the beginning of cycle (BOC) where the values are approximately 75% more negative. This can be seen as being a benefit because in most safety analyses, the MTC must be negative at all times in life. However, the problem associated with MOX cores is that MTC can become too negative during the

cycle (as the fission products build up in the core) which reduces safety margins in steam line break calculations. In general, the MTC becomes more negative as.the Pu content increases but not to a great extent.

Table III provides similar results for a MOX-UO_2 core with a MOX core fraction of approximately 45%. It should be noted that the MOX contents have now increased since the proportion of the 4.50 ^{235}U assemblies has now reduced and hence the MOX assemblies have to compensate for the shortage in cycle length. The loading pattern used for the 45% MOX core fraction can be seen in Figure 6.

TABLE 111. Whole Core Results for 45% MOX Core Fraction

PU CONTENT	TOTAL FISSILE	UO2 4.5, 4.0	Case A 8.50 5.46	Case B 7.00 4.87	Case C 7.80 5.16	Case D 6.10 4.51	Case E 4.10 3.94
CYCLE LENGTH (GWd/tHM)		15.45	15.44	15.43	15.48	15.42	15.25
Critical Boron Conc.	BOC	1610	1620	1600	1550	1600	1940
	EOC	10	10	10	10	10	10
F-DELTA-H	BOC	1.467	1.526	1.532	1.570	1.532	1.727
	EOC	1.329	1.366	1.362	1.356	1.363	1.370
		HIGH LOW	*UO2 MOX*	*UO2 MOX*	*UO2 MOX*	*UO2 MOX*	*UO2 MOX*
BATCH BURNUP (GWd/tHM)	3rd dwell	45.5 48.4	44.5 48.4	44.8 48.1	44.8 48.3	44.9 47.8	44.6 47.1
	2nd dwell	31.5 35.5	31.2 32.7	31.3 32.6	31.4 32.5	31.5 32.5	31.1 33.2
	1st dwell	14.3 19.6	14.5 15.8	14.6 15.7	14.7 15.5	14.6 15.8	14.4 16.9
MTC (HFP) (pcm/F)	BOC	-8.24	-16.64	-16.87	-17.57	-16.69	-11.77
	EOC	-33.30	-35.45	-35.87	-35.51	-35.95	-35.27
Boron Coeff (pcm/ppm)	BOC	-6.56	-4.58	-4.77	-4.71	-4.91	-5.08
	EOC	-8.30	-5.72	-5.97	-5.77	-6.18	-6.99
Control Rod Worth (ARO cf ARI) (pcm)	BOC	8720	7070	7270	7200	7410	7520
	EOC	9542	7680	7910	7760	8074	8560
Delayed n-fraction	BOC	0.0061	0.0049	0.0049	0.0049	0.0049	0.0047
	EOC	0.0053	0.0046	0.0046	0.0046	0.0046	0.0046

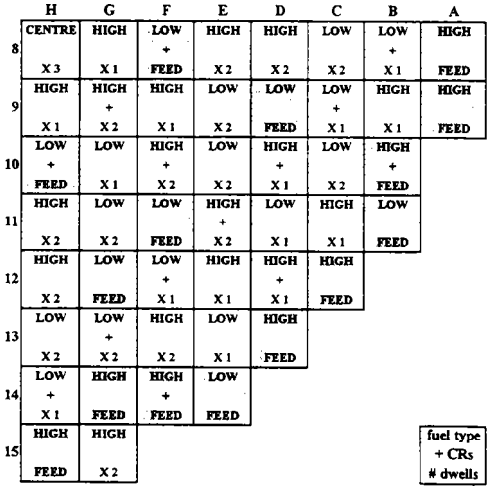

FIG. 6. Equilibrium loading pattern for 45% MOX Core Fraction

As expected, the introduction of a further 24 MOX assemblies into the equilibrium cycles has had a significant impact on the boron coefficient, MTC, delayed neutron fraction and control rods worth. The MTC is more negative for the higher core fraction; approximately 15% more negative, which coincides with the additional 15% MOX core fraction. The boron coefficient has also been affected to a similar extent with a further reduction of approximately 10%. The delayed neutron fraction has also decreased by approximately 10% in each case. All of these effects are core wide issues and are therefore independent of the loading pattern used and are simply due to the fact that

there is more Pu in the core. However, the reduction of control rod worth (by approximately 5 to 10%) is very much linked to the loading pattern. By keeping the.MOX assemblies (particularly the more highly burnt) from the control rod locations, it is possible to minimise the impact of a higher core fraction. Furthermore, the loss in control rod worth can be mitigated by, for example, using either enriched control rods, or introducing additional control rod banks into the core.

Similarly, the impact on the boron worth can be reduced by the use of enriched boron in the coolant. This would help particularly with the differential boron worth as there would be less duty on the boration system to achieve the same reduction in reactivity. However, the use of enriched boron will have little or no effect on the MTC issue since it is the total amount of neutron absorption that affects the MTC and that will be the same irrespective of the enrichment of the ^{10}B.

4. SUMMARY

This report has investigated the impact of MOX fuel made from differing Pu sources on core wide nuclear design parameters and the effect of increasing the MOX core fraction from 30% to 45%. Plutonium fissile fractions ranging from 64% to 96% have been investigated and some of the key concerns with respect to the core physics have been explored. The report has also provided an illustration as to the effectiveness and limitations of using a Lifetime Average Reactivity (LAR) approach to matching MOX fuel compositions to other MOX fuels or to UO_2.

The study has illustrated two key conclusions about the approach of matching the LAR of MOX to UO_2 at the assembly level; the effect of power sharing has a significant effect on the accuracy of LAR matching (it is possible to match reactivity but not power) and hence matching the LAR of a MOX assembly to that of a UO_2 assembly is not a viable approach. However, matching a MOX assembly with one Pu vector to that of another quality works very well, and is a rapid and effective way of calculating required MOX contents, but a match at the whole core level must be completed initially.

For the 30% MOX core fraction, it has been shown that the core physics parameters generally show only modest variations from one fissile quality to another. This suggests that in terms of overall core physics performance, the implications of loading MOX fuel with any Pu vector that may arise from current reactors is not too pronounced. The effect of loading a 45% MOX core fraction results in a further impact on the core physics parameters, but this can be mitigated by use of alternate, yet proven technology e.g. enriched control rods and enriched boron in the coolant.

BNFL has many reprocessing customers throughout the world and therefore potentially a diverse range of Pu sources to manufacture into MOX fuel. With the growing experience in MOX fuel irradiation around the world using many different Pu sources, it is important to understand the whole core effects of such a wide variety of fissile compositions and to ensure the continued safe and efficient utilisation of MOX fuel.

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PLUTONIUM RECYCLING IN PWR

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Abstract

Two concepts of 100% MOX PWR cores are presented. They are designed such as to minimize the consequences of the introduction of Pu on the core control. The first one has a high moderation ratio and the second one utilizes an enriched uranium support. The important design parameters as well as their capabilities to multirecycle Pu are dicussed. We conclude on the potential interest of the two concepts.

1. INTRODUCTION

Recycling plutonium in standard PWRs using 100% MOX loadings brings about a degradation of the control mean efficiency. Different solutions exist, in order to mitigate this effect, and have to be analyzed and compared. One possibility could be to increase the moderation ratio (Highly Moderated Reactor concept). Another way of improvement, while using a standard PWR, is to limit the plutonium content in the reactor, which means that to reach the target burnups, enriched uranium must be used (MIX concept). Other possibilities, like the APA [1] (Advanced Plutonium Assembly) concept are, as well, under study at CEA. This paper presents the analysis of the MIX and HMR concepts performed with the codes APOLLO2 (preparation of the group constants) and CRONOS2 (3D diffusion and fuel management).

One must notice that the MIX and HMR belongs to different strategies. The MIX belongs to a 'dilution' strategy, i.e. each reactor of the park contains Pu. In particular, it is shown that a PWR core with a 100% MIX loading characterized by a 2% Pu content has a zero Pu mass balance. On the other hand, the HMR is designed to burn as much Pu as possible and must be fed with Pu coming from UOX cores.

2. HYPOTHESIS

The MIX has been developped so that Pu can be recycled in a standard 1300 MWe reactor without any modifications, whereas the HMR has been designed so that it is compatible with the 1450 MWe EPR (European Pressurized Reactor) vessel.

A 3x18 months fuel management is considered, corresponding to average discharge burnups of, respectively, 45 Gwd/t and 66 Gwd/t for the MIX and for the HMR. The Pu isotopic composition considered (Table I) represents the average Pu that will be available in France around 2015; it takes into account the Pu coming from UOX as well as MOX. The results obtained with different fuel managements and Pu isotopic compositions are presented in Ref. [2] and [3].

TABLE I. PLUTONIUM ISOTOPIC COMPOSITION CONSIDERED IN THE STUDY

Pu^{238}	Pu^{239}	Pu^{240}	Pu^{241}	Pu^{242}
2.7	56.0	25.9	8.1	7.3

3. ASSEMBLY CHARACTERISTICS

3.1. The MIX assembly

The main interest of the MIX concept is that no modifications of neither the assembly nor the vessel are necessary. Thus, the data concerning the geometry of the MIX assembly come from the standard 1300 MWe EdF (Electricité de France) reactors, i.e. a 17x17 lattice with a 1.26 cm pitch and 25 water holes. Each assembly contains 535 kg of heavy metal (HM), and the specific power is 36.8 W/g. The U^{235} enrichment necessary to meet the fuel cycle length requirement is calculated for different Pu contents. It is found to be an almost linear function of the Pu content, going from 4.0% U^{235} for 0% Pu (i.e. UOX fuel) to 0.25% U^{235} (i.e. tail uranium) for 9.5% Pu.

As shown on Fig. la and b, the reactivity coefficients (fuel and moderator temperature coefficients and boron efficiency) deteriorate up to 4% Pu, and then stabilize, except for the boron efficiency which keeps decreasing. In order to keep some control margin, the boron efficiency should not be lower (in absolute value) than -4 pcm/ppm which sets the limit on the Pu content at 4%. The uranium support must be enriched up to 2.5% U^{235} in order to fullfill the fuel cycle requirement (3x18 months).

3.2. The HMR assembly

A parametric study carried out to assess the effect of the moderation ratio demonstrates the strong impact on the reactivity coefficients. For example, in a 100% MOX core, the increase of the moderation ratio from 2 (standard PWR) to 4, brings about an increase of the boron efficiency by a factor 2.5, and a 20% decrease on the doppler coefficient. The moderator temperature coefficient becomes less negative as well. The main cause is the difference in the Pu inventories: increasing the moderation ratio provokes a 60% decrease of the Pu mass in the assembly (for a similar fuel management). Furthermore, the moderation ratio must be as high as possible in order to improve plutonium consumption and reduce minor actinide production

However the safety criterion concerning the nucleated boiling crisis sets limits on the surface heat flux released by the fuel rods. Thus at constant power, a geometry of the 17x 17 type cannot be retained for moderation ratios higher than 3.1. But in turning to geometries of the 19 x 19 type (while retaining the external dimensions of the fuel assemblies), a moderation ratio of 4 can be reached. The latter configuration was chosen; it is characterized by a fuel pellet diameter of 6.12 mm and a 1.13 cm pitch. In this situation the neutron spectrum is thermalized but it remains characteristic of a MOX fuel. The specific power is high (56.6 W/g) and the Pu content necessary to meet the fuel cycle length is 9.7% (tail uranium is used, i.e. 0.25% U^{235}). Compared with a 17x17 assembly with a moderation ratio of 2, the 19x19 assembly chosen contains 40% less HM, i.e. 3 12 kg per assembly. Among the various solutions studied, the lattice containing 81 guide tubes (Fig. 2) presents the best power flattening within the assembly owing to the regularity of the lattice.

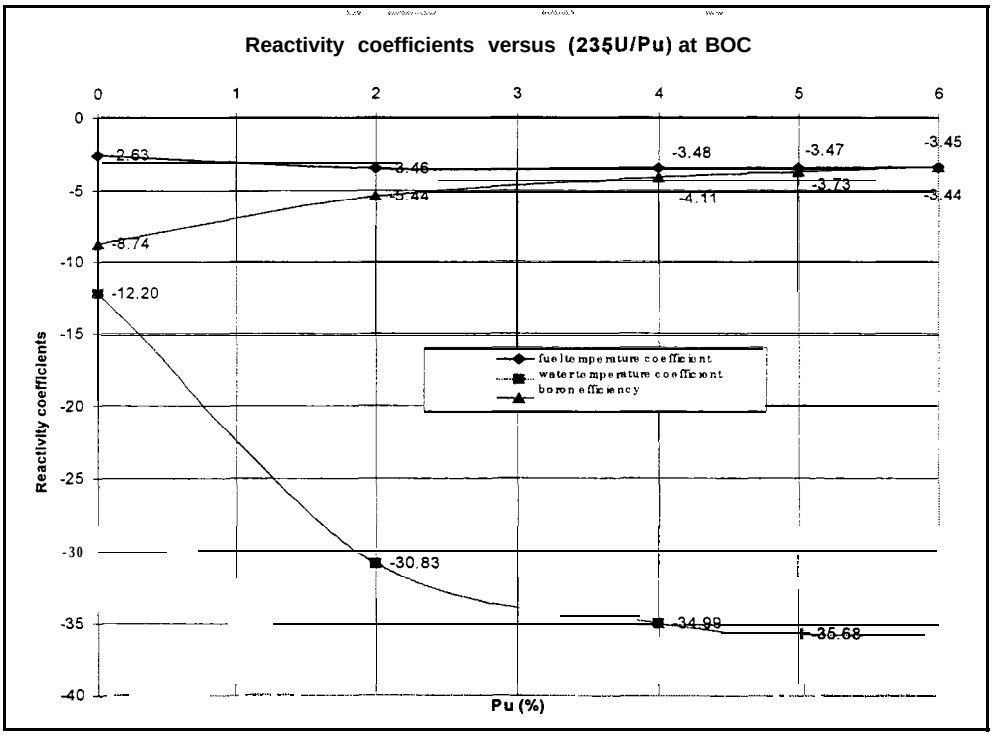

FIG. IA. Reactivity coefficients (at 150 MWd/t) vs. the Pu content

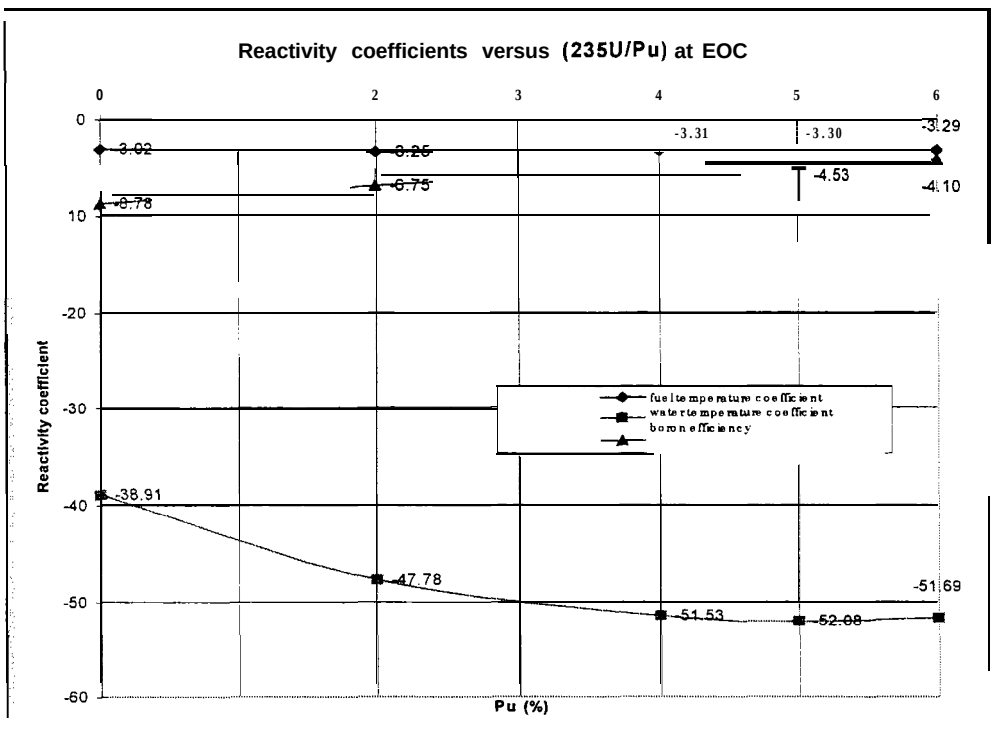

FIG. 1 B. Reactivity coefficients (at 45000 MWd/t) vs. the Pu content

4. MASS BALANCES

The Table II gives the Pu and MA (Minor Actinides) consumption for the two concepts. Furthermore, for the MIX, the needs in natural uranium as well as SWU (Separation Work Units) are compared to those of a reference 1300 MWe UOX core (3x18 months).

In the MIX, the gain in terms of SWU is important compared to the UOX (factor of 2). Furthermore, the U^{235} residual enrichment is not negligible, i.e. 1.14%; only 55% of the U^{235} is actually fissionned compared to 80% in the UOX. The MIX Pu burning rate is 25 kg/TWhe, however 41% is transmuted into MA (10.3 kg/TWhe). The HMR uses tail uranium, thus the needs in natural uranium and SWU are moot points. The interest of the HMR is highlighted by its capability to fission Pu. Of the 80.3 kg/TWhe of Pu destroyed, only 12.8% is transmuted into MA (10.3 kg/TWhe). The parametric study showed that the higher the moderation ratio, the smaller the MA quantity produced. At each cycle (i.e. 18 months) , one third of the assemblies are changed, which means that 34.2 tons and 24.9 tons of HM are reloaded, respectively, in the MIX and in the HMR.

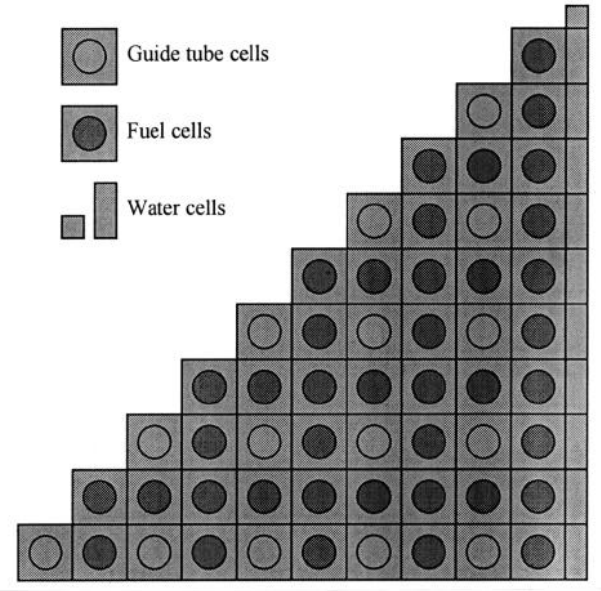

FIG. 2. 19×19 assembly, 81 guide tubes, $V_{mod} / V_{fuel} = 4$

TABLE II. MASS BALANCES FOR THE MIX, HMR AND A REFERENCE UOX WITH 3X18 MONTHS FUEL MANAGEMENT

Reactor Type	*Teneur Pu (%)*	*U^{235} enri. (%)*	*U^{235} resi. (%)*	*Δ Pu (kg/TWhe)*	*Δ A.M. (kg/TWhe)*	*U_{nat} (kg/tHM)*	*SWU (kg/tHM)*
UOX	0	4.0	0.85	+ 30.1	+ 3.6	8152	5839
MIX	4.0	2.53	1.14	- 25.0	+ 10.3	4956	2903
HMR	9.7	0.25	-	- 80.3	+ 10.3	-	-

5. CORE CHARACTERISTICS

The MIX and HMR cores contain, respectively, 193 and 241 of the assemblies described above. The soluble boron (B_{nat}) concentrations, the reactivity coefficients and the control rods' efficiency (from full power to hot shutdown) are given in Table III.

In a UOX core some of the control rods are weakly absorbant ('grey' clusters) whereas some are strongly absorbant ('black' clusters). This is designed in order to limit the perturbation on the flux, and limit the probability of any unstabilizing xenon-induced oscillations. Since the xenon capture rate is significantly smaller in a Pu loaded core than in a UOX core, we consider that all the clusters are made of strongly absorbant materials. For the

MIX the control and shutdown rods (65 clusters) are made of $B_{nat}4C$[1]. For the HMR the 33 control rod clusters are made of Hafnium whereas the 72 shutdown rod clusters are made of $B_{nat}4C$.

When the reactor is brought from its hot shutdown state to its cold shutdown state, characterized, respectively, by isothermal core temperatures of 296°C and 20°C, the reactivity increases. The safety authorities require that the reactor must be at least subcritical by 5000 pcm when it is in its cold shutdown state; as a consequence boron must be added to the moderator but should not, however, exceed the limit of 2500 ppm because recrystallization might then occur in auxilliary circuits. The calculations performed for the most penalizing situation, i.e. BOC, showed that 1470 ppm are sufficient to meet the safety criteria in the HMR, whereas the MIX needs 2645 ppm. Slightly enriched soluble boron or burnable poison should then be used in the MIX.

TABLE III. CORE REACTIVITY COEFFICIENTS AND CONTROL RODS' EFFICIENCIES

	MIX		HMR	
	BOC	EOC	BOC	EOC
C_{boron} (ppm)	1820	0	1500	0
α_{Tfuel} (pcm/°C)	-3.3	-3.4	-2.2	-2.1
α_{Tmod} (pcm/°C)	-24.9	-55.0	-15.1	-57.6
α_{Cboron} (pcm/ppm)	-3.8	-4.6	-5.8	-8.0
Reactivity at hot shutdown (pcm)*	-6210	-5780	-9685	-11070
Most effective cluster (pcm)	970	1210	3910	4620

* calculated as $(k_{eff}-1)/k_{eff}$ - all clusters inserted

6. ACCIDENTAL SITUATIONS

6.1. Control rod ejection

This accident has been analyzed at full power and zero power (critical reactor in both cases). For both the MIX and the HMR, the reactivity inserted is larger at zero power than at full power. The reactivity inserted is not very large, 380 pcm (0.7$) for the MIX and 320 pcm (0.9$) for the HMR. The same calculations performed with a UOX core (control clusters in Silver-Indium-Cadmium, shutdown clusters in B4C) results in a reactivity insertion of 280 pcm (0.5$).

6.2. Cooling transient

An unexpected valve opening leads to an overcooling of the core and to an increase of the reactivity. The reactor initially in its hot shutdown state, with the most effective cluster supposedly blocked out of the core, should not go critical during the 15 minutes necessary to the operator to stop the cooling. The calculations were performed at EOC since the moderator temperature coefficient is more negative. The analysis showed that this accident leads to an approximately 2500 pcm reactivity increase for the two concepts; neither cores go critical. However, if penalties of 10% are taken into account for all the parameters involved (reactivity

[1] Because of the swelling occuring in B4C rods, the control clusters should be changed every 3 years

coefficients, temperature, and moderator density) then the cores can go critical. For comparison, the reactivity increase calculated for a UOX core is about 1200 pcm.

7. MULTIRECYCLING CAPABILITIES

7.1. Multirecycling Pu in the HMR

The HMR is initialy loaded with Pu coming from UOX reactors (Fig. 3), i.e. containing about 64% fissile Pu. The Pu degrades very rapidly and at the end of the first cycle it contains already only about 39% fissile Pu. As a consequence, the Pu content must be increased as well; It goes from 10.1% for the first cycle to 13.7% and 16.1% for the second and third cycles. After 10 cycles, 25% Pu are needed, which is not realistic vis-à-vis the void coefficient; hence the number of recycling might be limited to two. Furthermore, the proportion of the Pu transmuted into MA, instead of fissionning, increases: 15% for the first cycle, 21% and 24% for the second and third cycles. The fraction of HMR necessary to balance the Pu production from UOX remains about constant: between 15 and 20%. The gain in terms of SWU is about 20% compared to a 100% UOX park.

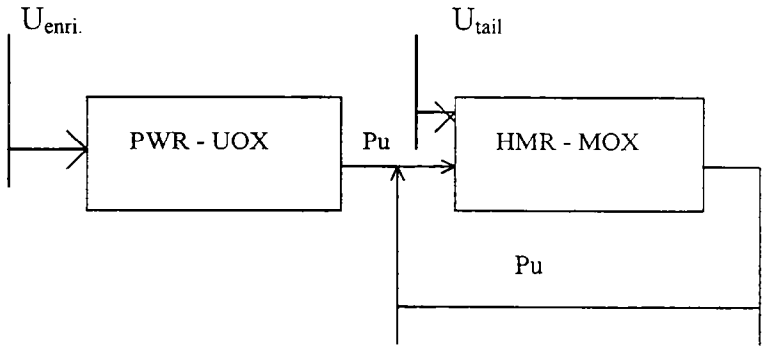

FIG. 3. Pu multirecycling scheme with the HMR

7.2. Multirecycling Pu in the MIX

As said in the introduction, the Pu management with the MIX implies the dilution of this Pu in all the reactors of the park (Fig. 4), i.e each reactor takes care of its own Pu. The Pu content corresponding to a zero Pu mass balance is about 2%; above this value the reactor burns Pu, whereas below this value it produces Pu. At equilibrium, the U^{235} enrichment needed to meet the fuel cycle length requirement (3x18 months - 45 Gwj/t) is about 3.4% compared to 4% for a UOX 3 x18 months. The gain in terms of SWU is about 20% compared to a similar park made up of only UOX cores. Hence, from a physics point of view, Pu management in MIX cores is an attractive solution for the short term.

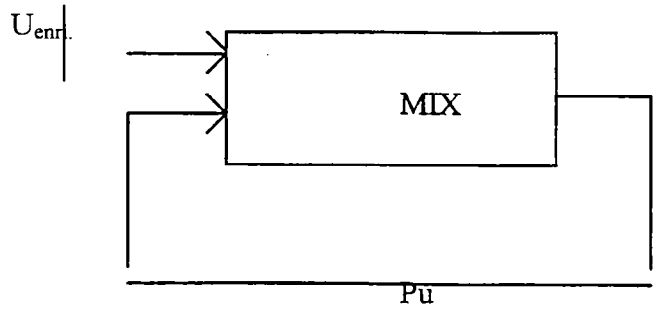

FIG. 4. Pu multirecycling scheme with the MIX

8. CONCLUSIONS

Introducing large amounts of Pu in a standard PWR core brings about penalties, especially on the control mean efficiency. Two solutions allowing to recycle Pu in a PWR and in the same time limit the consequences on the core control are presented. The first one (HMR) uses a high moderation ratio together with a tail uranium matrice and the second (MIX) uses a standard 17x17 assembly and an enriched uranium matrice. The two cores are characterized, and no major problems forbidding their realizations show up. In the MIX strategy, the Pu is diluted in all the reactors of the park, whereas the HMR must fed by Pu coming from UOX. The Pu multirecycling capabilities are assessed and show that the Pu content must be increased rapidly in the HMR because of its degradation; only two recycling might be allowed. In the MIX, the multirecycling is feasible: the Pu content stabilizes at about 2% and the U^{235} enrichment is 15% lower than in the corresponding UOX core.

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ADVANCED MIXED OXIDE FUEL ASSEMBLIES WITH HIGHER PLUTONIUM CONTENT FOR LIGHT WATER REACTORS

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Abstract

The MOX introduction in LWRs (PWR and BWR) was started in Germany with initial steps of design in the early 70ies. The process of commercial utilisation of Pu recycling was based on these designs and initial MOX insertion at the Obrigheim-plant KWO (PWR) and the Gundremmingen-plant Unit A KRB-A (BWR). The needs of testing and validation of used methods could mainly be fulfilled by the insertion of initial **MOX-**FA reloads. Experiments with this early MOX-FAs have been conducted under fully realistic power reactor conditions for neutronic/nuclear and fuel/technological aspects.
Optimising fuel cycle costs by increasing the final burnup leads to reduced generation of plutonium. Under properly defined boundary conditions thermal recycling in MOX-FAs reduces further the amount of Pu which has to be disposed of to final storage.
Increasing the final burnup requires higher initial enrichments of U-fuel to be matched by an advanced design of MOX-FAs with higher Pu contents. The neutronic design of these MOX-FAs has to consider the licensing status of NPPs concerning the use of MOX fuel and the evolution of U fuel enrichment and burnup level.
The Siemens Nuclear Fuel Cycle Division, with more than 20 year's experience in the production of MOX fuel, has designed several advanced MOX FAs of different types for PWRs (14x14 to 18x18) as well as for BWRs (9x9 and 10x10) with averaged contents of fissile plutonium up to 5.85 w/o. Some reloads of this kind are at present under irradiation in different NPPs.

1. INTRODUCTION

For several years a shift towards higher burnup has taken place in all countries using light-water reactors (LWRs) in the effort to minimise amounts of radioactive waste, especially to reduce the number of fuel assemblies which have to be disposed of, to save resources of fissile material by reducing the numbers of new fuel assemblies, and to minimise fuel cycle costs to improve the economics of nuclear power generation.
This trend towards higher burnup is accompanied by the thermal recycling of plutonium from reprocessed fuel. Several countries including Belgium, France, Germany, Japan and Switzerland are already using MOX (Mixed-Oxide) FAs (Fuel Assemblies) in LWRs or intend to do so [l]. For more than two decades, Germany has been gaining practical experience in the thermal recycling in both boiling water reactors (BWRs) as well as especially in pressurised water reactors (PWRs) [2]. This was in accordance with the national Atomic Energy Act. Until mid 1994, this was the only established method of nuclear waste disposal.
The experience of SIEMENS (KWU) covers more than 20 years of MOX FA design (neutron physical and mechanical), production and insertion in BWRs as well as especially in PWRs, as can be seen in the Fig. 1. Thermal recycling of plutonium started under commercial conditions at Obrigheim Nuclear Power Plant (KWO) in 1972 (PWR) and at Gundremmingen Nuclear Power Plant Unit A (KRB-A) in 1974 (BWR). After the introduction of the improved production methods OCOM (Optimised Co-Milling) and A (U, Pu) C (Ammonium-Uranyl-, Plutonyl-Carbonate) in 1980 MOX FAs have been used in other plants such as Neckarwestheim Unit 1 (GKN I) since 1982 and Unterweser (KKU) since 1984 [3], [4]. Up to 30 000 MOX fuel rods have been inserted per year in these reactors, some of them spending six irradiation periods in the reactor core.

As the national German fast breeder program had been cancelled, thermal recycling was the only way of using large quantities of plutonium for the energy production and helps to avoid accumulating separated Pu and the final disposal of plutonium.

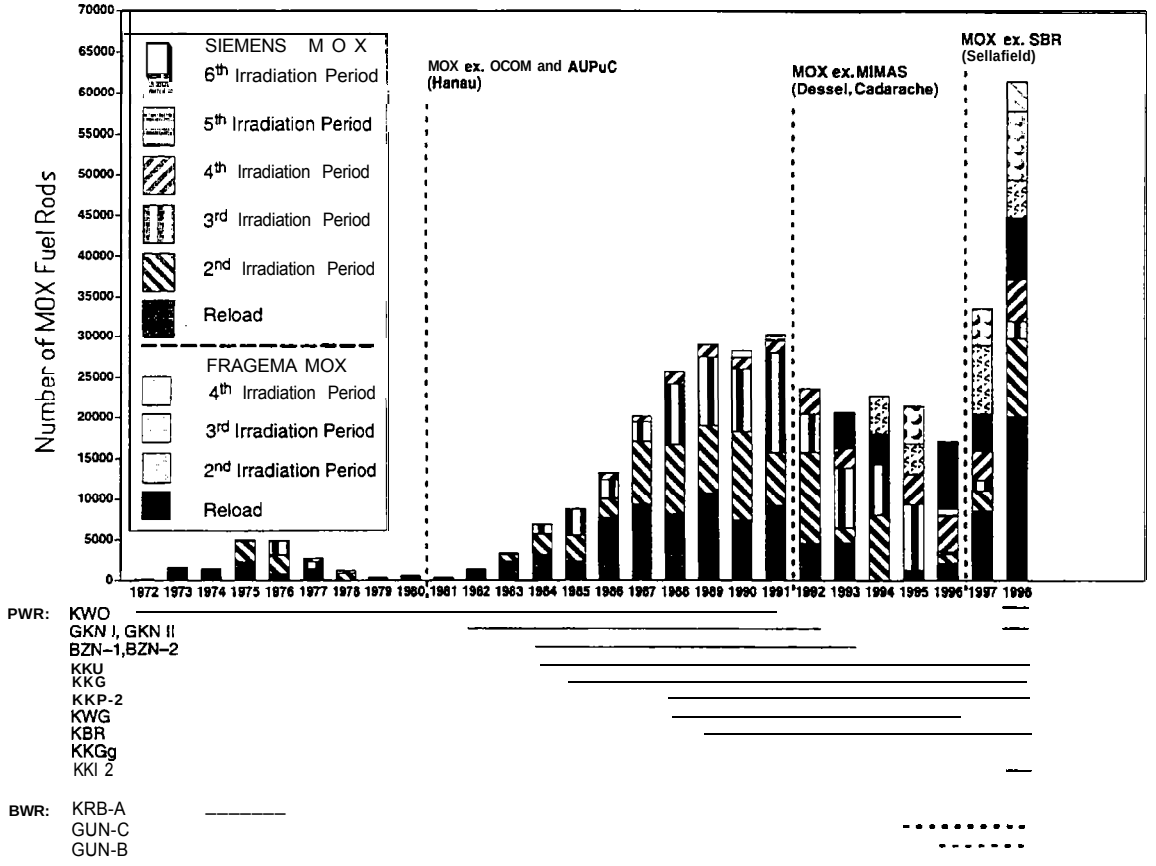

FIG. 1. Core Management Experience with commercial MOX Insertion in B WR and P WR by Siemens KWU Group (Status End of 1998)

The basic idea of thermal recycling is to insert MOX FA into normal LWR cores. As it is common practice to use MOX FAs together with U FAs in the same core it has to be the principal aim of nuclear MOX FA design to make them as compatible as possible with U FAs, so that they can be used instead of U FAs without any restrictions. Changes in cycle length are to be avoided. In order to achieve this, the fissile material content has to be dimensioned to obtain MOX FAs, which have the same burnup potential as the U FAs (burnup equivalence). Fuel management studies must be carried out to confirm that equilibrium cycle lengths of cores with MOX FAs are the same as those of cores without MOX FAs.

2. PLUTONIUM BALANCE

The debate and the public discussions on some aspects of the thermal plutonium recycling are often confused, above all with respect to the plutonium balance. The essential issue is the question, whether thermal recycling actually increases or decreases the amount of plutonium, which has to be handled. Especially uncertain definitions and general conditions within the complex system of the fuel cycle mostly cause this confusion. Quantitative statements can only be made with scenarios having properly defined boundary conditions.

In the following scenario, a PWR with 1300 MW electrical energy generation (1300 MWe) operating 320 equivalent full power days (efpd) per year is considered and discussed. The annual production of electric power is set to 10 billion kWh, which is a typical value for German nuclear power plants of the 1300 MWe class. Nuclear calculations to analyse the plutonium generation and to obtain a plutonium balance are done using the zero-dimensional burnup code KORIGEN [5]. In these calculations the cross-section libraries are based on the JEF nuclear data generated by the FZ Karlsruhe (former: Kern-Forschungszentrum-Karlsruhe). Some input parameters such as burnup dependent cross sections of actinide nuclides and spectral parameters were calculated with the Siemens (KWU) standard design program system SAV90 [6] including the spectral code FASER.

In general, increased burnup can be achieved by increasing the initial enrichment. If the cycle length remains unchanged, the number of new (fresh) fuel assemblies which have to be reloaded every year has to be reduced. In cores without MOX FAs, for example, the reload batch of 64 FAs (initial enrichment of 3.1 w/o 235U) can be reduced to 48 FAs with an initial enrichment of 3.7 w/o ^{235}U without any influence on the cycle length. Due to longer irradiation in the core, the total amount of plutonium generated in any FA increases. However, in terms of the amount of plutonium unloaded every year in the spent fuel assemblies of one batch, the smaller batch size results in a reduced final quality and quantity of plutonium. A scenario without thermal recycling is listed in the upper part of Table I. Increasing the burnup from 35 MWd/kg to 45 MWd/kg reduces the amount of unloaded fissile plutonium from 205 kg to 165 kg (- 20%) per year; the total amount of plutonium is reduced from 312 kg to 264 kg (-15%) per year.

Thermal recycling can further reduce the amount of plutonium generated per year, as is apparent from Table I. The plutonium generated in 54 U FAs is just enough to fabricate 10 MOX FAs with a burnup equivalent content of fissile plutonium Pu_{fiss} = ($^{239}Pu + ^{241}Pu$). Natural uranium Unat is used as carrier material for the MOX FAs. The isotopic composition of the plutonium in the MOX FA corresponds to the plutonium produced in an U FA with an initial enrichment of 3.1 w/o ^{235}U and a burnup of 35 MWd/kgHM considering a decay of 241Pu of seven years between discharge of U FA and MOX FA reloading. The quality of the used plutonium, defined as the sum of the mass fractions of the fissile plutonium isotopes 239Pu and ^{241}Pu relative to the sum over all Pu isotopes, is 66 %. It is assumed that only spent U FAs are reprocessed and that burnt MOX FAs are consigned to the final storage. As can be seen from the Table I, the amount of plutonium, which has to be disposed of in the case of recycling, is reduced to 107 kg Pu_{fiss} and 195 kg Pu_{tot}, respectively.

TABLE I. PLUTONIUM BALANCES FOR DIFFERENT BURNUP AND WITH/WITHOUT RECYCLING (PRODUCED BY A PWR OF 1300 MW ELECTRICAL POWER)

	Pu to be disposed of to final storage			
	Without recycling (once through cycle)		**with recycling (one MOX generation)**	
Reload	**kg Pufiss**	**kg Putot**	**kg Pu_{fiss}**	**kg Putot**
64 U FAs	205	312		
48 U FAs	165	264		
54 U FAs				
10 MOX FAs			107	195
42 U FAs				
6 MOX FAs			94	172

If U FAs with 3.7 w/o 235U are used to achieve higher burnup, 42 U FAs and 6 MOX FAs have to be loaded. Again an equilibrium scenario is assumed. Owing to the higher burnup, the quality of plutonium resulting from the reprocessing of the corresponding U FAs has decreased to 62 %. As it is a basic aim of MOX FA design to produce compatible and burnup equivalent MOX FAs, the MOX FAs have to be designed for higher burnup in this case (containing the same burnup potential as the U FAs with an 235U content of 3.7 w/o) and to compensate for lower Pu quality. Also here the carrier

material depleted Uranium Utails was chosen, which allows the plutonium content to be increased in the MOX FA. In this case of recycling, 94 kg Pufiss and 172 kg Pu_{tot}, respectively, have to be placed in the final storage, assuming again that MOX fuel is not reprocessed. These trends of reducing amounts of Pu to be disposed of continue for higher reload enrichments and higher batch burnup values.

Although only the once-trough MOX fuel cycle is discussed here, reprocessing of spent MOX FAs has been demonstrated. The plutonium of the so-called second recycling generation can be used together with plutonium from reprocessed U FAs. The worsened Pu quality (- 55 % - 60 % depending on burnup and mixing ratio between plutonium of the first and the second recycling generation) can also be compensated by increasing the absolute content of fissile Pu isotopes in the MOX FA to about 7 w/o.

3. CURRENT STATUS OF LICENSING IN GERMANY

The present status of MOX licensing for German nuclear power plants has given rise to certain differences caused by the different procedures adopted by utilities and state authorities. An overall-view of the current licensing status of MOX licenses, which are in use or have been granted for German LWRs is given in Table II. The numbers of MOX FAs per reload or the total numbers in the core given in the table are restricted only by the licenses and not by technological limitations.

TABLE II. CURRENT STATUS OF MOX LICENSING FOR LWRS IN GERMANY

Reactor type	Plant-Name	Status of License	Pu_{fiss}-Content in w/o	Number of MOX-FAs per Reload	MOX-FA-Content in the Core in %
PWR:	KWO	in use	3.8	8	26
	GKN I	in use	3.04		9
	GKN II	in use	3.8 [1)]		37
	KKU	in use	3.5 [4)]	16	33
	KKG	in use [*)]	3.07 [1)]	16	33
	KK12	in use	equivalent to 4.0 w/o ^{235}U	24	50
	KWG	in use	3.2	16	33
	KBR	in use	equivalent to 4.0 w/o ^{235}U	- [3)]	- [3)]
	KKP-2	in use	4.65	24	37 [2)]
	KKE	granted [*)]	equivalent to 4.0 w/o ^{235}U	16	35
	KWB A	in preparation*)	equivalent to 3.5 w/o ^{235}U	24	42
	KWB B	in preparation [*)]	equivalent to 3.5 w/o ^{235}U	24	42
	KMK	in preparation	-		
BWR:	GUN B/C	in use [*)]	2.57/3.6	2x64	38
	KKB	in preparation	-		
	KKK	in preparation	-		

[1)] changes in the carrier material and/or Pu-quality can be compensated
[2)] temporary restriction
[3)] according to the amount of Pu-generation in the plant (up to ~ 16 MOX FAs per reload)
[4)] max. nominal Putiss content of a fuel rod
[*)] modification or extension in preparation

Based on the principles established for former MOX designs of FAs of the types 14x14, 15x15, and 16x16, a "standard" MOX FA was designed (Fig. 2) for use at five 1300 MWe plants (KKU, KKG, KWG; KKP-2, and KBR) and has been in service since the mid-80s with good operating results [7]. The average fissile plutonium content is 2.91 w/o in three FR (fuel rod)- types with different fissile plutonium contents. Four additional water rods (i. e. cladding tubes filled with water in connection with the water of the primary circuit) at the centre of the FA increase the moderation there in order to flatten the power distributions.

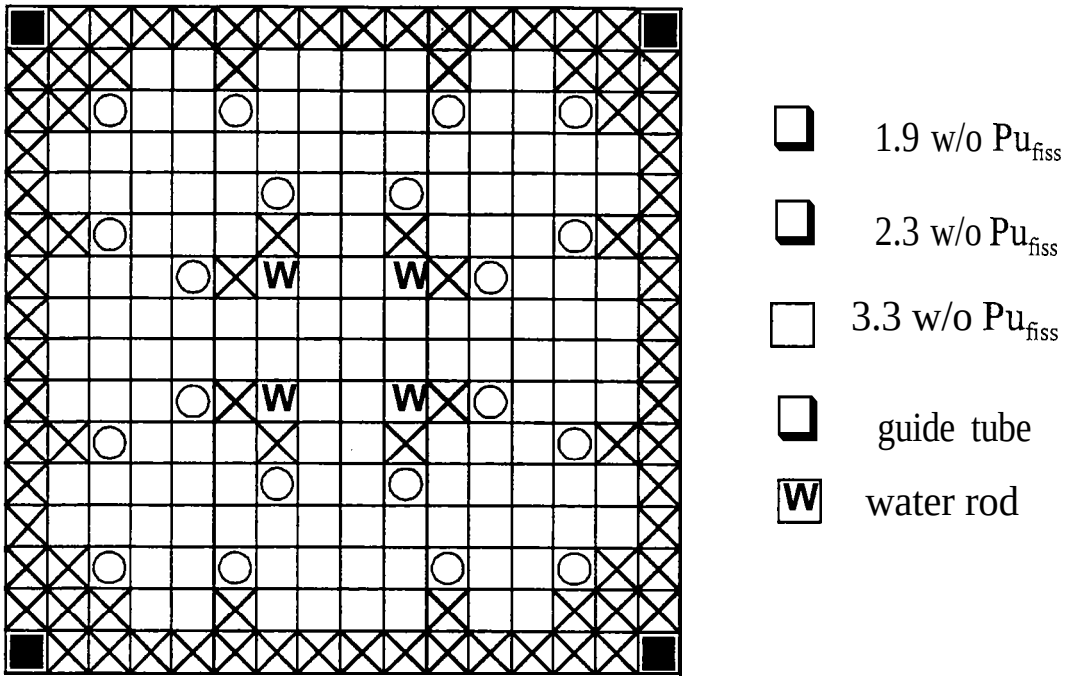

FIG. 2. Standard MOXFA (16x16), carrier material U_{nat}, 2.91 w/o average Pu_{fiss} content

As it has been common practice in Germany in the past to license Unat as carrier material, planned changes to depleted uranium could not be compensated under some existing licenses by increasing the fissile plutonium content. In this case burnup equivalent MOX FA design could not be realised up to now. Here new licenses are necessary, which are in preparation, in licensing procedure or already granted. Lower Pu quality has to be treated in the same way, where burnup equivalence requires compensation for higher contents of the neutron-absorbing isotopes ^{240}Pu and ^{242}Pu by increasing the Pu_{fiss} content.

4. MOX FA DESIGN WITH RAISED Pu_{FISS} CONTENT

The above mentioned standard MOX FA had to be redesigned for several reasons:

- The enrichment of U FAs loaded together with MOX FAs is increasing. In order to obtain equivalent MOX FAs, the content of fissionable plutonium nuclides has to be increased too. Usually, this requires a change in geometrical arrangement of the fuel rods with different Pu_{fiss} contents within the FA.
- Instead on U_{nat}, depleted uranium (Utails) is proposed as carrier material. The lower content of the fissionable nuclide ^{235}U in the matrix material has to be compensated by increasing the Pu_{fiss} content. In this case the available plutonium can be concentrated in fewer FAs.
- Changes in the Pu quality are caused by higher burnup of the reprocessed U FAs. This requires higher Pu_{fiss} contents to compensate for the effects of the neutron absorbing plutonium isotopes ^{240}Pu and ^{242}Pu, respectively.

4.1 **MOX FA designs used to date**

Designs for all boundary conditions have been established.
Examples are given in the following chapters. In all cases of MOX FAs for PWRs the main features developed by Siemens

- maximum three different fuel rod types (Pu_{fiss} contents)
- water rods in the central region of the FA

are established also international as a proven design.

4. I. I MOXFA designs compatible to enrichments of U FAs up to 4.0 w/o ^{235}U

A first design with raised plutonium content was made for a NPP with 14 x 14 rod lattice in 1987 to match the increase in enrichment of the reload U FAs to 4.0 w/o 235U. Eight MOX FAs of that design were inserted in 1988. Based on the carrier material Unat, an average content of fissile material of 3.8 w/o Pu_{fiss} is used. In this case there is no need for the use of water rods.
A further design was made for a NPP with a 16 x 16 rod lattice, triggered by the change in carrier material from Unat to Utails with compensation of the lower ^{235}U content by a higher Pu_{fiss} content at the same time. The enrichment of U FAs has been remained unchanged in this case. A FA design with an averaged Pu_{fiss} content of 3.48 w/o (carrier material Utails with 0.25 w/o 235U) was realised. The mentioned 14 x 14 MOX FA and the 16 x 16 MOX FA are schematically shown in Fig. 3.

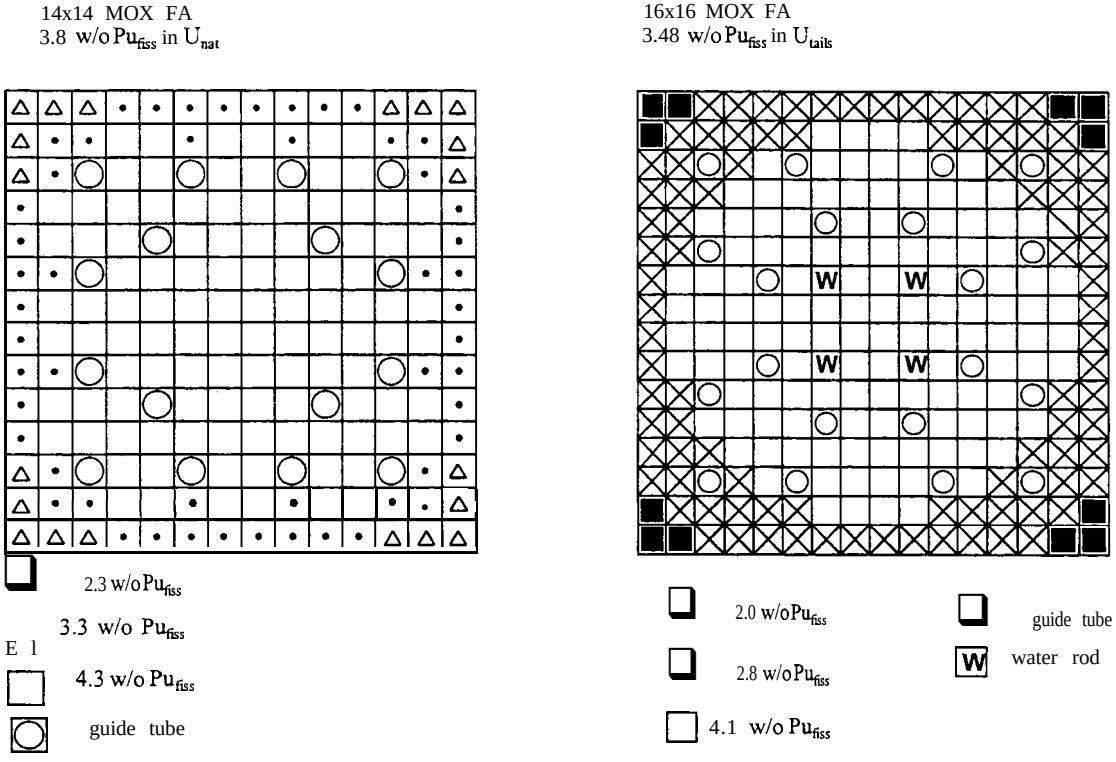

FIG. 3. *Different MOXFA designs for higher burnup and use of U_{tails} as carrier material*

Further MOX FA designs have been drawn up for other PWRs on the basis of U FA enrichments up to 4.3 w/o 235U and are shown in Fig. 4.
For the German 1300 MWe NPPs with 16 x 16 rod lattice, a MOX FA compatible with U FAs with an enrichment of 4.0 w/o 235U has been designed. With depleted Uranium (with 0.25 w/o ^{235}U content) as carrier material the averaged Pu_{fiss} content of the MOX FA is 4.2 w/o.
For the 1300 MWe NPPs with an 18 x 18 rod lattice, MOX FAs compatible to enrichments of U FAs of 4.0 w/o 235U have been designed. The neutronic design calculations lead to a burnup equivalent MOX FA design with an averaged Pu_{fiss} content of 4.6 w/o with the carrier material Utails(0.25 w/o ^{235}U).

4. I.2 *MOXFA designs compatible to enrichments of UFAs exceeding 4.0 w/o ^{235}U*

For 14 x 14 rod lattices of Westinghouse type reactors, a newly designed MOX FA using Utails with 0.25 to 0.30 w/o ^{235}U as carrier material and an averaged Pu_{fiss} content of 4.75 w/o fulfils the required burnup equivalence to U FAs with 4.25 w/o ^{235}U. Because of the instrumentation tube at the centre of the FA it is not necessary to increase the moderation by adding water rods.

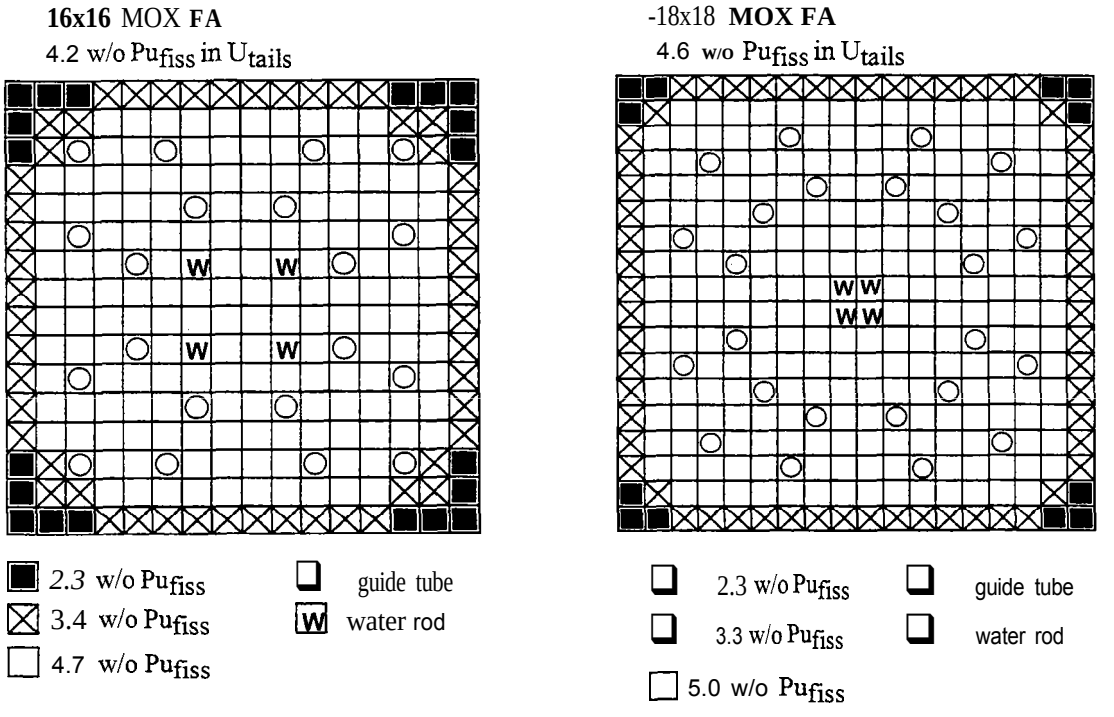

FIG. 4. Different MOX FA designs for higher burnup for 16x16 and 18x18 FAs

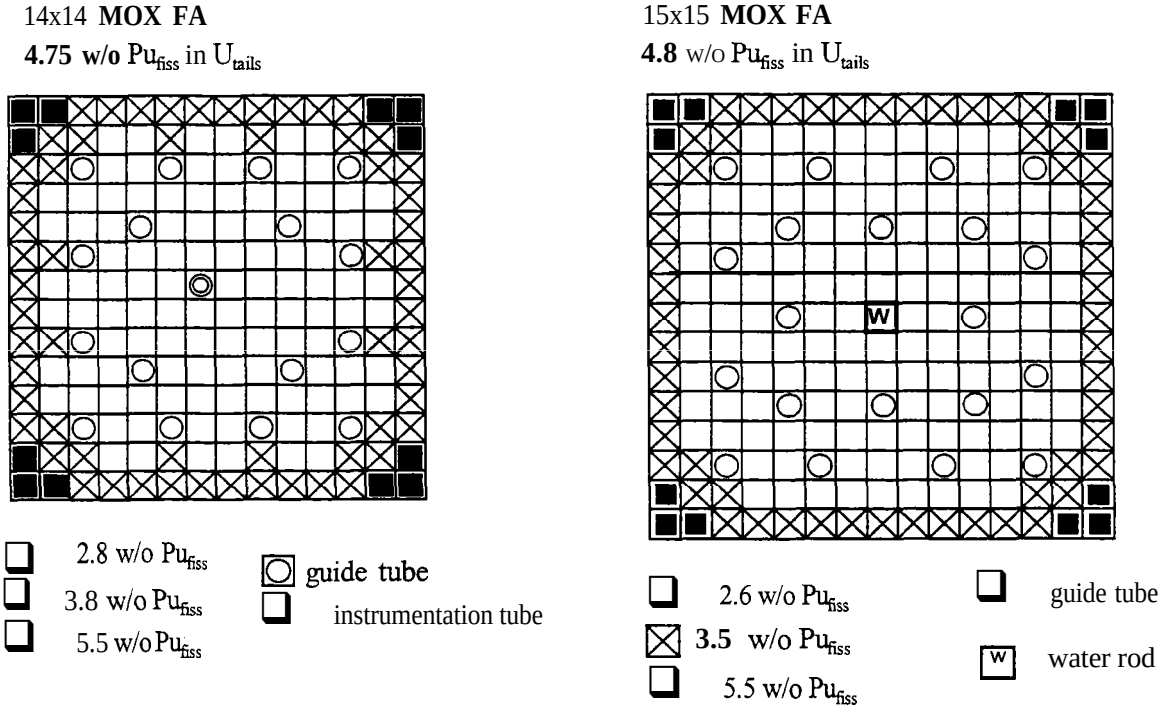

FIG. 5. MOX FA designs for higher burnup for 14x14 and 15x15 MOX FAs

For the 15 x 15 rod lattice type a MOX FA with the up to now highest content of Pu_{fiss} has been designed. The requirement of burnup equivalence to U FAs with 4.3 w/o ^{235}U can be fulfilled with an average content of 4.8 w/o Pu_{fiss}, using Utails with 0.25 w/o 235U as carrier material. In the 15 x 15 rod lattice FA one central water rod is sufficient to flatten the radial power distribution. The MOX FA designs mentioned above are shown in the Fig. 5.

4.1.3 MOX FA designs for B WRs

For thermal recycling of plutonium in BWRs MOX FAs of the 9 x 9 and 10 x 10 rod lattice type have been designed.
The MOX FA for the insertion in a BWR is in general much more complicated because of the much higher heterogeneity in comparison to PWR. The 9 x 9 -1 BWR MOX FA contains 6 different MOX fuel rod types and 1 additional Gd poisoned U fuel rod type to avoid power peaks around the water channel and to reduce the initial reactivity. The averaged Pu_{fiss} content is about 3 w/o with carrier material Utails (0.25 w/o 235U). As an example for the progress in design of BWR MOX FAs a design for a 10 x 10 rod lattice type (ATRIUM 10TM) has been performed.
The BWR 9 x 9 BWR MOX FA and the ATRIUM 10™ MOX FA are shown in the Fig. 6.

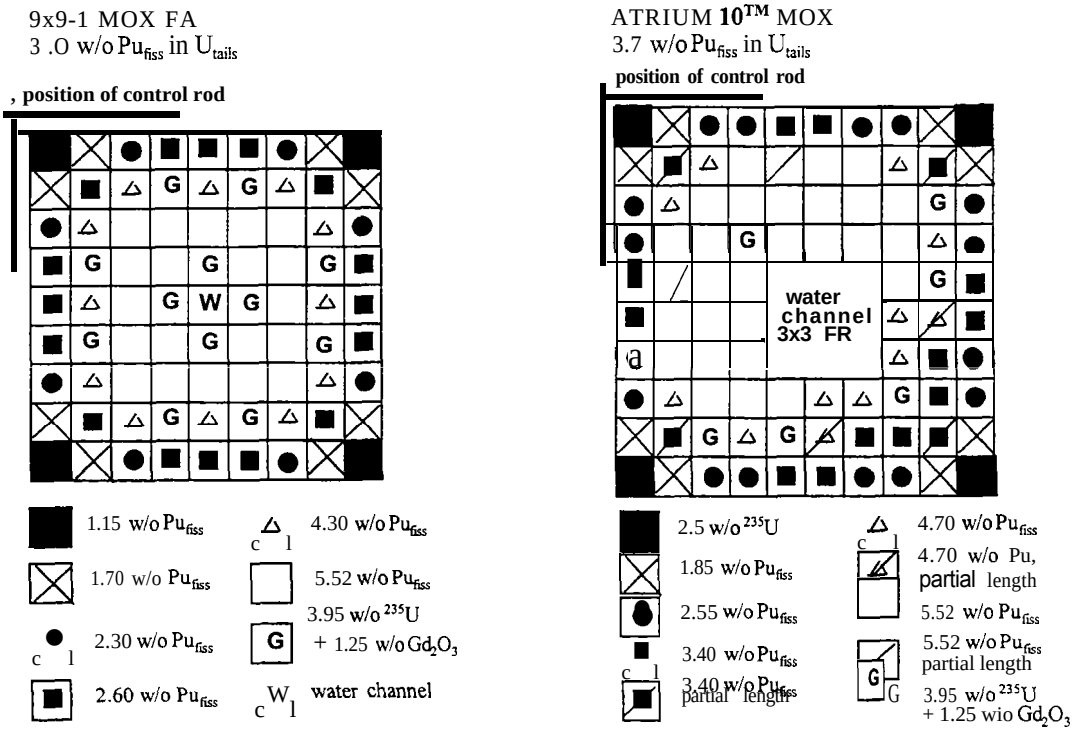

FIG. 6. Different MOX FA designs for B WRs (9x9 and 10x10 rod lattice type)

5. CORE PERFORMANCE WITH MOX FA IN PWR

An equilibrium core based on full low leakage loading with 88 MOX FAs (45 % of the core) of a 1300 MW_e NPP with 16 x 16 rod lattice type has been investigated in a licensing study. The equilibrium core has a reload batch of 20 MOX FAs and 24 U FAs (majority is Gd poisoned).
Based on this study a license was granted for reload of MOX FAs with 4.2 w/o Pu_{fiss} in Utails of Fig. 4.
The important cycle characteristics are listed in Table III. The coolant or moderator temperature coefficient (MTC) tends to more negative vaiues for increasing piutonium content in the core. The Doppler coefficient is hardly influenced by plutonium. This is of importance with respect to the shutdown margin.

The boron worth decreases with increasing number of MOX FAs in the core. The boron control system must therefore handle larger concentration differences during reactor operation. The critical boron concentration must be raised during loading operations to keep the reactor at a required level of subcriticality. The use of boric acid with enriched ^{10}B can increase the effective boron capacity.

TABLE III. U-MOX EQUILIBRIUM CYCLE (PWR WITH 1300 MW ELECTRICAL POWER)

MOX FA loading number / % of the reload batch (core)	20 / 45	
number of reload MOX and U FAs	20 / 24	**Effect compared with Uranium core**
MOX FA type	16x 16	
Pu_{fiss} content in w/o	4.2	
^{235}U content of carrier material	0.25	
^{235}U content of U FAs in w/o	4.0	
cycle length in efpd	315	**same**
MOX FA burnup in MWd/kgHM averaged discharge burnup maximum FA burnup	 54 60	 **about same**
initial boron concentration (without Xenon) in ppm	1775	**lower**
boron worth at BOC in pcmlppm	- 5.5	**lower (U core ~ 8.3)**
MTC at EOC in pcm/K	- 73	**higher (U core ~ 55)**
Net control rod worth at EOC in % Ap	5.6	**lower or same**

As regards the net control rod worth for stuck-rod configurations, the data depend more on the loading scheme than on the MOX FA fraction in the core. Thus MOX fractions of up to 50% without need for increasing the number and the effectivity of control rod system were found possible.
The primary operating results include information on cycle length, power distribution, reactivity coefficients, and control rod worth of cores containing MOX FAs. The reliability of the design methods is validated by measurements of these quantities. The neutron physics experience [7] is based on start-up measurements, in-service cycle monitoring and specific measurements required under licensing commitments.

6. CONCLUSIONS AND OUTLOOK

- It is shown that, under properly defined circumstances, increasing of initial enrichment and final burnup combined with thermal recycling lead to decreasing amounts of plutonium to be disposed of.
- MOX FAs can be adapted without problems to be compatible with the design and enrichment of new U FAs, actual plutonium qualities and different carrier materials.
- MOX FAs inserted together with U FAs of an initial uranium enrichment exceeding 4.3 w/o ^{235}U can be designed, as was demonstrated in design studies for a FA with 14 x 14 rod lattice type. With averaged Pu_{fiss} content of 5.84 w/o with the carrier material U_{tails}, the burnup equivalence (in the manner mentioned above) to U FAs with about 4.6 w/o ^{235}U is achieved. First drafts of a 17 x 17 MOX FA for the European Pressurised Water Reactor (EPR) result in a comparable content of fissile plutonium of ≈ 6.5 w/o.

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SAFETY AND LICENSING OF MOX VERSUS UO_2 FOR BWRS AND PWRS: ASPECTS APPLICABLE FOR CIVILIAN AND WEAPONS GRADE Pu

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Abstract

This paper reviews the safety and licensing differences between MOX and UO_2 BWR and PWR cores. MOX produced from the normal recycle route and from weapons grade material are considered.

Reload quantities of recycle MOX assemblies have been licensed and continue to operate safely in European LWRs . In general, the European MOX assemblies in a reload are <40% of all fresh assemblies, and operate on annual cycles to burnups <45,000 MWD/MTHM.

In the U.S., fuel vendors have undertaken studies to examine the feasibility of disposing of weapons grade plutonium in LWRs. These studies included review of the safety parameters of MOX assemblies versus UO_2. These studies indicated that no important technical or safety related issues have evolved from these studies.

The general specifications used by fuel vendors for recycled MOX fuel and core designs are as follows:

- MOX assemblies should be designed to minimize or eliminate local power peaking mismatches with co-resident and adjacently loaded UO_2 assemblies. Power peaking at the interfaces arises from different neutronic behavior between UO_2 and MOX assemblies.
- A MOX core (MOX and UO_2 or all-MOX assemblies) should provide cycle energy equivalent to that of an all-UO_2 core. This applies, in particular, to recycle MOX applications. An important consideration when burning weapons grade material is rapid disposition which may not necessarily allow for cycle energy equivalence
- The reactivity coefficients, kinetics data, power peaking, and the worth of shutdown systems with MOX fuel and cores must be such to meet the design criteria and fulfill requirements for safe reactor operation.

Both recycle and weapons grade plutonium are considered, and positive and negative impacts are given. The paper contrasts MOX versus UO_2 with respect to safety evaluations. The consequences of some transients/accidents are compared for both types of MOX and UO_2 fuel.

1.0 INTRODUCTION

The key to successful operation with MOX assemblies is in the neutronic design of the fuel assemblies and the core as it affects steady state characteristics like power distributions and shutdown margins, the core behavior under accident conditions, and fresh and spent fuel storage.

Table l-l presents typical isotopic compositions of recycle and weapons grade plutonium. The composition differences are sufficient to alter the operational behavior and safety analysis, primarily as related to temperature and void coefficients as well as on-site handling.

TABLE 1 TYPICAL ISOTOPIC COMPOSITIONS OF RECYCLE AND WEAPONS GRADE PLUTONIUM

	ISOTOPE, WT. %				
	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
Recycle Pu ("Reactor Grade")	1	59	24	11	5
Weapons Grade	<0.1	93	6	1	<0.1

2.0 NUCLEAR DESIGN CONSIDERATIONS

The neutronic behavior of mixed oxide fuel differs from that of UO_2 fuel designed for equivalent service on a number of points. These differences have an important effect on the nuclear and core design for cores with MOX fuel. They also have significant safety implications. The main points of difference are discussed in the following sections. In general, for most parameters, the differences are greater for recycled as opposed to weapons grade MOX. Table 2 summarizes the impact of both recycle and weapons grade MOX versus UO_2 on nuclear parameters.

2.1 REACTIVITY REDUCTION VERSUS BURNUP

The reactivity reduction with burnup is slower for MOX fuel than for UO_2 fuel (PWRs and BWRs). This applies particularly to recycled MOX. As a result recycled MOX assemblies would definitely have the following characteristics relative to equivalent UO_2 assemblies:

- Much flatter reactivity loss versus burnup, leading to
  - Less power sharing in MOX assemblies at low assembly burnups
  - More power sharing in MOX assemblies at higher assembly burnups

2.2 IMPORTANCE OF ABSORPTION CROSS-SECTIONS

The absorption cross-sections of the main plutonium isotopes are about twice as important as for U-235 in the thermai energy spectrum. As a consequence:

- Xenon poisoning is lower and the xenon transient induced power distribution oscillation hazards are diminished
- Control worths tend to be lower, and control clusters (PWR) or blades (BWR) tend to be less efficient
- The soluble boron worth is substantially lower

Again, the influence of recycled MOX on these items would be greater than weapons grade MOX.

TABLE 2 IMPACT OF MOX VERSUS UO_2 ON PARAMETERS

PARAMETER	ITEM AFFECTED	EFFECT OF MOX		IMPACT		COMMENT
		RECYCLE Pu	WEAPONS GRADE Pu	POSITIVE	NEGATIVE	
Reactivity Behavior with Burnup	Power Sharing MOX/UO_2	Flatter Depletion: ▪ less power fresh MOX low burnup ▪ more power high burnup	Behavior closer to UO_2	Higher local peaking at low burnup with no impact on margins	At higher burnup: ▪ more fission gas release ▪ more clad corrosion	Less impacts with weapons grade MOX
Absorption cross-section at Thermal Energies	1. Xenon Poisoning 2. Control rod worth 3. Soluble Boron worth 4. Assembly design	Higher cross-sections of plutonium give lower thermal flux harder spectrum and less efficient thermal region	Less of an impact due to the lower plutonium fissile loadings to compensate for Pu-240 neutron absorption	Lower, less oscillation hazard	Lower, less efficient fixed shim Lower, adversely affects cold shutdown safety, post LOCA behavior	On one-to-one basis weapons grade Pu core would behave more like UO_2 core than recycle Pu core regarding affected items
Epithermal Absorption Resonances	1. Moderator Temperature Coefficient 2. Doppler Coefficient 3. Void Coefficient	More numerous resonances in epithermal range and fission/capture cross-section resonances lower than at thermal	Some evidence of somewhat greater impact on coefficients	1,3 More negative: ▪ improved cycle stretch 2. More negative - better response to rod drop events	1,3.More negative: ▪ exacerbate over-cooling events, or pressurization transients ▪ increases hot-to-cold swing	On one-to-one basis coefficients somewhat more negative than equivalent recycled MOX core if weapons grade MOX employed

TABLE 2 (Continued) IMPACT OF MOX VERSUS UO_2 ON PARAMETERS

PARAMETER	ITEM AFFECTED	EFFECT OF MOX		IMPACT		COMMENT
		RECYCLE Pu	WEAPONS GRADE Pu	POSITIVE	NEGATIVE	
Delayed Neutron Fraction	Transient Power Increases	Smaller	Smaller than recycled Pu		Faster power/flux rises in uncontrolled rod withdrawal, overcooling events	Withdrawal event rise eased by lower rod worth, more negative Doppler
Decay of Pu^{241}	Fuel Reactivity (at longer than anticipated times between manufacturing and operation)	Important due to relatively high Pu^{241} content	Nil		Reactivity loss (less cycle energy) due to decay of fissionable Pu^{241} for recycle Pu cases	
Pressure Vessel Fluence	Vessel Damage Rate	Higher fast flux at low-moderate burnups	Similar		Accelerated PWR vessel damage if MOX assemblies are placed in critical peripheral locations	Not of particular concern for BWRs
Batch Isotopic Variability	Manufacturing Related Uncertainties	Relatively large variability batch to batch	Little or no variability batch to batch		Increased engineering uncertainties built into MOX fuel/core for recycle Pu cases	

2.3 MIXED MOX and UO_2 CORES

The difference in neutronic behavior between MOX and UO_2 assemblies, unless designed against, results in flux rises (high power peaking) in the plutonium fuel rods at the interfaces between uranium and plutonium PWR assemblies. Since MOX assemblies will reside next to UO_2 assemblies, flux rises that can occur in the plutonium rods at the assembly interfaces are minimized by plutonium enrichment shaping in PWR assemblies.

2.4 ABSORPTION CROSS-SECTIONS OF THE PLUTONIUM ISOTOPES

The absorption cross-sections of the plutonium isotopes are characterized by absorption resonances more numerous and much higher in the epithermal energy range (0.3 to 1.5 eV) than those of uranium isotopes. The ratio of fission to capture cross-sections of these resonances is generally lower than at thermal energies. For MOX cores, this leads, in general, to moderator, void, and fuel temperatures (Doppler) coefficients which are more negative than for equivalent UO_2 fueled cores.

There is some evidence that on a one-to-one basis, the net effect of the use of weapons grade plutonium as opposed to recycle plutonium would be to make feedback coefficients somewhat more negative. This is more than likely a consequence of the different isotopic compositions as shown in Table l-l and different spectral hardening for the same fuel cycle service between recycle and weapons grade MOX.

The more negative coefficients together with the reduced boron worth tend to exacerbate core over-cooling transients in PWRs or core overcooling and pressurization transients in a BWR.

2.5 DELAYED NEUTRON FRACTION

The delayed neutron fraction is smaller for ^{239}Pu (0.22%) than for ^{235}U (0.67%), thus a MOX core will react faster in a prompt critical event than a core fuelled with UO_2 assemblies. Accident kinetics in uncontrolled rod withdrawal or excessive cooling situations will differ, and flux increase will be faster. Since ^{241}Pu has a delayed neutron fraction close to ^{235}U, as MOX fuel burns the aforementioned impact will become closer to that of an all-UO_2 core.

2.6 DECAY OF PU-241 INTO AM-241

Normally the effect of the decay of ^{241}Pu into ^{241}Am on reactivity over the period of time between manufacture and reactor startup is correctly accounted for by the reload core designer. If the MOX fuel does not startup on or near the intended date, it would have an adverse effect on core reactivity one the MOX fuel irradiation does start. This is a consequence of the buildup of the absorbing isotope ^{241}Am. This issue would not arise with weapons grade derived MOX.

Taking all of the above into consideration, the MOX fuel vendors have been providing MOX fuel and core designs which, at most, have required only minor changes in equipment and systems.

3.0 SAFETY CONSIDERATIONS OF MOX VERSUS UO_2

The thermal hydraulic design for a MOX core is unchanged from a UO_2 core. However, the differences in the nuclear parameters as outlined above have very significant effects on the safety evaluations. Table 3 summarizes some safety information for PWRs.

3.1 SEVERE TRANSIENT EVENTS

The most severe transients for the PWR cores are breaks in the secondary side and rod ejection. Unscheduled opening of a secondary valve (safety valve, steam generator relief valve, or

turbine by-pass steam line valve) is a category 2 event while a double ended main steam line break is a category 4 event. Both events are within the design basis core overcooling transients.

3.1.1 BREAKS IN THE SECONDARY SIDE

The rupture of a steamline induces a secondary side blowdown that intensifies heat exchange with the primary system and lowers the primary temperature and pressure. The rise in core moderator density increases reactivity because of the negative moderator coefficient. The negative Doppler

TABLE 3 PWR REACTOR SAFETY EVALUATION COMPARISON OF MOX AND UO_2

EVENT	SOURCE	EFFECT OF MOX IN CORES
Overcooling Transients (Breaks in Secondary Side) 1. Category 2 Secondary Valve opening	EdF	– Margins to recriticality lower – No recriticality for 30% MOX core – Major impact with MX is lower boron worth
2. Category 4 Double Ended Main Steam Line Break (Consequences may be less severe with weapons grade Pu due to more negative Doppier Coefficient)		– Prompt critical in 30% MOX core (less favorable kinetics) – More severe power rise (peak <10 % rated power) – DNBR below dryout conditions – No system changes required
		– Recriticality does not occur for envelope of MOX cores
	Siemens	– Margin less than for equivalent UO_2 cores No system changes required – Problems with 16 x, 100% MOX, and 18x, 50% MOX – recriticality can occur – larger tanks of enriched b may be needed (avoid dryout)
Rod Ejection Accident (Consequences may be less severe with weapons grade Pu and higher fuel burnups)	EdF	– Margin to limits lower with 30% MOX core – iess favorabie kinetics overshadows iower rod worth – Event terminated within limits (only modest fuel burnup)
	CABRI Test	– MOX rodlet at ~ 100 cal/gram failed violently at 50-55 GWD/MTHM. Test may not be representative.
LOCA (Breaks in Primary System)	EdF	– Minor changes for 30% MOX core case: – initial boron reserve increased from 2000 to 2400 ppm – minimum concentration in sump water raised from 1300-1500 ppm – instruments and control of steam generator atmospheric relief system duplicated and emergency supplied
	Siemens	– German LOCA requirement <10% rods fail in LOCA – For MOX, failure threshold IO-20 watt/cm lower than UO_2 due to higher fission gas release (at modest fuel burnups)

coefficient also contributes to the reactivity increase. If the most reactive control rod cluster is stuck at its fully withdrawn position, criticality could occur. The worst condition occurs at end-of-cycle where the moderator coefficient is most negative, and the boron worth is smallest.

For the category 2 event, inadvertent secondary valve opening, the licensing criterion is to preserve the clad integrity. In effect, in France, this criterion imposes a constraint that the core remain subcritical, during and after all automatic safety systems have been exhausted. This is generally achieved by continual injection of boron.

For the category 4 accident, double ended main steamline break, the rapid increase in reactivity induces a power excursion, limited mainly by Doppler feedback, and also by the boron safety injection. The licensing criterion in category 4 involves a release limit. However, for the steamline break, clad integrity must be assured because of the containment bypass. This imposes a design constraint that dryout shall not occur.

3.1.2 ROD EJECTION ACCIDENT

The ejection of a control cluster in a PWR can only occur in the event of a control rod mechanism pressure housing rupture, which exerts a violent upward pressure, due to the primary pressure, on the drive shaft. Rod ejection is a rapid reactivity insertion event, producing a sudden power escalation in the primary system, accompanied by severe power distribution distortion.

For a given core design, the key parameters that control this event are control assembly worth, negative magnitude of the Doppler coefficient, and kinetics parameters (delayed neutron fraction, prompt neutron lifetime). The former two are generally more favorable for a MOX core, whereas the latter one is less favorable.

The local power build-up in this event is such that it can cause Departure from Nucleate Boiling (DNB) and a marked increase of the energy contained in the fuel with attendant risks of clad and pellet damage.

EDF calculations of this event indicate that a 30% MOX core relative to an all UO_2 core entails a reduction in margins to limits, since the less favorable kinetics parameters of the MOX core control. Nevertheless, EDF notes that the event in the MOX core has passed licensing scrutiny since:

- The calculated energy stored at the hot spot remains below the limit value
- The clad temperature is below the embrittlement level (does not exceed 17% oxidized thickness), and
- The peak center-line temperature of the hottest pellet does not reach mixed oxide melting temperature.

3.1.3 LOSS OF COOLANT ACCIDENT (LOCA)

French LOCA studies have shown that for certain types of piping breaks, borated water would be injected into the core and steam removed. Such a process would ultimately lead to a high boron concentration in the core and a low concentration in the sump water. The minimum boron concentration in the sump water was raised from 1300 ppm for an all UO_2 core to 1500 ppm for a 30% MOX core to compensate for the lower boron worth with the latter.

From a practical standpoint, in the event of a LOCA, the operator is required to ensure safety injection switchover at regular intervals so that the top and bottom of the core are successively treated. The purpose of this is to achieve a more homogeneous distribution of the boron concentration. In order to avoid overfrequent switchovers, the initial boron reserve content has been increased from 2,000 ppm to 2,400 ppm. EDF notes that with the indicated modifications, the post-LOCA behavior of a 30% MOX core is similar to that of a UO_2 core.

3.1.4 MAIN STEAM LINE BREAK IN A GERMAN PWR

The main steam line break is identified as the limiting transient for Siemens MOX cores. The worst scenario for this event is for it to occur at the end-of-cycle where the moderator coefficient is highest by about a factor of two from beginning-of-cycle. Siemens calculations show that recriticality does not occur in the current envelope of MOX cores analyzed, even at end-of-cycle, although the margins are less than for equivalent UO_2 cores.

Siemens indicated that for MOX cores with higher assembly loadings, e g., 16x16 designs with 100% MOX or 18x18 KONVOI designs with 50% MOX assemblies, the steam line break event could present a problem. Initial analyses show that with the very high negative moderator temperature coefficient in these cores, recriticality can occur. Although this may be the case, the Siemens calculations indicated no large power rises even with current boron systems.

3.2 FUEL STORAGE POOL CONSIDERATIONS

In Germany, the safety of spent fuel storage requires reanalysis when using MOX fuel. The reasons for this are the higher reactivity level of spent MOX assemblies relative to UO_2 assemblies at the same burnup, and the fact that the decay heat from MOX assemblies is higher after some days from discharge.

4.0 BWR SAFETY

As with the PWR, with MOX cores and for equivalent fuel designs BWRs have moderator and Doppler coefficients that are more negative than an all UO_2 core. The void coefficient is also more negative and the delayed neutron fraction is smaller. Table 4 summarizes the BWR safety information.

A transient analysis performed for KKIsar-1 with a 54% MOX core indicates that the DOPPLER coefficient controls the events producing prompt criticality. The limiting transient for the 54% MOX core is the same as for an all UO_2 core, i.e., the core pressurization event caused by a turbine trip without by-pass, with scram on neutron flux level (first scram on valve position is bypassed).

The turbine trip without bypass transient determines the magnitude of the MCPR at higher power levels. This transient was analyzed for Gundremmingen with 9x9 MOX assemblies (38% of the core) and resulted in an MCPR operating limit of 1.3 1 – 1.32 versus 1.30 for an all UO_2 core. Thus there would be a minimal loss of operating flexibility.

4.1 BWR LOCA

In German internal pump plants, and particularly with 9x9 fuels, LOCA (MAPLHGR) is not limiting. Nevertheless with MOX in the core, the accident could require a change in boron concentration in the auxiliary shutdown system used in the event of a LOCA.

Siemens' evaluation of the KKIsar-1 equilibrium core (54% ATRIUM™ 9 MOX) showed that the worth of the boron in the auxiliary shutdown system decreases by 23% for MOX cores versus all UO_2 cores. This, in effect requires an increase in boron concentration from 600 to 900 ppm to maintain the same post-LOCA behavior as an all UO_2 core.

It has been calculated that the residual heat from MOX and UO_2 cores is approximately the same. Therefore, existing residual heat removal systems would be adequate in the event of a LOCA.

ABB Atom has performed licensing calculations for a core containing 50% of their SVEA-96 UO_2 and 50% Siemens MOX assemblies. The fundamental result of the ABB Atom analysis was that

TABLE 4 BWR REACTOR SAFETY EVALUATION COMPARISON OF MOX AND UO_2

Event	Source	Effect of MOX in Cores
Core Pressurization Transient (Turbine trip without bypass)	Siemens	- Scram on neutron flux (valve position scram bypassed) - MCPR margin same or slightly less than in UO_2 core for 3 8-54% MOX cores with 9x9-l and ATRIUM 9 - MCPR margin reduced slightly with 100% weapons grade MOX core (l-2%), but adequate margin available
	GE	
Rod Drop Event	Siemens	- Deposited energy well below limit for fuel (4 15 Kjoules for 54% MOX ATRIUM 9 core versus 711 Kjoules allowable - Large margin to limit for weapons grade plutonium
	GE	
ATWS	Siemens	- Not limiting
	GE	- Response of 100% MOX core about same as UO_2 core with weapons grade plutonium
LOCA	Siemens	- MAPHLGR not limiting in German internal pump plants, particularly with 9x9 fuels - Boron concentration in auxiliary shutdown systems needs to be increased in order to maintain same post LOCA behavior as with all-UO_2 core - use of enriched Boron is an option - Residual heat removal systems are adequate
	GE	- Radiological effect of LOCA appears to favor cores with MOX - 100% weapons grade MX core will not degrade LOCA consequences of all-UO_2 core - Decay heat on shutdown with MOX lower than with UO_2 (lower fission product inventory)
Stability	Siemens	- Theoretically MOX cores should be less stable due to more negative feedback - Analyses showed no important differences – rod pattern changes can have a greater effect
	GE	- Less stable with 100% weapons grade MOX - Slight increase in the exclusion zone - Some probably minor impact on long-term solutions for U. S. BWRs
Handling damage to an Irradiated Assembly	**Siemens**	- Little difference in dose equivalent exposures between MOX and UO_2 cores at 37 GWD/MTHM

the core could operate with no restrictions. They found, however, as did Siemens, that the boron concentration in the auxiliary shutdown system would have to be increased.

4.2 STABILITY

Because feedback mechanisms tend to be more negative with MOX fuel, it would appear that core stability behavior should be somewhat poorer in a MOX containing core, as compared with an all UO_2 core of the same basic fuel design.

Siemens has analyzed the ATRIUM™ 9 core in KKIsar-1 and the calculations show no important differences in stability behavior between MOX and all UO_2 cores. For the 9x9-1 fuel in Gundremmingen Siemens' investigation found that the impact on stability relative to an all UO_2 core

is small and that control rod pattern changes have a greater effect on stability than changing from UO_2 to MOX.

4.3 TRANSIENT ANALYSIS -PLANT OPERATING LIMITS US BWRs

One purpose of the transient response analysis is to set plant operating limits to avoid the possibility of departure from nucleate boiling (DNB) for events expected to occur during the plant lifetime. For BWRs this has been measured by the Minimum Critical Power Ratio (MCPR), which is the ratio of the bundle power at which DNB is expected to occur on the hottest rod in a fuel bundle to the bundle's current power level. With departure from nucleate boiling MCPR at 1 .0, statistical considerations are added which account for manufacturing and measurement tolerances, including uncertainties in the thermal-hydraulic correlations developed by test programs, to determine the Safety Limit MCPR (SLMCPR).

For BWRs the SLMCPR has been established to be 1.07. It is defined such that if the most limiting fuel bundle in the core is calculated to have an MCPR of 1.07, 99.9% of the fuel rods in the core will not experience DNB. This is the acceptance criterion approved by the U.S, NRC.

Those transients identified which can occur due to a single equipment failure or single operator error are analyzed to determine the most limiting transient in terms of the change of critical power ratio which can occur. This ACPR is added to the SLMCPR to determine the Operating Limit MCPR (OLMCPR). The core is designed and operated such that adequate MCPR operating margin exists.

One of the more limiting transients for BWR cores is the Load Rejection with No Bypass (LRNBP) event. Even though this event involves multiple failures, a long-standing GE-USNRC agreement exists to consider this an initiating event as one of those in Regulatory Guide 1.70, Chapter 15. The primary nuclear dynamic parameter influencing this transient, other than scram reactivity, is the void coefficient. Collapse of voids due to primary system pressurization results in a power increase.

Since the MOX fuel design has a larger negative dynamic void coefficient than the reference design, a larger ACPR is predicted due to the void collapse associated with the transient.

4.3.1 STABILITY

The more negative void coefficient with the full MOX core adversely impacts the core stability. As an illustration, the calculated decay ratios at natural circulation and rated rod line are 0.69 and 1 .01 for UO_2 and MOX cores respectively.

Utility requirements for interim corrective actions (ICAs) to protect against reactor instabilities include an exclusion zone bounded by the 45% flow and the 80% load line. Some plants have even more restrictive ICAs.

Protection from reactor instabilities is provided in the BWR by control which excludes a zone of pump speed/flow operation on the reactor power/flow map. For the MOX core evaluated, it would be necessary to increase this zone slightly beyond that specified for a UO_2 core.

4.3.2 ANTICIPATED TRANSIENT WITHOUT SCRAM (ATWS)

ATWS events are a special class of transients also analyzed in BWR safety analyses. Because of their low probability of occurrence, acceptance criteria for ATWS events are somewhat different than those for normal transients. These include:

- Core coolability
- Peak nuclear boiler pressure below ASME service Level C (1,500 psig)
- Containment parameters below limiting conditions.

According GE, the response of a BWR with MOX to an ATWS event is not expected to change. If an isolation occurs, the increased void coefficient can lead to a slightly higher power spike, however, the remaining heat discharged to the suppression pool is a function of the makeup injection rate and is unaffected by the void coefficient.

4.3.3 OVERPRESSURE ANALYSIS

In BWRs the number of safety/relief valves is normally set by requirements of the ASME Code for overpressure protection. This analysis is a special transient in which the Main Steam Isolation Valves are suddenly closed, and the normal main Steam Isolation Valve position scram is assumed to fail, which results in a reactor scram due to high neutron flux. In addition no credit is given for safety/relief valves opening in the power actuated relief mode. Credit is given only for the spring mode. The acceptance criterion for this analysis is a peak vessel pressure of less than 1,375 psig (9.48 mega-pascals).

For the full MOX core design the peak vessel pressure attained during the Main Steam Isolation Valve Failure transient is 8.80 mega-pascals, which is comparable to the results from the UO_2 reference core design.

4.3.4 ACCIDENT ANALYSIS

Reactivity accidents, such as rod drop or rod ejection (dominated by the fuel Doppler coefficient and control rod worth) are evaluated in BWR safety analyses. Experience with these calculations has shown that they tend to be insignificant when compared with the heat deposition limits associated with the event. Despite the smaller beta of MOX fuel, rod worth values are significantly lower than an equivalent uranium core. Thus, the severity of this event is reduced in comparison with the UO_2 based analyses contained in the safety analysis. Therefore, rod drop accidents are not of concern.

For LOCAs, nuclear dynamic feedback plays an insignificant role, since the plant is presumed to scram at the start of the event. The only other core design dependent input is the decay heat level for the fuel after the scram. The decay heat level for the MOX core is expected to be slightly less than for a UO_2 core. In any case, the BWR LOCA analysis is dominated by the inventory loss during the initial blowdown, and the results for a UO_2 core show an acceptable response. Thus, it is concluded that a MOX core will not degrade the LOCA results.

5.0 CONCLUSION

Within the framework of current operations with MOX fuel in Europe, the consequences of limiting transients, while somewhat more severe with recycled MOX, are still acceptable with no system changes required. Rod ejection has adequate margin to current limits with MOX. This may become an issue if limits are reduced based on Reactivity Insertion Accident experiments, particularly at higher burnups. The consequences of some transients/accidents may be less severe with the weapons grade plutonium because of a more negative Doppler coefficient (relative to recycle plutonium).

There are no real obstacles to safety or licensing when MOX fuel, whether derived from recycled commercial power plant fuel or weapons grade plutonium, is prudently designed and deployed. In certain cases reactivity control systems may require some modification, but both PWRs and BWRs are capable of using MOX fuels.

WEAPONS GRADE PLUTONIUM DISPOSITION IN PWR, CANDU AND FR

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Abstract

In the frame work of the *AIDA/MOX phase I /I/* program (1994-1997) between France and Russia, the disposition of plutonium in reactors was studied.

The LWR (Light Water Reactor), FR (Fast reactors), CANDU (Heavy Water Reactors), HTR (High Temperature Reactors) options for using excess dismantled weapons plutonium for peaceful commercial nuclear power generating purposes offer some advantages over the remaining options (storage).

The *AIDA/MOX phase 1* program covers different topics, among which are the neutronic aspects of loading reactors with weapon-grade plutonium.

The conclusions are that the weapon plutonium consumption is similar in the different type of reactors. Only, the use of inert matrices allows to increase the mass balance for a same denaturing level.

The use of Thorium as a matrix or special isotopes to increase the proliferation resistance prove to be insufficient.

1. INTRODUCTION

In the frame work of the *AIDA/MOX phase I* program (1994-1997) between France and Russia, the disposition of plutonium in reactors was studied.

The LWR (Light Water Reactor), FR (Fast reactors), CANDU (Heavy Water Reactors), HTR (High Temperature Reactors) options for using excess dismantled weapons plutonium for peaceful commercial nuclear power generating purposes offer some advantages over the remaining options (storage) :

-fission of a significant fraction of the initial weapons plutonium (at least 30%),
- high radioactivity of the final product,
- isotopic denaturing of the residual plutonium,
- conservation of natural ressources : 50 metric tons of WG-Pu are able to produce 350 TWHe (one year production for France),
- no additional nuclear wastes.

The *AIDA/MOX phase I* program covers different areas, among which are the neutronic topics of loading reactors with weapon-grade plutonium.

This paper will present the studies applied to PWR and FR in order to improve the degradation of plutonium during irradiation and will compare different core concepts in term of denaturing and mass balances.

The first part (§ 2,3 and 4) will present a summary of sensitivity studies performed for each reactors.

The second part (§ 4) is an approach for an optimisation : how to improve the reprocessing.

The last part (§ 5) is a comparison of different reactors.

2. THEORETICAL BACKGROUND

The plutonium inventory change in a MOX fuel results from a production term, by captures on ^{238}U, and from a consumption by fissions on (fissile) Pu isotopes and captures on Pu isotopes. The consumption is improved by reducing the source term (^{238}U) or increasing the reaction rate (flux, and/or cross sections). Due to large flux levels, fast reactors could be favourable, but due to large cross sections, a thermal neutron spectrum could be also favourable. Thus, we analyse the plutonium consumption in different kinds of reactors : standard PWR, CANDU and FR and with different matrices for plutonium.

Also, we may improve the proliferation resistance by using highemitters of α particles , y or neutrons. For example, in the depletion chain of heavy nuclides for a MOX fuel, the ^{238}Pu, ^{236}Pu are α emitters, the ^{242}Pu is a poison. Then, in order to enhance the resistance, we add ^{237}Np or ^{241}Am, or use a support of reprocessed uranium.

The calculations were performed with the code system APOLL02-DARWIN-CRONOS2 for the PWR cores, ERANOS,-DARWIN for FR reactors and TRIPOLI 4 for the Monte-Carlo route, all based on data from JEF2 libraries.

3. FR CAPABILITY EVALUATION

Sensitivities studies were performed firstly in order to assess weapon Pu burning rates and isotopic degradation in fast reactors.

The parameters were (1) the size of the core (from 250 MWe to 1500 MWe), (2) the fertile support for plutonium (either uranium or thorium), (3) the plutonium content in the fuel (including Pu burner designs with large Pu contents), (4) and some addition elements to plutonium, intended to enhance the radiation or heat release of spent fuel plutonium, and thus proliferation resistance, as it is mentioned in the theoretical background.

Reactivity coefficients, isotopic degradation, and mass balances were also assessed.

3.1. Uranium /weapon Pu fuels

The plutonium burning is assessed for three reactor types: a small one, like the Phénix reactor (250 MWe), a large one, like the Super-Phtnix reactor (1200 MWe), and fmally a large plutonium burner (1500 MWe, Pu content increased by reducing the fuel volume fraction in the core, according to CAPRA design recommendations).

Compared to a loading with reactor-grade plutonium, the plutonium content is reduced because of the higher plutonium quality. Hence, the plutonium consumption rate is reduced. The sodium void effect is smaller (smaller content in plutonium even isotopes), and the Doppler constant is stable. Linear pin ratings are also almost the same.

In order to obtain a $^{240}Pu/Pu$ content around 20% at the end of the fuel residence time, the burn-up required ranges between 11 and 14% (see table 1).

Table 1 : $^{240}Pu/Pu$ content vs burnup and irradiation time in $(U,Pu)O_2$ fast reactors

$^{240}Pu/Pu$	Small size (250 MWe)	Large size (1200 MWe)	Large Pu Burner (1500 MWe)
15 %	7.6 at% or 570 efpd	6.7 at% or 750 efpd	9.6 at% or 560 efpd
20 %	12.3 at% or 950 efpd	10.8 at% or 1250 efpd	14.2 at% or 800 efpd

The shorter residence times being for the higher Pu contents in the fuel (less conversion from ^{238}U to ^{239}Pu).

The overall plutonium consumption is also directly linked to the initial plutonium content, as shown in table 2 (for a weapon-grade Pu loading). A yearly weapon-grade plutonium consumption of ≈400 kg/year (load factor = 0.8) is achievable in a large Pu burner.

3.2. Thorium /weapon Pu fuels.

This study has been performed on a small core design (250 MWe, like the Phénix reactor), by changing the $(U,Pu)O_2$ fuel to $(Th,Pu)O_2$ with weapon-grade plutonium.

Table 2 : Plutonium balance (EOL-BOL) vs core size and desig fast reactors

	Small size	Large size	Large Pu Burner
Initial Pu content (w%)	17	14	28
Fuel residence time (efpd)	540	900	855
Pu balance (kg/year)	-27	-10	-400

The average Pu/(Th+Pu) content in the core is 21 w%. With respect to the reference core, the sodium void reactivity is greatly reduced, to less than 1$, for a voiding of the fuel zones. This is due to the favourable neutronic characteristics of ^{232}Th (less fissile at high energies than ^{238}U) and ^{233}U (smaller increase of neutron production for high energies than ^{239}Pu). The Doppler constant is almost the same, as well as the delayed neutron fraction .

The overall plutonium consumption is 82 kg/TWhe (i.e. 140 kg/year), far from the maximum of ≈110 kg/TWhe because a significant part of the ^{233}U produced contributes to the fissions.

The degradation of the plutonium isotopic composition remains low, despite the absence of internal ^{239}Pu breeding: a 12% content in ^{240}Pu/Pu is reached at end of life conditions (540 efpd), corresponding to a burnup of 5.9 at%.

Furthermore, the amount of uranium produced is 45 kg/TWhe (i.e. 80 kg/year), this uranium being itself a good weapon material (≈96% ^{233}U/U). However, it includes small amounts of proliferation resistant ^{232}U (hard gamma emitters).

3.3. *Other elements mixed to plutonium.*

The basic idea is to make the spent fuel plutonium more resistant to proliferation by increasing its specific decay heat and neutron source.

The most active plutonium isotope is ^{238}Pu, with a large decay heat and neutron source (see table 3).

Table 3 : Specific decay heat and neutron source of plutonium isotopes

	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
Specific decay heat (W/kg, a + β)	567	2	7	3	0.1
Neutron source (n/g/s, sp. fission)	2.6 10^5	3	8.7 10^4	—	1.8 10^5

Three solutions have been investigated: addition of ^{237}Np or ^{241}Am to the fuel, or utilisation of reprocessed uranium as a fertile support to plutonium (with ^{236}U, which yields ^{237}Np by capture).

The use of reprocessed uranium is the less efficient solution, due to the low ^{236}U content (^{236}U/U = 0.5%), and to the indirect chain connecting ^{236}U to ^{238}Pu (two successive neutron captures are required).

Similarly, the addition of ^{237}Np is more efficient than the addition of ^{241}Am to the fuel. However, addition of an absorbing element such as ^{237}Np or ^{241}Am to the fuel leads to an increase of the sodium void reactivity and to decrease the Doppler constant.

3.4. Summary and conclusions for fut reactors.

A single irradiation of weapon-grade plutonium in a fast reactor does not denature much the isotopic composition of the plutonium. Nevertheless, large-size plutonium burners can burn significant quantities of plutonium.

The use of thorium as a fertile support to plutonium does not increase significantly the degradation of weapon-grade plutonium, but oniy its consumption.

The use of special elements to improve the proliferation resistance at reprocessing is not efficient enough.

4. PWR CAPABILITY EVALUATION

The parameters investigated were the moderation ratio, the fuel type (with or without U, use of Np, Am, U), the irradiation time, the reactivity coefficients, the degradation kinetics of plutonium, and the mass balances.

4.1. *(U,Pu)O_2 fuels*

First, we compare reactor grade and weapon grade plutonium irradiated in the form of standard MOX fuel in a PWR for a same fuel management. Table 5 gives a comparison of the main reactivity coefficients.

Table 4 : Reactivity coefficients, (U,Pu)O_2 fuels in standard P WRs

	WG	RG
Initial Pu content (w%)	7.4	4
Boron efficiency (0 MWd/tHM) (10^{-5}/ppm)	-3.3	-4.8
Boron efficiency (45 MWd/tHM) (10^{-5}/ppm)	-3.6	-5.5
Global void defect (%)	-33	-53
Doppler coefficient (10^{-5}/°C)	-3.2	-3.1

The main impact is on the initial plutonium content and on the initial fissile plutonium content.

At the end of the fuel life, the weapon grade plutonium consumption is about 35% of the initial plutonium, compared to 26% for a civil grade. The mass balances are given in table 6.

Table 5 : mass balances, (U,Pu)O_2 fuels in standard P WRs

	civil grade	weapon grade
Mass balance Pu (Kg)	-2100	-1540
Mass balance (Kg/TWhe)	-51	-38

The plutonium consumption is directly linked to the initial plutonium content . The plutonium degradation vs irradiation tune is shown in figure 1 for a standard PWR. A burn-up of -15 GWd/tHM is needed to reach -20% of ^{240}Pu in the plutonium.

In order to enhance the plutonium degradation and proliferation resistance, neptunium was added to the fuel, and also americium, or uranium provided by the reprocessing of irradiated uranium. The heat emission is strongly increased, by a factor 5 to 10 depending on the initial Neptunium content. The neutron emission, partly to ^{238}Pu, is increased by 25%.

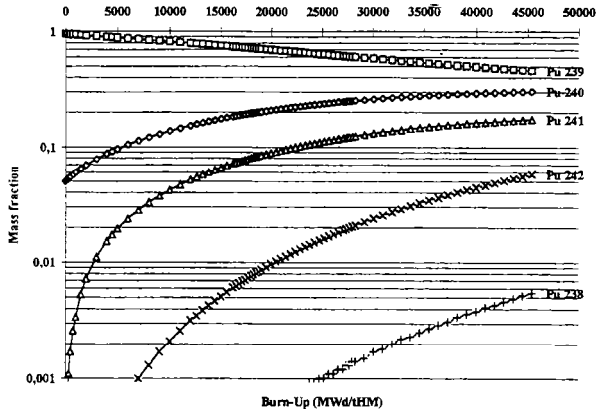


Figure 1 :

The impact of an addition of americium instead of neptunium is lower than for neptunium insertion in standard MOX fuel.

4.2. *Fuels without uranium*

Another way to improve the plutonium consumption and degradation is to use an inert matrix support or a thorium support. This cuts the production term by ^{238}U.

The evaluation of reactivity coefficients is summarised in table 9.

Table 6 : Pu without U fuels in PWRs : reactivity coefficients

	Inert matrix	Thorium matrix
Initial Pu content (g/cm3)	0.5	0.6
Boron efficiency (0 efpd) (10^{-5}/ppm)	-4.1	-3.5
Boron efficiency (1200 efpd) (10^{-5}/ppm)	-13	-5.1
Global void defect (95 %) (%)	-3	-30
Doppler coefficient (0 efpd) (10^{-5}/°C)	-1.2	-3.3
Doppler coefficient (1200 efpd) (10^{-5}/°C)	-0.6	-3.0

Using thorium allows to improve these reactivity coefficients, the weak points of such fuels without uranium being the void.

The plutonium degradation is important. Table 10 shows the plutonium degradation during irradiation.

Table 7 : Pu without U fuels in PWRs : Pu degradation

		Inert matrix		thorium matrix		
Time efpd	100	*300*	*600*	100	*300*	*600*
$^{239}Pu/Pu$ (%)	*84*	*65*	*33*	*90*	*82*	*67*
$^{240}Pu/Pu$ (%)	*12*	*23*	*39*	*8*	*14*	*24*
$^{241}Pu/Pu$ (%)	*3*	11	21	2	4	8
$^{242}Pu/Pu$ (%)	1	1	7	0	0	1

In 300 efpd, the ^{240}Pu fraction reaches more than 20%. This is an efficient way to enhance the degradation in short irradiation time but not the resistance to reprocessing, due to the small ^{238}Pu content.

The overall plutonium composition is very important, reaching some 110 kg/TWhe in the inert matrix case.

4.3. *Summary and conclusions for thermal reactors.*

The plutonium degradation (^{240}Pu = 20%) is reached after 20 GWd/tHM for a consumption of about 1.8 metric tons.

The reactivity coefficients for a MOX fuel using weapon plutonium are in the usual range for the standard UOX and MOX fue1s.

The Pu w/o U fuel allows to improve the consumption but they need R&D.

The use of added elements to improve reprocessing resistance is not efficient enough.

4. CANDU CAPABILITY EVALUATION

CANDU reactors presents a heterogeneous structure : the coolant and the moderator are different zones. Thus, the neutron spectrum (Figure 2) is thermal, heterogeneous allowing a large fission microscopic cross-section of ^{239}Pu (Table 8).

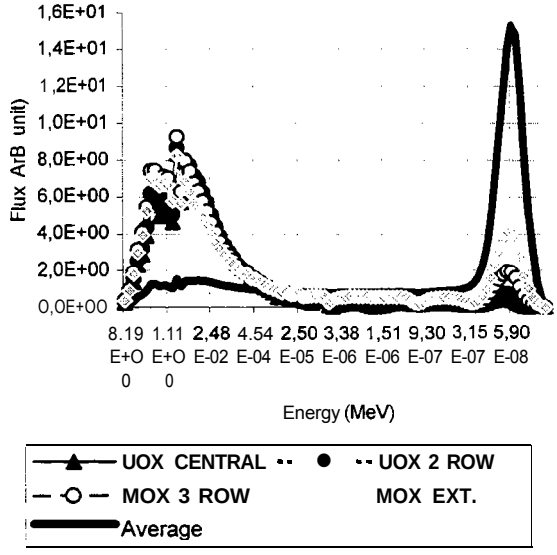


Figure 2 : Neutron spectrum in CANDU

Table 8 : ^{239}Pu fission cross-sections

Fast Reactor		PWR		CANDU
Weapon Grade	Reactor Grade	Weapon Grade	Reactor Grade	Weapon Grade
1.1 barn	1.8 barn	38 barn	42 barn	80 barn
Flux : 10^{15}		Flux : 10^{14}		10^{14}

The large fission plutonium cross section allows a strong denaturing within a short irradiation tune.
In fact, for a standard fuel management length (300 efpd), the output isotopic plutonium vector is the following (^{239}Pu : 57%, ^{240}Pu : 35%, ^{241}Pu : 7%, ^{242}Pu : 1%). The degradation of the plutonium vector is fast.

In the CANDU, the initial fissile inventroy is small. For a CANDU with MOX fuel and WG Pu, the inventory of Plutonium disposed is small (≈ 1 metric Tons). The initial inventory is smaller compared to other reactors but it correspond at one year of operation.

The reactivity coefficients were not evaluated.

The CANDU reactor has a good potential in term of plutonium disposition and mass disposal capacity. The reactivity coefficients were not evaluated by ourself.

5. IMPROVEMENT OF REPROCESSING RESISTANCE

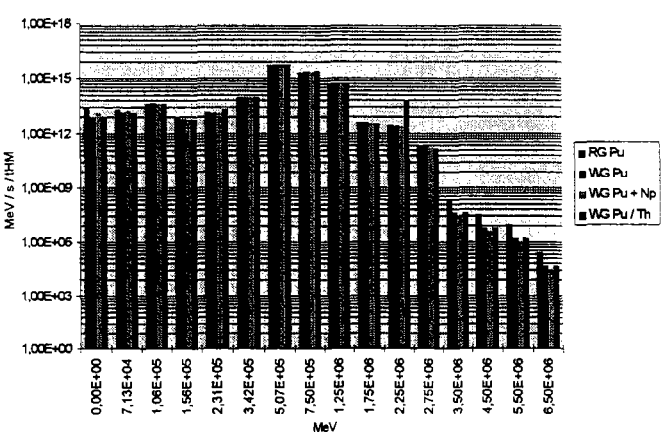
In the previous chapters, we have presented some results in term of reprocessing resistance, especially by introducing neptunium, americium or by using reprocessed uranium as a support for the MOX fuel. The table 9 gives the increase of decay heat in case of ^{237}Np, ^{241}Am adding.

Table 9 : Impact of adding materials to improve the reprocessing resistance : Decay Heat (w/kg), Neutron emission (n/s. kg)

	PWR		
	std	1% Am	1%Np
Decay Heat	8	38 (x5)	56 (x7)
neutron emission	$3.8\ 10^{7}$	4.2 10'	5.1 10'

The impact on the cycle is strong (factor 10). The tendencies are very similar in FR. Nevertheless, the increase is not significant enough to improve the reprocessing resistance.

The same conclusions can be drawn for the doses when thorium or neptunium is added, compared to a standard MOX fuel.



The impact on the cycle operation is not strong enough when we add Np or Am in order to improve the reprocessing resistance.

7. *COMPARISON* OF DIFFERENT REACTORS.

Finally, we compare the plutonium disposition in different reactors, based on the following parameters :

Mass consumption per day
Inventory in the core per electricity unit
Irradiation time for a minimum plutonium degradation (20% $^{240}Pu/Pu$ total)

	Core concerts			Fuel concept
	PWR MOX	FR MOX CAPRA	'CANDU	PWR inert matrice
Initial inventories (Kg/MWth)	1.3	1.6	0.5	1.5
Pu Consumption (Kg/efpd)	1.8-1.5** (0.6 for 30% MOX)	1.6	1.2	4
Denaturing*	600	950	100	300

Time of irradiation (e&d) to obtain 20% $^{240}Pu/Pu$ total
*** Depending on fuel management*

For a HTR, the Initial inventories and the Pu Consumption are respectively :1.3 Kg/MWth and 0.6 Kg/efpd.

The WG plutonium loaded mass per year are presented following per reactors :

EPR 100%MOX (4250 MWth) : 1600 Kg
EPR 20% MOX . 325 Kg
VVER-1000 30 %MOX[1] : 270 Kg
VVER- 1000 43 %MOX[2] : 460 Kg
BN- 600 18%MOX[3] 240 kg
BN- 600 20%MOX[4] 300 kg
BN-600 100%MOX 1300 Kg
BN-800 100%MOX 1600 Kg
CANDU (2160 MWth) 1000 Kg
HTR (600 MWth) 250 Kg

[1] : *3 years cycle for MOX fuel*
[2] : 2 *years cycle for MOX fuel*
[3] : 67 *MOX subassemblies in BN 600*
[4] : *84 MOX subassemblies in BN 600*

These values are given for indications only. They are subject to change various the fuel management.

This panorama of solutions, some of them being feasible with small effort, shows that the plutonium disposition is efficient and allows a fast reduction of WG plutonium.

5. CONCLUSIONS

The weapon grade plutonium disposition is efficient when using standard reactors. Anyway, due to their larger velocity of consumption, FR may be used. Special designs can improve the Pu consumption, by increasing the Pu content or removing the uranium support. Using neptunium improves also the reprocessing resistance.

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SAFEGUARDS ON MOX ASSEMBLIES AT LWRs

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Abstract

Operating within the framework of the New Partnership Approach (NPA) for unirradiated MOX fuel assemblies in LWRs, the IAEA and EURATOM have gained experience in safeguarding 13 LWRs licensed to operate with MOX assemblies. In order to fulfil SIR requirements, verification methods and techniques capable of measuring MOX assemblies under water have been and are still being developed. These encompass both qualitative tests for the detection of plutonium (gross attribute tests) and quantitative tests for the measurement of the amount of plutonium (partial defect tests) and are based on gamma and neutron detection techniques. There are nine PWR and two BWR where the reactor and the spent fuel pond can be covered by the same surveillance device. These are Type I reactors where the reactor and the pond are located in the same hall. In these type of facilities relying on surveillance during the MOX refuelling is especially difficult at the BWRs due to the depth of the core pond. There are two PWR type facilities where the reactor and the spent fuel pond are located in different halls and cannot be covered by the same surveillance device (Type II). An open core camera has not been installed during refuelling and therefore indirect surveillance is currently used to survey MOX loading. Improvements are therefore required and are under consideration. After receipt at the facility, there are a few facilities which must keep the received fresh MOX fuel in wet storage, not only for a short period prior to refuelling, but for more than a year, until the next refuelling campaign. In these cases timely inspections for direct use fresh nuclear material require considerable inspection effort. Additionally, where human surveillance of core loading and finally core closure are necessary there is also a large demand for manpower. Either an agreement should be reached with the operators to delay the MOX loading until the end of the fuelling campaign, or alternative approaches should be sought to optimise inspection efforts. State of the art technology in containment and surveillance devices and systems, as well as ongoing developments in NDA techniques have proven feasible for implementing an integrated safeguards approach in these types of facilities. In an unattended mode of safeguards, it is proposed to survey the loading of unirradiated MOX assemblies in complex design facilities using gate monitors (radiation detectors) combined with digital surveillance. This decreases the probability of failure of containment and surveillance during refuelling periods and reduces the manpower effort of the inspectorate. If there is a loss of continuity of knowledge during loading of MOX assemblies, it is proposed that NDA techniques be implemented, based on gamma spectrometry and neutron yield measurements to differentiate irradiated MOX from irradiated LEU assemblies. One such technique which is under development employs a high resolution CdZnTe, SPD 3 10 Z 205 which uses the neutron emission relative to burn-up (Cs-137 signal) to differentiate between irradiated MOX and LEU assemblies. This assures that MOX loading has occurred and re-establishes the continuity of knowledge.

I. Introduction

Since 1973 when the first MOX assembly was received at an LWR in an EURATOM facility till now 13 reactors are safeguarded: 11 PWR and 2 BWR are licensed to use MOX.

The Mixed Oxide Fuel (MOX) has in order of 8 kg of Pu (1 Significant Quantity) per assembly in BWR and up to 36 kg of Pu per assembly in PWR (4 SQ)

Pu concentration = 4% - 8%

Pu Quality ≅ 70%

For safeguards purposes we have classified the reactors in 2 types:

- Type I: Reactor and Spent Fuel pond can be fully covered by the same Containment /Surveillance (C/S) device and are both in the same containment.

- Type II: Reactor and SF can not be fully covered by the same C/S device, usually are in separate containment.

BWRs of type I, due to the deep core pond, offers difficulties to be covered by adequate direct surveillance of MOX loading, and reactors type II require enhanced C/S and use of radiation detectors (NDA).

The use of MOX fuel is enforced by contractual obligations to take back the plutonium from reprocessing contracts, often the number of assemblies exceeds the limits to be loaded in one refuelling campaign; therefore the MOX assemblies remain in the SF pond for a long time requiring inspection effort and adequate use of NDA to cover safeguards timeliness for direct use material.

The MOX assemblies are usually not loaded in one batch, therefore inspector manpower and enhanced C/S are also required to cover the refuelling period and to have assurance of loading of the MOX assemblies into the core.

To cover from a loss of continuity of knowledge of C/S and to have credible assurance that the fresh MOX has been irradiated, “state of the art” NDA has been developed using neutron measurements and gamma spectrometry.

2. Criteria Requirements

2.1 Examination of records and reports, during each inspection, for correctness and internal consistency and, for the IAEA, comparison with state reports.

2.2 Physical Inventory Verification

a) Fresh MOX fuel under C/S (surveillance)

The C/S is evaluated and the assemblies are item counted, verified by serial number identification where applicable and measured with 10% detection probability for gross defects.

For assemblies under single C/S (seals) seal verification is performed with medium detection probability. Additionally, item counting, verification by serial number identification where applicable and re-measurement with 10% probability are performed on sealed items selected for re-measurements.

b) Assemblies not under C/S are item counted and verified with a high detection probability for gross and partial defects and by serial number identification where applicable.

2. 3 Verification of domestic and international transfers:

Item counting and verification with high detection probability for gross and partial defects, either at the shipping or receiving facility. Continuity of knowledge can be maintained using C/S.

2.4 Verification for timely detection

Verifications of fresh MOX fuel are carried out 12 times per calendar year at monthly intervals.

3. Safeguards Approach implemented

Of particular importance is the requirement to verify, the down-grading from unirradiated to irradiated direct use material by core loading. Currently most fresh MOX fuel assemblies are verified by NDA for gross and partial defects at the fabrication plant and shipped under seal. Only if the fabrication plant is not under safeguards, the verification is routinely done upon receipt at the reactor.

Safeguards measures at the reactor are normally limited to maintain C-o-K (continuity of knowledge) of the verification at the fabrication plant by application of C/S measures until the MOX assemblies are loaded into the reactor core and the core is closed. NDA re-measurement at PIV for gross defects is only necessary if some MOX assemblies are not loaded into the core but remain in the storage after refuelling. However, MOX assemblies must be re-verified within a timeliness period whenever C/S evaluation is not conclusive.

Regarding the implemented C/S measures three operational situations must be distinguished.

3.1 Receipt and transfer to storage

a) An inspector must be present to remove the seal(s) on the shipping cask. Only in exceptional cases the inspector must verify the received assemblies. Such a need arises when the MOX assemblies were not verified at the fabrication plant or not shipped under seal or if the sealing was inconclusive. Whenever possible this is done prior to the transfer into wet storage.

b) During the unpacking and transfer of MOX assemblies to their final storage position, C-o-K is generally maintained by human surveillance, complemented by temporary C/S measures and standard pond surveillance which is switched to shorter intervals.

3.2 Fresh MOX Assemblies in Storage

For safety reasons, fresh MOX assemblies are normally stored in wet storage in dedicated positions of the SF pond separated from other stored items. Only in a few cases MOX assemblies are stored in dry storage. Safeguards measures are:

a) In wet storage

Underwater TV cameras with short intervals (fig. 1). In addition standard pond surveillance is switched to shorter intervals.

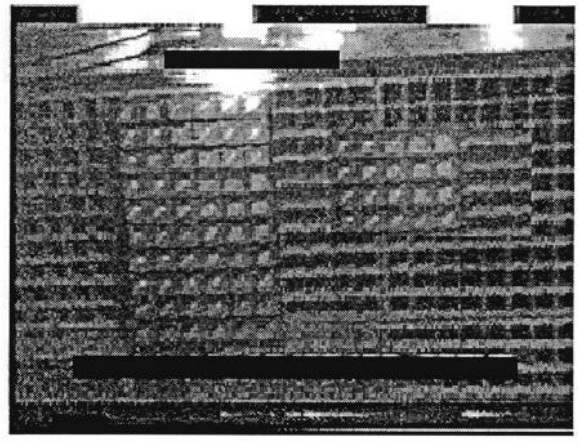

Fig. 1

Under water survey of fresh MOX assemblies

b) In dry storage

Each MOX assembly is sealed separately.
C/S measures in MOX storage must be evaluated in monthly intervals. If C/S evaluation is not conclusive positive re-verification for gross and partial defects with high detection probability is required. This is no longer possible as soon as the core loading commenced. In order to enable prompt follow-up action in case of any problem it is desirable to perform a review of surveillance records on site or to have remote monitoring.

3.3 Core Fuelling

The current intention is to apply surveillance for tracing the path of each fuel assembly to the core and to confirm that it has not been removed from there until the core is closed and standard C/S measures for the core are re-applied. Two types of reactor designs must be distinguished.

(i) Type I reactor:

Spent fuel pond, transfer gate and core pit are all inside the reactor containment and can be covered by the same system of surveillance measures (fig. 2).

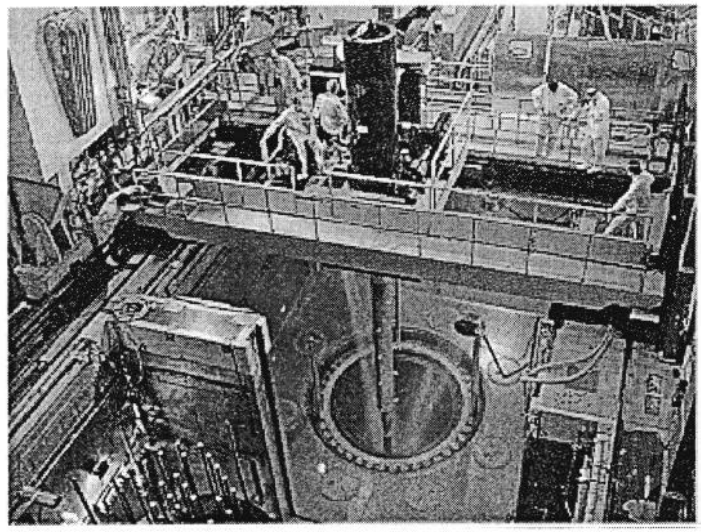

Fig. 2
Type I reactor

(ii) Type II reactor:
The spent fuel pond is a separate building connected by a transfer channel with a temporary reactor pond, and the reactor pit. Separate systems of C/S measures must be applied to cover the spent fuel pond, the transfer channel, the temporary reactor pond, and the reactor core pit.

Regarding the surveillance coverage of MOX loading to the core different approaches have been implemented:

a) Human Surveillance of Core Loading
Inspectors are present 24 hours a day starting from the first MOX fuel transfer until core closure. They observe the loading of the MOX assemblies into the core and must ascertain that none of them is removed, concealed as the discharge of spent fuel or a dummy. Human surveillance is complemented by the underwater surveillance of the wet MOX storage and by standard LWR surveillance which is switched to a higher recording frequency of effectively one minute. In some cases additional cameras are installed. For Type II reactors 2 inspectors per shift may be necessary to cover storage pond and reactor core

b) Unattended Surveillance with open core camera

This is the "classical" approach implemented under the New Partnership Approach. An unattended surveillance system consisting of underwater surveillance of the MOX storage area in the pond, of the open core and (for type II reactors) the transfer channel is installed which is complemented by the standard LWR surveillance system switched to higher recording frequency. This arrangement should ensure that the movement of a MOX assembly can be followed during surveillance review from the storage position in the core. It should also be possible to confirm that MOX assemblies are not moved back from the core to the pond (fig. 3).

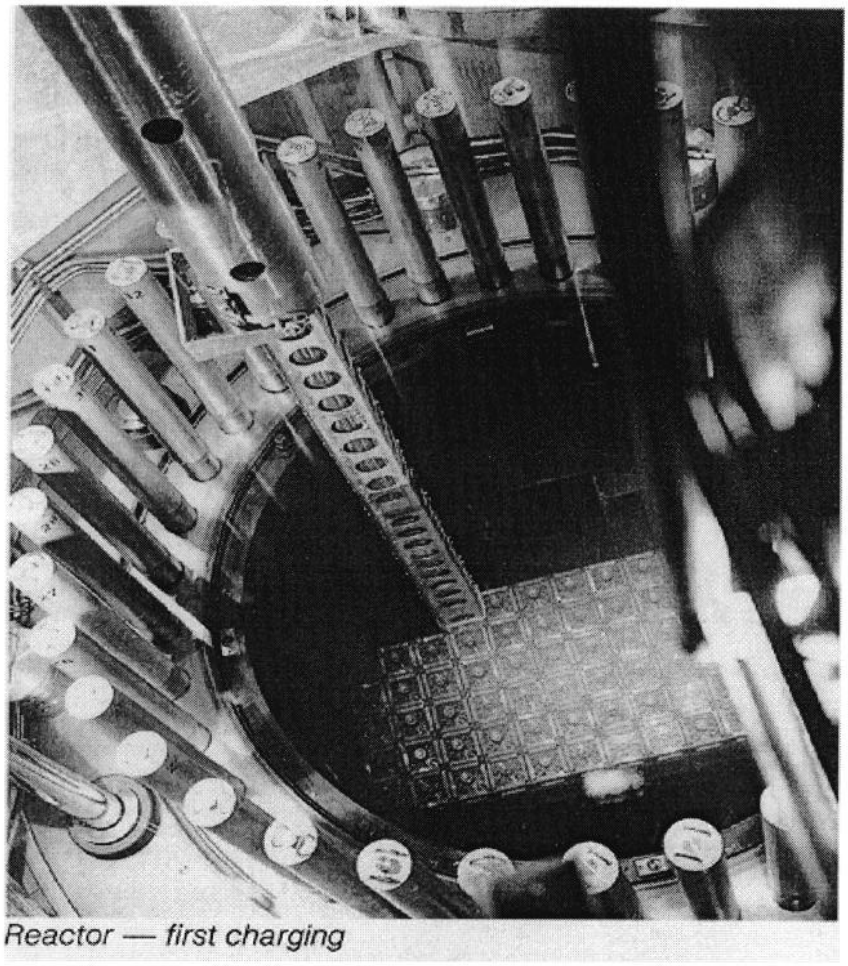

Fig. 3
Open core camera at PWR

4. Technical Safeguards measures for MOX LWR Type assemblies

Criteria shall be verified for Gross and Partial defects.

Gross (Attribute) defect test - a qualitative test. For a positive result of a gross defect test, the detection of plutonium presence is sufficient.
Partial defect test - a quantitative test. The amount of plutonium in the verified assembly must be measured with a specified accuracy.

A spent (irradiated) MOX assembly stored in a spent fuel pond is so-called "self-protected" by extensive gamma-radiation and contains indirect use material. Only a gross defect test is required for a spent MOX assembly. However, if the spent MOX assembly is to be transferred from the plant into the dry storage (difficult to access area) it must be verified for partial defects as any other spent fuel assembly.

4.1 Methods and techniques for fresh MOX LWR type assemblies underwater verification

The main plutonium specific features which can be used for gross and partial defects tests of MOX assemblies are:

- well identified gamma-lines of different plutonium isotopes;
- spontaneous fission neutrons of even (mainly Pu-240) plutonium isotopes.

It should be noted that any kind of underwater MOX fuel assembly measurement in a spent fuel pond is always intrusive, because it requires movement of the assembly or, at least, partial withdrawal from its position in the rack. Without movement (measuring from the top) even the gross defects test is not feasible, because of the long distance to the fuel zone. However, for reasons of safety fresh MOX assemblies are often not stored adjacent to each other. In these cases - if no spent fuel assemblies are stored in the vicinity - in situ gross defects tests can be done, by inserting a detector into an empty storage rack location next to the assembly to verify.

For gross defects tests one relatively simple and easy to use method is being introduced in safeguards inspection practice - the use of a watertight collimated compact spectrometric CdZnTe detector [1] which is lowered under water on a long cable from the pond bridge or from the edge of the pond. The detector is positioned as closely as possible to the side of assembly, which is partly withdrawn from the rack. The gamma spectrum is accumulated and displayed by an energy calibrated miniature MCA. The energy resolution of the detector is good enough to detect underwater at least one plutonium gamma peak at 208 keV (Pu-241). Excessive scattering of gamma-radiation in water can however negatively affect the measurements, if the detector is not well collimated or is not close enough to the assembly. Another possible disturbing factor is the proximity of spent fuel assemblies which could obscure the entire gamma spectrum. A neutron gross counter (He-3-tube) is being considered to be added to complement the gamma measurement.

A more sophisticated and powerful method suitable for gross and partial defects tests is based on the well known neutron coincidence counting technique and realized in the Under Water Neutron Coincidence Counter (UWCC) [2,3]. The detector consists of a solid polyethylene body with two polyethylene fingers and the distance between the fingers is suitable for either a PWR or a BWR size assembly. Each finger contains two high sensitivity He-3 tubes. The body is fixed at the bottom of a long rigid watertight pipe. Through this pipe neutron signal cables come to a standard neutron coincidence shift register electronics (JSR-12). The device is capable to determine the axial density of even plutonium isotopes ("Pu-240eff"), which could be transformed into the total plutonium density, provided the isotopic

composition is known (normally it is verified at the fuel assembly fabrication plant). The measurement accuracy with appropriate calibration is about 2-3%.

4.2 Spent LWR Type MOX Assembly Verification

A gross defects test for the normal LEU LWR type assemblies is usually performed by the Improved Cerenkov Viewing Device (ICVD) from the spent fuel pond bridge. The method is fast, non-intrusive and well established. It is not applicable, however, for MOX spent fuel, as it is not capable to distinguish between MOX and standard LEU spent fuel assemblies. The only easy indicator to detect the difference between spent LEU and MOX fuel assemblies is the neutron emission rate, which is significantly (factor of 10) higher for MOX assemblies, when the irradiation history of both types of assemblies is comparable. One of the standard safeguards techniques for LWR spent fuel assembly verification is the fork detector (FDET), which is used if ICVD application is not possible (very long cooling time, bad water transparency), or some additional information is required,

The FDET detector mechanical construction is the same as the mentioned above UWCC , but He-3 tubes are substituted by fission chambers. The He-3 tubes are too sensitive to high gamma radiation fields. Also, the FDET contains one ionization chamber to monitor the gross gamma activity of the verified assembly. Thus the FDET detects two spent fuel attributes: neutrons (arising mainly from Cm-242 and Cm-244 isotopes) and gammas (mainly from fission and activation products). A part of the attribute (gross defect) test, the FDET detector is capable to provide a consistency check with operator declared burn up and cooling time.

The FDET is able to distinguish between MOX and LEU spent-fuel assemblies by neutron count rate comparison, unless the irradiation history of MOX and LEU assemblies is completely different. A one-cycle MOX assembly produces approximately the same amount of neutrons as a standard LEU assembly of three irradiation cycles. If it is the case, the gross gamma activity measurement helps, because a 3-cycle LEU assembly produces significantly higher gamma radiation than a l-cycle MOX assembly.

However even a combination of both neutron and gross gamma measurements fails to distinguish MOX and LEU assemblies, when the cooling time is short, because with short cooling time (a few weeks) the gross gamma emission does not reflect the whole irradiation history, but only the last weeks of operation. The solution has been found through introduction of spectrometric (instead of gross) gamma measurements [4,4], using miniature spectrometric CsZnTe detector. It was experimentally shown, that such a detector, if adequately shielded and collimated, is capable to detect quantitatively the 662 keV peak from Cs-137 even for short cooling times. Because of a very long decay time, the Cs-137 fission product activity is proportional to the burn up, which obviously is much higher for a 3-cycle LEU assembly than for a l-cycle MOX assembly. At present the method and device are in the process of development (Reference 4). A partial defect test for a spent fuel assembly is formulated as a confirmation that no more than 50% of fuel is removed from the assembly with

a confidence level of 3σ. This requirement does not look strong, however, it is not a straight forward task for any kind of spent fuel. At present a number of possible solutions are under consideration. One of them could be based again on the combination of neutron and gamma spectrometry measurements. Assuming that the neutron emission is proportional to the amount of fuel and is a power function of the burn up, and on the other side, the Cs-134/Cs-137 activity ratio, after correction for irradiation history and cooling time, is linearly proportional to the burn up and presumably does not depend on the amount of pins removed from the assembly, the calibration curve N = F (Cs-137/Cs-134) could be created for any given type of fuel. This approach is under investigation. A second approach is the development of under water tomography of spent fuel assemblies [ref. 5] into a practical method which can potentially detect the removal of single rods in a measurement time of less than 30 min.

5. Experience achieved

In order to judge the effects of the efforts undertaken we should differentiate between detection goal attainment at all (quantity component) and timely detection goal attainment (timeliness component).
The timeliness goal for fresh MOX fuel is very demanding and seldom attained whenever a primary C/S measure failed to be conclusive and fresh MOX spent fuel must be re-verified. Priority is given to attaining the quantity component.

- It is agreed that MOX assemblies which will be shipped to a reactor which is under safeguards will be submitted to safeguards already prior to shipment (i.e. from MOX fuel fabrication plants in nuclear weapon states). They will be verified at the fabrication and shipped under seal.

- At a reactor of type II the operator was reluctant, on safety grounds, to accept the installation of underwater surveillance for the open core during refuelling.

- At one large BWR a good resolution of the open core surveillance was impossible to achieve. The core pond is too deep to maintain the continuity of knowledge on the small BWR MOX fuel assemblies in the large deep core pit with about 700 assemblies positions. An alternative approach implemented for indirect confirmation (survey of canalgate) that the declared number of MOX assemblies was loaded into the core was partially accepted; due the operational needs, to remove core fuel assemblies from the core to the dismantling station, does not assure that no unirradiated MOX assemblies were returning back from the core (fig. 4).

In order to solve the problems currently still preventing attainment of the quantity component at the type II reactors and large BWR, action has been initiated leaving 3 options to be followed:

a) Human surveillance of the open core

Inspectors present during the whole period of MOX loading observe the loading of the fresh MOX and keep the knowledge of MOX position in the core to confirm that MOX assemblies are not moved out of the core.

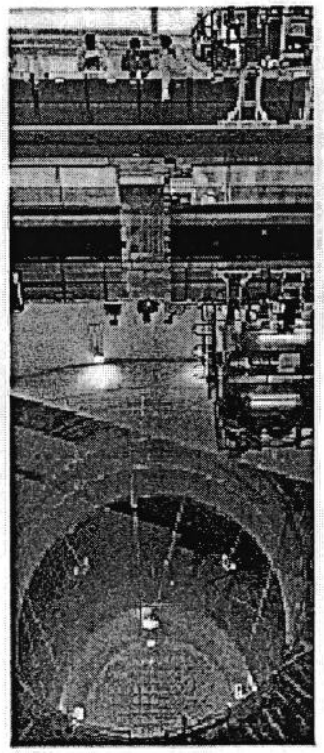

Fig. 4
BWR with deep core pond

If possible, an agreement should be made with the operator that MOX is loaded at the end of the campaign to reduce the otherwise very high inspection effort.

b) Open Core Camera

This requires positioning of a relatively large number of surveillance cameras, including underwater cameras looking into the reactor core. These arrangements should ensure that the movement of a MOX assembly can be followed during the surveillance review from the storage position to the dedicated position in the core. It should also be possible to confirm that MOX is not moved back from the core to the pond. The interpretation of surveillance records might be difficult for type II reactors (involvement of two bridges and the presence of a channel gate where an assembly may be hidden).

c) "Combined approach"
Cameras are installed for observation of the water surfaces in the pond and reactor area and for observation of the movement of MOX assemblies from the storage position to the channel gate. Before the loading starts the inventory of the pond is established. Upon completion of loading and sealing/fixing the channel gate, the core control is performed and the pond inventory is verified (including verification of fresh assemblies and of any dummies in the pond). These measures confirm the loading of MOX into the core by difference.

6. Needs for further Research and Development (R&D)

In the area of Containment and Surveillance (C/S) remote monitoring techniques will be considered for already installed C/S equipment to reduce the necessary number of inspections for timeliness purposes, in particular if MOX fuel is stored for longer periods and to facilitate timely re-verification in case of inconclusive C/S results. In addition new C/S measures are currently being investigated for spent MOX fuel safeguards measures. These include:

a) An unattended gate monitor in the transfer channel for monitoring the flow of MOX assemblies between the spent fuel pond and the reactor core. The Gate Monitor is an integration of NDA equipment and new generation digital surveillance-instrumentation on an adequate platform to perform front-end triggering and back-end review of the collected data. A support programme task will be defined for the development activities. Investigations are carried out about the feasibility of gate monitor in the transfer channel for ensuring that no unirradiated MOX assemblies were returning back from the core. Special He-3 tube detectors, radiation resistent, are being tested for this purpose. The main users will be reactors of type II and BWR using MOX fuel.

b) Underwater surveillance equipment utilizing the existing and authenticated digital surveillance technology will need to be developed and tested. Remote monitoring fresh MOX fuel is on storage will reduce the number of inspections for timeliness purposes, it also facilitates the detection of inconclusive C/S for timely re-verification.

c) Improved sealing methods have been identified as a need for better MOX safeguards measures. Therefore the feasibility of an ultra-sonic seal for fuel assemblies should be further investigated. An existing task of the EURATOM Support Programme should be further sponsored by other Support Programmes to assure timely success.

Regarding the large BWRs investigations are carried about the feasibility to lower down new, particularly small, underwater cameras into the reactor pit for better view of the open core during refuelling.

A method is being developed to re-verify MOX fuel assemblies loaded to the core after the next irradiation cycle when a surveillance failure happens during or shortly after core loading. It is based on the use of neutron yield measurements and ratio of Cs-137/134 (gamma spectrometry) using CdTe miniature detectors.

7. Conclusion

From the previous discussion, it can be understood that the Safeguards measures for MOX LWR assemblies is still an on-going and developing field in safeguards. A good deal of work has been done and effective measures exists as a result, but the complete and effective safeguarding of MOX assemblies requires further development.

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MANAGEMENT OF THE MOX FUEL CYCLE

(Session V)

Invited Paper

THE TRANSPORTATION OF PuO_2 AND MOX FUEL AND MANAGEMENT OF IRRADIATED MOX FUEL

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Abstract

Information on the transportation of PuO_2 and mixed-oxide (MOX) fuel, the regulatory requirements for transportation, the packages used and the security provisions for transports are given. The experience with and management of irradiated MOX fuel and the reprocessing of MOX fuel are described. Information on the amount of MOX fuel irradiated are provided.

1. INTRODUCTION

Spent fuel management comprises the technical operations that begin with the discharge of spent fuel from a power reactor and end either with the reprocessing of spent fuel and recycling of plutonium and uranium in new mixed oxide fuel (closed cycle) or with the direct disposal of the spent fuel elements in a geological repository (open, once-through cycle).

A third approach is the deferral of a decision, the 'wait and see' strategy with interim storage, which provides the ability to monitor the storage continuously and to retrieve the spent fuel later for either reprocessing or direct disposal.

Countries like Canada, Sweden and USA retain their plutonium in the spent fuel and are planning to put their fuel in long-term storage followed by final disposal in deep geological formations. At least part of the fuel is reprocessed from countries like Belgium, France, Germany, Japan, Russia, Switzerland and UK. Countries with smaller nuclear programmes are currently deferring a decision on which of the strategies to select and are storing their fuel.

Plutonium from reprocessing is recycled in MOX fuel in Belgium, France, Germany, Japan, Russia and Switzerland.

Plutonium recycling and MOX fuel production are mature industries in Europe. PuO_2 powder and MOX fuel rods and assemblies are transported regularly. There is also wide experience in operating reactors with MOX fuel.

2. TRANSPORTATION OF PuO_2 AND MOX FUEL

Plutonium and MOX fuel have been safely transported for almost 40 years, mainly by road but also by the sea and air modes.

PuO_2 powder is filled in cans containing about 3 kg plutonium dioxide. According to the IAEA "Regulations for the Safe Transport of Radioactive Material" PuO_2 and MOX fuel must be packaged in accident resistant "Type B" containers which also take into consideration the fissile characteristics

of the contents. BNFL, COGEMA and others have developed packages for PuO_2 powder transport. The BNFL 1680 and the COGEMA FS47 permit the shipment of large quantities of plutonium oxide in powder form. Similarly, packages designed for one or more MOX fuel assemblies have been produced and are currently in use.

2.1 Regulatory requirements

Shipments of plutonium and MOX have to fulfil the highest physical security requirements.

All packages and transport operations must comply with applicable national and international transport safety laws and regulations which are, in practically all cases, based on the transport safety regulations recommended by the IAEA. In addition to having been widely incorporated into national laws and regulations, the IAEA recommendations have also been introduced into international regulations including the:

- UN Committee of Experts on the Transport of Dangerous Goods "Recommendations on the Transport of Dangerous Goods - Model Regulations" (the "Orange Book")
- International Civil Aviation Organization (ICAO) - Technical Instructions for the Safe Transport of Dangerous Goods by Air
- International Maritime Organization (IMO) - International Maritime Dangerous Goods Code
- ADR - European Agreement concerning the International Carriage of Dangerous Goods by Road
- RID - European Agreement concerning the International Carriage of Dangerous Goods by Rail

Physical protection requirements are basically laid down in the IAEA document INFCIRC 225 , Rev. 3 and are further detailed in national regulations and guidelines which are classified "restricted". For MOX shipments a large number of technical and administrative requirements have to be fulfilled.

In addition, for maritime transports, the "Code for the Safe Carriage of Irradiated Nuclear Fuel, Plutonium and High-Level-Wastes in Flasks on board of Ships" (the INF Code) is typically applied to all transports covered by this voluntary code. The IMO is currently taking the necessary steps to make the INF Code mandatory under the provisions of the Safety of Life at Sea Convention.

2.2 Packages

Plutonium transport packaging calls for diverse packaging types suitable for its many forms, ranging from powder to complete MOX fuel assemblies for LWRs and FBRs. Each type of package and design must satisfy the complete set of regulatory design and performance requirements. Although the material may be in very different forms the containment performance requirements for the packaging are the same. All packages for PuO_2 and MOX transport which contain "category I" quantities of plutonium must be designed to meet the Type B criteria. That means they are safe under both normal and accident conditions of transport. Additionally, their designs must be approved to account for the fissile nature of the contents.

For the transport of fresh fuel assemblies it is particularly important to ensure that the fuel is not subjected to any unacceptable shock loads or vibration. Continuous recording of vibration and shock loads are taken using special equipment such as recording accelerometers.

2.3 Transport

Type B(U) F packages are used for transportation of plutonium powder and fresh MOX fuel. Additionally, for road transport, special security trucks are typically used.

Irradiated MOX fuel moved from NPPs to reprocessing facilities is transported in Type B(U)F packages which are heavily shielded and which may be co-loaded with uranium fuel due to residual heat and neutron radiation considerations.

2.3. I Belgium

The transport of plutonium oxide and MOX fuel has been performed in Belgium for more than 30 years. The transport of PuO_2 has reached an industrial stage with the introduction of the French FS 47 packaging. Its handling at the reprocessing plant and at the fuel manufacturing plants, among those Belgonucleaire (Dessel), did not face any difficulty. So this system became standard. The safety margin offered by this packaging is largely in excess of the IAEA regulatory requirements. it has been demonstrated, for instance, that it can withstand extreme external pressures up to 1000 bar and that the seals of the containment envelope would not be affected by a 1000° C fire for a period of one and a half hour.

For smaller quantities of plutonium, TNB 145 packagings can be used too. This design was licensed in Belgium at the end of the 1979's and has been validated in several countries. It is used for the transportation of various types of materials including fissile material such as uranium and plutonium oxide. Various sizes of such drum like packagings are available, depending on the size and quantities of the material to be transported. The maximum allowed quantity of plutonium is 4.5 kg.

The transport of fresh MOX fuel assemblies is performed using packagings specially designed for such purpose. The TNB 176/FS 69 packagings are at present used for the transport of fuel assemblies in Belgium, France, and Switzerland.

In the same manner as for plutonium oxide, stringent security rules apply to the transport of MOX fuel assemblies. Depending on the type of security vehicle used for the transport, up to 8 MOX fuel assemblies can be transported at a time. Detailed procedures have been set up and approved by the Competent Authorities of the various European countries concerned.

Packagings for PuO_2 and MOX fuel assemblies

	Packaging	
	FS 47	**TNB 176/FS 69**
Content	**PUO_2**	**MOX fuel**
Capacity	**19 kg**	**2 assemblies**
Typical Activity (PBq)	**7.5**	**11**
Heat dissipation (max) (kW)	**0.3**	**1.2**
Weight (t)	**1.5**	**5 / 6.6**

2.3.2 *France*

The transport of plutonium and MOX fuel in France is performed in accordance with stringent requirements. France adopted the applicable international regulations which also take into account the IAEA regulations. To meet these regulations safety is usually provided by the package. Different packages have been developed and are used in France for the transport of material containing plutonium: FS-47 for PuO_2 powder, FS-67 for samples and FS-65 and FS-69 for fresh fuel elements.

Most of the plutonium powder is transported from the reprocessing plants to MOX fuel fabrication facilities for LWRs and FBRs (Belgonucleaire in Dessel, Cogema in Cadarache and Melox in Marcoule and in the past also Siemens in Hanau). More than 70 tons have been transported. The plutonium oxide powder separated at La Hague is loaded into stainless steal cans containing about 3 kg each. These cans are loaded into a leak-tight stainless steel canister. For transportation this

container is placed in the FS 47 packaging with an overall weight of 1.5 tons and a capacity of about 19 kg of plutonium oxide. Ten FS 47 packagings are placed in a specially designed container providing additional security protection. A security truck is used for the transport of this type of packaging.

Since 1987, MOX fuel assemblies have been used in 900 MW type NPPs in France. MOX fuel rods are transported from the Cogema Cadarache facility to FBFC Dessel facility for assembling. MOX fuel assemblies are transported from the FBFC Dessel and Melox plants to the nuclear power plants. Two types of B(U)F packages are used which are transported in a security truck. The FS 69, designed for 2 PWR 900 MOX assemblies is equipped with neutron shielding and accommodates 1.2 kW thermal power resulting from the plutonium decay heat. Up to four FS 69 packages can be grouped in a protective container. Up to now 992 MOX fuel assemblies have been transported in the FS 69 container.

In 1994 COGEMA decided to develop a new container (FS 65) for MOX fuel serving both, 900 and 1300 MW PWR and BWR. The FS 65 can load 1 PWR 900 assembly, 2 BWR assemblies or 3 14 rods for PWR 1300 in the FS 65-1300 type. Up to now about 60 000 MOX fuel rods have been transported in the FS 65 and about 30 000 MOX fuel rods in the FS 65-1300.

As there will be up to 28 reactors at the end of the century loading MOX fuel a new packaging type, the MX 8, is under development to reduce the number of transports. This container and its basket are designed for 8 MOX assemblies.

2.3.3 *Germany*

2.3.3.1 Transport of fresh MOX fuel.

For road transport in Germany a combination of security systems is used. overview of equipment)

- an armoured transport vehicle
- *an armoured escort vehicle
- an armoured control center
- *communication lines between vehicles, control center and police
- a vehicle tracking

Drivers, guards and control center operators have to undergo an extensive training and have to be licensed by the BfS.

The armoured transport vehicle “SIFA” is a tractor/trailer combination.

Class I shipments with this SIFA have been performed since 1982. More than 150 shipments from 3 manufacturers to 9 destinations have been carried out.

For shipments between UK and Germany an approach for sea transport has been developed which avoids loading/unloading procedures for container. The safety vehicle picks up the MOX fuel at the fabrication facility at Sellafield, brings it to the port, is loaded onto a Ro-Ro (roll on roll off) ship. and drives it from the German port to the final destination. For MOX scrap from Hanau it goes the other way around. The first transport of this kind took place in October 1996. 6 road/sea/road shipments have been performed since.

2.3.3.2 *Transport of irradiated MOX fuel:*

MOX fuel transported from German NPPs to reprocessing has been co-loaded with uanium fuel due to decay heat and neutron radiation considerations.

Specific transport systems for irradiated MOX fuel was licensed in Germany include:

Transport cask CASTOR-KRB-MOX

License:	D/4193/B(U)F, Rev. 0, 9.9.1986
Capacity:	16 MOX BWR FA
Max. burnup:	14 GWd/t HM
Decay time:	min. 5 years
Enrichment:	max. 2.5 % Pu in U_{nat}

This cask was used for the transport of MOX fuel from Germany to the CLAB facility in Sweden.

Transport cask CASTOR S1

This is a wet transport cask for the transport of spent fuel, including MOX fuel, from German NPPs to the reprocessing plant in Sellafield, UK.

License:	D/4229/B(U)F-85, Rev. 8 validation in France and UK
Capacity:	6 PWR fuel assemblies including 2 MOX fuel assemblies
Max. burnup:	50 GWd/t HM for MOX fuel
Decay time:	min. 2 years
Enrichment:	max. 4.1 % U-235 for Uranium fuel, max. 3.3 % Pu_{fiss} + max. 0.72 % U-235 for MOX fuel

120 transports were made with 4 casks including 15 transports with MOX fuel.

Transport and storage cask CASTOR V/19

License:	D/4323/B(U)F-85, Rev. 1 and D/4312/B(U)F-85, Rev. 2 storage license for Ahaus and Gorleben
Capacity:	19 PWR FA including 4 MOX FA
Max. burnup:	55 GWd/t HM for MOX fuel
Decay time:	min. 10 years
Enrichment:	max. 4.05 % U-235 for Uranium fuel, max. 3.95 % (Pu_{fiss} + U-235) for MOX fuel
Heat load:	max. 39 kW

3 casks are stored each in Ahaus and Gorleben with Uranium fuel only.

Transport and storage cask CASTOR V/52

License:	D/4319/B(U)F-85, Rev.1 storage license for Ahaus
Capacity:	52 BWR FA including 16 MOX FA
Max. burnup:	50 GWd/t HM for MOX fuel
Decay time:	min. 10 years
Enrichment:	max. 4.6 % U-235 for Uranium fuel, >4.2 % U-235 proof of min. 5 GWd/t HM burnup` max. 5.7 % (Pu_{fiss} + U-235) for MOX fuel or 4.9 % Pu_{fiss} and min. 0.2 % U-235.
Heat load:	max. 40 kW

3 casks are stored in Ahaus with uranium fuel only.

2.3.4 Japan

Fresh MOX fuel has been transported to the FBR reactors Monjyu and Joyo and the ATR reactor Fugen using type B(U)F and type B(M)F packages with a cylindrical design. The transports applied to the first core for Monjyu, about 450 MOX assemblies for Joyo and more than 500 MOX assemblies for Fugen.

Later this year it is planned to transport MOX fuel for thermal reactors from the UK to Japan. A modified ship will be used for the escorted shipment.

2.3.5 *Switzerland*

With the exception of small quantities of separated plutonium for research purposes, plutonium is shipped into Switzerland in the form of MOX assemblies only. A number of transport systems have been used over the last 20 years such as

- the French safety vehicle and FS 69 container
- the German safety vehicle "SIFA" with Siemens container.

2.3.6 UK

Plutonium has been safely transported by BNFL both nationally and internationally for the last 35 years. The transport operations have been carried out in accordance with national and international law which is based on regulations recommended by the IAEA.

The current practise in the UK is to use small MOX packages which fit into secure containers. A protection system is used to extend the time required to gain access to the cargo. With EUROMOX BNFL is designing a new system for secure transportation to cover a wide range of LWR fuel types and sizes within Europe.

MOX fuel shipped from the UK to Germany is transported by sea using a UK tractor unit and the German high security trailer. The MOX fuel is transported from Sellafield to the port and loaded on a German vessel as mentioned before.

For the transport to Japan late this year the fuel will be transported by rail from Sellafield to the port. A modified vessel will be used for the escorted shipment.

3. MOX FABRICATION

MOX fabrication is a mature technology in Europe. To better match the plutonium output from reprocessing and to reduce the present stockpile, MOX production capacity and utilization are in the stage to be increased.

Except for Germany (since termination in 1991) and Switzerland, the countries with reprocessing or plutonium recycling have their own MOX fabrication. In the 1950s plutonium from reprocessing was intended for feeding FBRs. In the 196Os, R&D activities started on the utilization of plutonium in MOX fuel for LWRs. The ALKEM facility in Germany for LWR MOX and FBR fuel, Belgonucleaire in Belgium, CEA facility in France, UKAEA in UK and PNC at Tokai Mura in Japan for FBR fuel started operation. The UK also demonstrated MOX utilization in the Windscale AGR.

Because of delays in the deployment of the FBR programme, the utilization of MOX fuel in LWRs became more and more important. Today MOX fuel fabrication and utilization reached a level of about 200 t HM/year and will further increase with the commissioning of BNFL's Sellafield MOX Plant (SMP). (Table 1 MOX fuel fabrication capacity)

While in France MELOX is a basic 17x17 PWR plant for EDF type fuel, Cadarache will cover PWR and BWR mainly for Germany and fast reactor fuel production. Belgonucleaire in Dessel, Belgium, fabricates MOX fuel for Belgium, France, Germany and Switzerland and SMP, UK is designed to make PWR, BWR and fast reactor fuel. Adequate capacity is provided in Japan to cope with the advanced breeder reactor and advanced thermal reactor requirements.

TABLE 1. MOX FUEL FABRICATION CAPACITY 1998

Country	Site	Plant	tHM/y
Belgium	Dessel	PO	35
France	Cadarache	CFC	35
	Marcoule	MELOX	120
India	Tarapur	AFFF	5
Japan	Tokai	PFPF	15
Russian Fed.	Chelyabinsk inside RT 1		
UK	Sellafield	MDF	8
Total			218

4. MANAGEMENT OF IRRADIATED MOX FUEL

Testing of MOX-fuel in reactors has taken place from the early days. In the last decade their performance approached that of conventional UO_2 fuel. Its use has increased enormously over recent years and will continue to do so. Many reactors will go up to 30 % of MOX-fuel in the core. At present European experience of MOX irradiation extends to 52 GWd/t rod average burn-up in commercial PWRs and up to 60 GWd/t in experimental assemblies. These irradiations show a good general behaviour of the MOX fuel.

Plutonium recycling in Germany started in the BWRs Kahl and KRB-A. The commercial MOX programme concentrated on PWRs starting with Obrigheim in Germany and Beznau-2 in Switzerland.

France started its MOX programme in 1987 with the 900 Mwe PWRs. Today MOX fuel is used in LWRs in Belgium, France, Germany and Switzerland. FBRs are in operation in France, India, Japan, Kazakhstan and Russia. (Table 2 Status of MOX fuel utilisation in thermal rectors)

TABLE 2. STATUS OF LARGE SCALE MOX FUEL UTILIZATION IN THERMAL REACTORS
Status end of 1998

	Number of Thermal Reactors			
	Operating [1]	Licensed to use MOX FAs[a]	Loaded with MOX FAs[a]	Applied for MOX license[b]
Belgium	7	2	2	
France	58	20	17	8
Germany	20	12	10	4
Japan	53	3	1	1
Switzerland	5	3	3	
Total	133	40	33	13

a. There are a number of reactors, notably in Europe and India, not included in this Table, which are licensed to use MOX fuel and have MOX fuel loaded on an experimental basis;

b. Technically capable reactors planned to be licensed.

4.1 Management of irradiated MOX-fuel in Belgium =

(Table 3 thermal reactors utilising MOX fuel)

Under experimental conditions MOX fuel from Belgonucleaire was inserted in the BR3 reactor in Belgium and Dodewaard in the Netherlands.

Doel-3 and Tihange-2 are licensed for MOX fuel reload. 56 MOX fuel assemblies have been loaded so far with 4.9% Pu_{fiss} in U_{tails}. The maximum FA burnup is 43,900 MWD/T HM (Table 4 experience with MOX reloads). Belgium also practices the recycling of the reprocessed uranium. As such, no stocks of usable fissile materials are built up.

Belgonucleaire evaluated for Eastern European countries the possibility of loading a WWER-1000 reactor with MOX-fuel. Calculations demonstrated the feasibility of the use of ex-weapon plutonium in MOX-fuel for this reactor type.

4.2 Management of irradiated MOX-fuel in France:

(Table 3 thermal reactors utilising MOX fuel)

France opted for the closed fuel cycle. After slowing down the FBR implementation, emphasis has been given on the plutonium recycling in PWRs. Every year, EDF unloads 1200 to 1300 tons of spent fuel. 850 tons are reprocessed by UP2 and 8 tons of plutonium are recovered. EDF's annual need of MOX fuel is about 120 tons.

EDF decided in 1985 to recycle plutonium in some of its PWR 900 units. A generic safety report was issued at the end of 1986 which demonstrated the feasibility of recycling MOX with a maximum ratio of 30 % MOX assemblies in each reload. This corresponds to 16 assemblies per reload. In 1987 the first MOX fuel was loaded into St. Laurent Bl and B2.

Today 17 of the 20 licensed reactors are loaded with MOX fuel. EDF applied for a license to load MOX fuel for another 8 reactors and in this year the number of plants loading MOX will increase to 19. Up to the end of 1998 992 MOX fuel assemblies were irradiated in France (Table 4 experience with MOX reloads). The licensed plutonium content is 7.08% to cope with the quality of Pu produced now by the reprocessing plant.

Every fuel loaded in a French PWR, including MOX fuel, is intended to be reprocessed and EDF has to demonstrate to the Safety Authority that reprocessing is feasible before loading any new type of fuel. After a cooling period of about 4 to 5 years in the power plant, MOX fuel is sent to La Hague reprocessing plant in the standard transport cask.. Four spent MOX fuel assemblies are loaded together with 8 spent U02 fuel assemblies. The MOX fuel assemblies are placed in the central positions of the cask internal basket and surrounded by the U02 fuel.

Some tons of used MOX assemblies have been reprocessed at La Hague (irradiated in German and Swiss reactors) to demonstrate the industrial feasibility of reprocessing and the possibility of recovering huge quantities of plutonium in the case of an eventual future fast reactor programme.

As used MOX fuel assemblies contain more plutonium (20 kg Pu) than used uranium assemblies (4 kg Pu), one could imagine it would be more beneficial to give them a priority in reprocessing. Nevertheless, second generation plutonium produced from reprocessing is rich in isotopes which make it less energetic in a LWR than first generation plutonium. As the MOX matrix is made with depleted uranium, reprocessed uranium separated from MOX assemblies cannot be recycled in LWRs.

The existing inventory of EDF's used UOz fuel (about 7000 fuel assemblies) permits it to choose for reprocessing those assemblies which contain the most easily handled plutonium in the MOX fuel fabrication plant. Therefore, in order to maximize the advantage of reducing the total inventory of used assemblies, it is not planned to reprocess MOX fuel in the near future.

TABLE 3. THERMAL REACTORS UTILIZATING MOXFUEL ON A LARGE SCALE

Status yearend 1998

	Licensed[a]	**Loaded[a]**	**Applied for licence[b]**
Belgium	Doel 3 Tihange 2	Doel 3 Tihange 2	
France	Blayais 1 Blayais 2 Dampierre 1 Dampierre 2 Dampierre 3 Dampierre 4 Gravelines 1 Gravelines 2 Gravelines 3 Gravelines 4 Tricastin 1 Tricastin 2 Tricastin 3 Tricastin 4 Saint-Laurent B 1 Saint-Laurent B2 Chinon B 1 Chinon B2 Chinon B3 Chinon B4	Blayais 1 Blayais 2 Dampierre 1 Dampierre 2 Dampierre 3 Dampierre 4 Gravelines 1 Gravelines 2 Gravelines 3 Gravelines 4 Tricastin 1 Tricastin 2 Tricastin 3 Tricastin 4 Saint-Laurent B 1 Saint-Laurent B2 Chinon B4	Blayais 3 Blayais 4 Cruas 1 Cruas 2 Cruas 3 Cruas 4 Gravelines C5 Gravelines C6
Germany	Brokdorf Grafenrheinfeld Grohnde Gundremmingen B Gundremmingen C Isar 2 Obrigheim Philippsburg 2 Unterweser Neckarwestheim 2 Emsland Neckarwestheim 1	Brokdorf Grafenrheinfeld Grohnde Gundremmingen B Gundremmingen C Isar 2 Obrigheim Philippsburg 2 Unterweser Neckarwestheim 2	Biblis A Biblis B Brunsbiittel Krümmel
Japan	Fugen Takahama 3 Takahama **4**	Fugen	Fukushima-Daiichi-3
Switzerland	Benau 1 Beznau 2 Gösgen-Däniken	Beznau 1 Beznau 2 Gösgen-Däniken	

[a] There are a number of reactors, notably in Europe and India, not included in this Table, which are licensed to use MOX fuel and have loaded MOX fuel on an experimental basis;

[b] Technically capable reactors planned to be licensed.

4.3 Management of irradiated MOX fuel in Germany:

(Table 3 thermal reactors utilising MOX fuel)

The first MOX fuel was loaded in 1966 in VAK, a small BWR reactor with 6x6 fuel assemblies. In total **113** assemblies containing MOX fuel rods were irradiated in this plant. In 1970

TABLE 4. EXPERIENCE WITH MOX RELOADS IN BWR AND PWR FROM 1981 TO 1998

COUNTRY/ REACTOR -/ FA-TYPE	NO. OF REACTORS	NO. OF MOX FA RELOADED	MAX. AV. PU_{FISS} IN W/O / CARRIER MATERIAL	MAX. FA - EXPOSURE AT EOC IN MWD/TM
Belgium				
PWR (17x17-24)	2	56	4.9 / U_{tails}	43900
France				
PWR (17x17-24)	17	992	7.08 / U_{tails}	40000
Germany				
PWR (18x18-24)	2	24	4.6 / U_{tails}	8000
PWR (16x16-20)	5	364	4.2 / U_{tails}	44900
PWR (15x15-20)	1	32	3.0 / U_{nat}	42000
PWR (14x14-16)	1	41	3.8 / U_{nat}	37000
BWR (9x9- 1)	2	116	3.0 / U_{tails}	32000
Switzerland				
PWR (15x15-20)	1	28	4.8 / U_{tails}	23000
PWR (14x14-17)	2	152	4.1 / U_{tails}	51000

VAK was followed by the BWR KWL and in 1974 by KRB-A in using MOX fuel. In 1972 MOX fuel was inserted in the 2 PWR reactors MZFR and KWO. All this fuel is now stored in CLAB in Sweden in exchange of Swedish fuel sent to La Hague for reprocessing.

In 1981 commercial MOX irradiation started in Germany. Until now, almost 800 MOX fuel assemblies were inserted in German NPPs, including the test assemblies, with a maximum burnup of 45 GWD/t HM. (Table 5)

Some of the commercial MOX fuel was sent for reprocessing and part of it was reprocessed in La Hague in 1992. Most of the German utilities utilizing MOX like PreussenElektra plan to send their fuel for reprocessing. Quite a number of fuel assemblies are waiting for the lifting of the transport suspension order in Germany.

4.4 Management of irradiated MOX-fuel in India:

India has adopted the philosophy of a closed fuel cycle and recycling of plutonium essentially in the fast breeder programme. The 40 MW_{th} Fast Breeder Test Reactor at the Indira Gandhi Centre with the PuC-UC fuel is a forerunner of the fast breeder prograrnme. Because of slow progress in the fast reactor programme, utilization of MOX fuel in thermal reactors has been considered. A study has been carried out by irradiating fuel assemblies consisting of 12 nat. UO_2 fuel rods in the outer periphery and 7 MOX fuel rods in the center in a PHWR reactor. Since the beginning of the 1980s irradiation experiments have been performed on short length MOX fuel rods in the pressurised water loop of the research reactor CIRRUS. After this MOX fuel development programme a few MOX assemblies have been loaded into one of the Tarapur reactors.

TABLE 5. COMMERCIAL MOX FUEL INSERTED IN GERMAN NPPs

Plant	Fuel Type	First Insertion	Total Number of FA accumulated	Maximum FA Burnup [MWd/kgHM]
KWO	14-16	1981	41	37
GKN-1	15-20	1982	32	42
KKU	16-20	1984	- 20	37
	16-20-4	1987	72	40
KKG	16-20	1985	16	40
	16-20-4	1987	44	45
KWG	16-20-4	1988	32	43
KKP-2	16-20-4	1988	32	45
KBR	16-20-4	1989	88	44
GKN-2	18-24-4	1998	8	15
GUN-B	9-1	1996	100	29
GUN-C	9-1	1995	16	34
Total			**501**	**45**

4.5 Management of irradiated MOX-fuel in Italy:

Some MOX fuel was irradiated in Italy before ending the nuclear programme.

Eight fuel assemblies were exposed in Trino until 1985 with a burnup of about 32 GWD/t HM and 63 FA in Garigliano with a maximum burnup of 28 GWD/t HM. There was one full MOX reload at the Garigliano plant.

These MOX assemblies were not sent for reprocessing. It is planned to store the MOX assemblies in the periphery of dry casks together with U assemblies.

4.6 Management of irradiated MOX-fuel in Japan:

(Table 3 thermal reactors utilising MOX fuel)

Japan has experience in burning MOX with two research reactors, Joyo and Fugen and the prototype fast breeder reactor Monju. Joyo used high enriched MOX fuel with 18% and 23%. About 380 irradiated fuel assemblies are in storage while 67 assemblies are still in the core. Fugen uses low enriched MOX fuel with 1.4% having 3 80 assemblies irradiated and 124 still in the core.

The LWR MOX fuel demonstration was started in 1986 at Tsuruga Unit-l for BWR and at Mihama Unit-l for PWR.

The 280 MW FBR Monju reached initial criticality in April 1994. Operation has been suspended because of a sodium leak accident in December 1995.

Japan will start plutonium utilization in LWRs this year by inserting MOX fuel fabricated by BNFL in the MOX demonstration facility.

4.7 Management of irradiated MOX-fuel in Russian Federation:

The information on the use of plutonium in Russia is very limited.

Work on integrating plutonium into the nuclear fuel cycle started in the mid 1970s. More than 300 MOX fuel assemblies have been irradiated in the BOR-60 reactor. The experience gained in this operation was the basis for the design of the MOX fuel inserted in the BN-350, which is now belonging to Kazakhstan and the BN-600. Some of this fuel was reprocessed in the RT-1 (Mayak) reprocessing plant.

The BOR-60 nuclear power plant was operated for 18 years with MOX and had a Pu consumption of 30 - 50 kg/year.

The WWER-1000 and BN-600 reactor designs are the main candidates to involve weapons grade plutonium in their fuel.

4.8 **Management of irradiated MOX-fuel in Switzerland:**

(Table 3 thermal reactors utilising MOX fuel)

In 1978 Beznau started its MOX recycling programme by loading 4 MOX assemblies into unit 1. Up to now, more than 150 MOX assemblies produced by Westinghouse, Belgonucleaire and Siemens have been loaded into both Beznau reactors. Due to slow build up of its own plutonium, NOK, the owner of Beznau, borrowed Pu from other parties. In this way it gained early experience in the use of MOX. The maximum assembly exposure is 43 GWD/t. Today's licensing allows 40% MOX or 48 MOX assemblies in the core. Lately, Goesgen also started to insert MOX fuel in their reactor. (Table 4 experience with MOX reloads).

The irradiated MOX fuel is stored in the reactor pools and it is not intended to send this fuel for reprocessing. In the case of dry interim storage the MOX fuel will be loaded into casks. Due to higher decay heat and neutron doses the number of MOX fuel assemblies per cask might be limited and a co-loading with uranium fuel might be necessary.

4.9 **Management of irradiated MOX-fuel in the UK:**

The UK has loaded some MOX fuel in the past. Experimental loadings of MOX fuel were made in the Windscale Advanced Gas Cooled Reactor in the 1960s. The fuel is being stored at Sellafield and is expected to be reprocessed at the Thorp facility. MOX fuel was also used in the Dounreay Fast Reactor (DFR) and the Prototype Fast Reactor (PFR). The fuel has been reprocessed in a mixed oxide reprocessing plant at Dounreay since 1979. The plutonium recovered was transferred to Sellafield for storage. Completion of the reprocessing is scheduled for around the year 2000. The UK government decided that the Dounreay facilities should close following completion of the existing reprocessing contracts.

5.0 **Other MOX irradiation activities**

The irradiation of the first Argentine prototypes of PHWR MOX fuel began in 1986. Four rods were irradiated in the HFR-Petten reactor in the Netherlands.

Canada fabricated about 150 CANDU MOX bundles from 1979 to 1987 containing more than 3 tons of MOX. Fabrication was suspended in 1987.

5. REPROCESSING OF MOX FUEL

In principle, reprocessing of MOX fuel is not much different from that of UO_2 spent fuel.

France started to gain reprocessing experience in 1967 with FBR fuel followed by Germany in the 1970s with LWR MOX fuel. Japan and UK also gained experience with the reprocessing of FBR fuel. Fuel from the Dounreay Fast Reactor (DFR) and the Prototype Fast Reactor (PFR) has been reprocessed in a mixed oxide reprocessing plant at Dounreay since 1979.

MOX fuel has been designed to be reprocessed. This leads to the monitoring of fresh pellet solubility during fabrication. France demonstrated in 1992 the feasibility of reprocessing MOX fuel in La Hague with a 4.7 ton campaign as mentioned before. The results were quite satisfactory.

6. CONCLUSIONS

There has been extensive experience in the transportation of plutonium powder and MOX fuel. Very rigorous transport safety requirements have been adopted by all participating countries and are based on the IAEA's "Regulations for the Safe Transport of Radioactive Material". MOX fuel is

irradiated in a number of Member States and several have considerable experience in reprocessing irradiated MOX fuel. The storage of MOX fuel prior to reprocessing or final disposal requires longer periods due to higher decay heat and slower decrease of the decay heat compared with uranium fuel.

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MOX FUEL TRANSPORT: THE FRENCH EXPERIENCE

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Abstract

The MOX fuel industry involves various specific materials to be transported at different steps of the process :

To supply to the fuel manufacturing plant with raw material : PuO_2 powder,
Before fuel assembling : MOX fuel rods,
After fuel assembling : MOX fuel assemblies.

UO_2 materials transports are not specific of MOX fuel industry. So, they are not described in this paper.

For these different materials, transport processes are mainly similar and using the same type of vehicle. The packaging themselves are dedicated to each material.

In this paper, we present firstly the common features. Then, material by material, we give information about :

The French experience,
The specific features.

1. COMMON FEATURES

The presence of significative quantity of PuO_2 in the materials to be transported leads to two main constraints :

For safety : highest level of the IAEA regulations for land transport.

As a consequence, type B/F packages are required. It means they shall meet the following functions :

- containment of the nuclear material
- shielding against radiations (gamma and neutrons)
- maintaining subcriticality conditions
- evacuation of residual heat

under normal and accidental conditions of transport.

The transport accidental conditions main specifications are :

- drop from 9 m height on an unyielding surface
- drop from 1 m height on a punch bar
- engulfing fire at a minimum of 800°C during 30 minutes

In comparison, for UO_2 fuel, type A/F packages are required. These packages have to meet less stringent requirements.

For security (or physical protections (PP)) : very stringent measures required by the government of the involved countries. These measures are taken at the equipment level and at the transport process level itsfelf.

Presently, transports are performed by road, using dedicated vehicles. A comprehensive transport organization has been set up by COGEMA and Transnucléaire. Transnucléaire operates a fleet of 7 trucks and 7 semi-trailors, adapted from standard vehicles. Packages are loaded into a security container, which contributes strongly to the security. During transport organization additional measures are taken, mainly to keep the confidentiality of the exchanged information. During the transport, the vehicle is accompanied by armed escorts.

Of course, the detailed measures are submitted to a high confidentiality level and cannot be disclosed in this paper.

2. PUO_2 TRANSPORT

In France, most of the PuO_2 transports are carried out from COGEMA La Hague reprocessing plant to the MOX fuel manufacturing facilities :

- BN plant in Dessel (Belgium),
- COGEMA plant at Cadarache in France.
- COGEMA Melox plant at Marcoule in France.

More than 70 tons of plutonium have thus been transported.

The packaging used for these transports is the so-called FS 47 (see figure 1). 10 FS 47 are transported in a security truck, representing an amount of around 150 kg PuO_2 per transport.

The powder is packed in stainless steel cans, crimped after filling. The cans are then stacked by groups of five in stainless steel canisters, the lid of which is welded. The canister is finally introduced in a specific cylindrical container (AA 227), with a screwed lid. One AA 227 container is loaded in a FS 47.

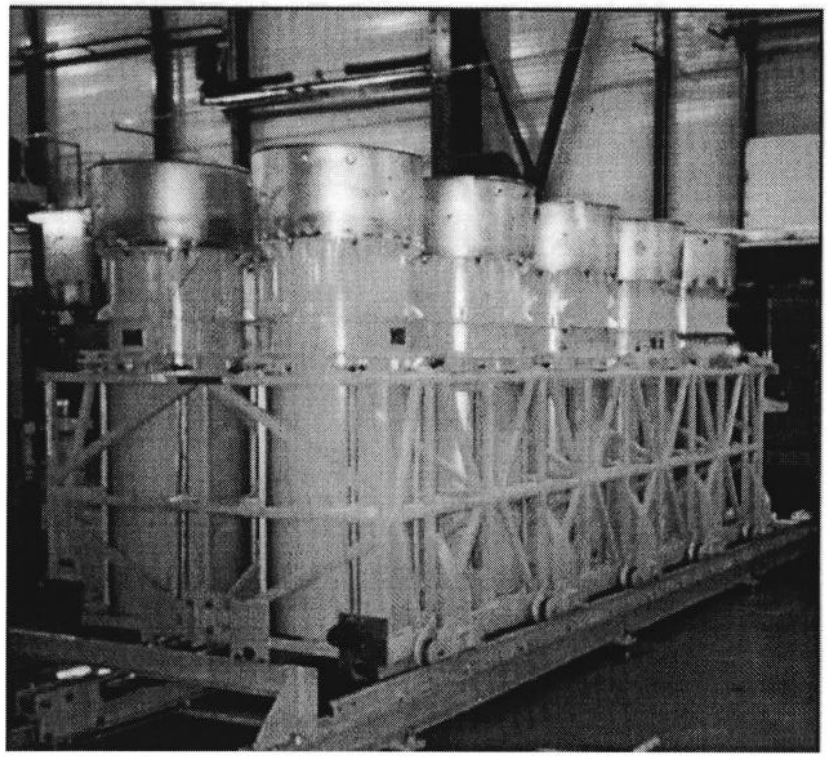

FIG.1. FS 47 Packaging

The FS 47 packaging is a cylindrical vertical packaging approximately 2 m high, 75 cm diameter, 1400 kg weight. The containment vessel, made in stainless steel, is equipped with a bolted closure system, protected by a wood shock absorber. The radiological protection is ensured by a layer of a compound. The insulation is ensured by a layer of plaster. These layers are crossed by cooling fins to improve the heat dissipation. So, as to increase the physical protection of all the process and to reduce the exposure to personnel, the operation of the FS 47 system is fully automated at La Hague and Melox plants.

4. MOX FUEL RODS AND ASSEMBLIES TRANSPORTS

In France, MOX fuel rods are transported from COGEMA Cadarache facility to FBFC Dessel facility for assembling.

MOX fuel assemblies are transported from FBFC Dessel and Melox plants to nuclear power plants.

Two types of packagings are used :

- FS 69, for delivery of 2 fuel assemblies. This packaging shares some features of the well known RCC packaging such as loading and unloading procedures. It consists in :
  - an external body composed of a lower part (caisson) and removable upper part (lid)
  - a cradle suspended to the caisson by means of elastic bearings, the number of which is in relation with the mass of the fuel assemblies, thus protected from external shocks and vibrations
  - a housing providing the two lodgements of the fuel assemblies, fixed to the cradle, able to tilt in a vertical position and equipped with two doors pivoting longitudinally to give access to the fuel assemblies

Up to now, more than 1000 MOX fuel assemblies have thus been transported in the FS 69 packagings.

(see figure 2).

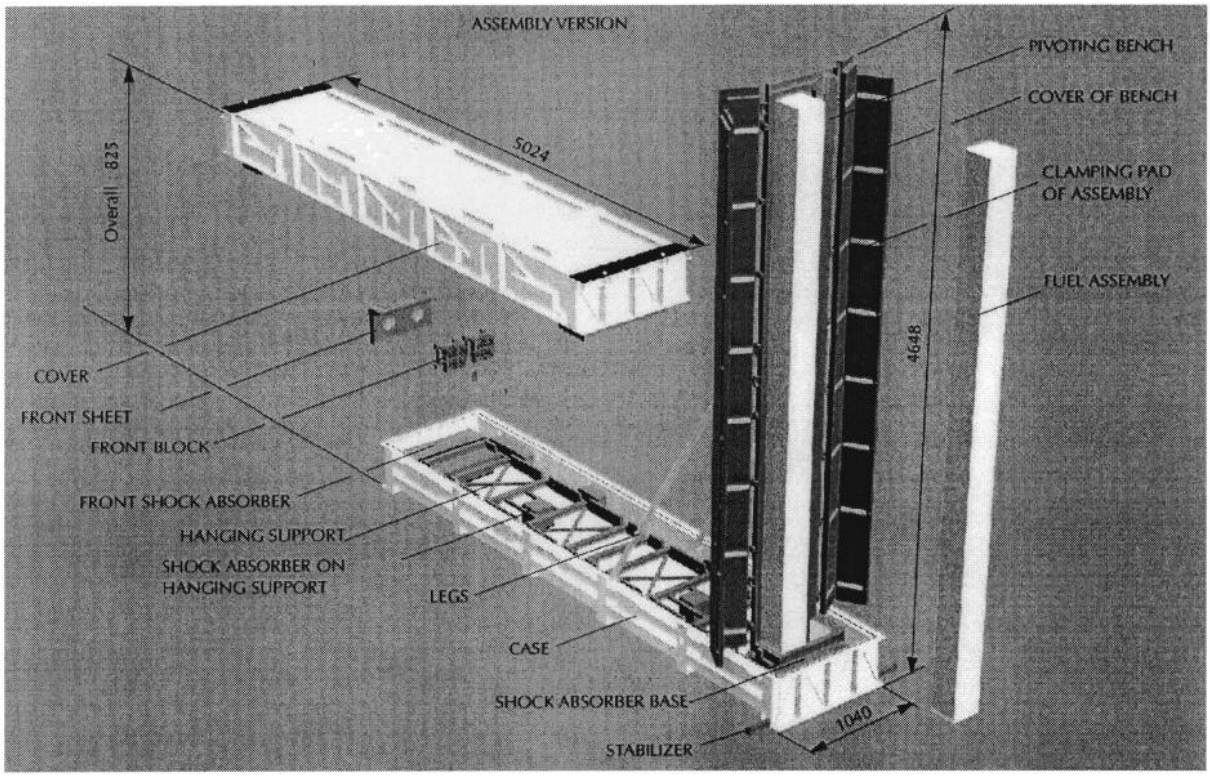

FIG. 2. FS 69 Packaging

FS 65 family either for rods or for assemblies.

Two versions of the packaging, the FS 65 and the FS 65-1300, equipped with internal arrangements allowing to transport various types of fresh MOX assemblies or rods, are used.

TABLE I : CHARACTERISTICS OF THE FS 65 PACKAGING

Contents	FS 65 1 PWR 900 assembly Or 2 BWR 900 assemblies	FS 65-1300 314 fuel rods max 1 PWR 1300 assembly
Thermal power	1000 W max	1100 W
Useful length of cavity	4670 mm	5000 mm
Useful diameter of cavity	500 mm	430 mm
Overall cross section	980 mm	930 mm
Overall length	5323 mm	5643 mm
Weight of the loaded package	5.6 tons	5.6 tons

The FS 65 (or FS 65-1300) packaging is composed of :

- A cylindrical body providing containment and shielding.
- An internal arrangement (basket), in which the contents are tightened.
- An outer frame mainly made of square aluminium tubes, to handle the packaging.
- An anti-vibration system connecting the body to the outer frame, to guarantee the fuel assembly integrity during handling and transport.

Up to now, about 30 000 fuel rods have thus been transported in FS 65-1300 packagings (see figure 3). Previously, about 60 000 fuel rods were transported in FS 69 packagings.

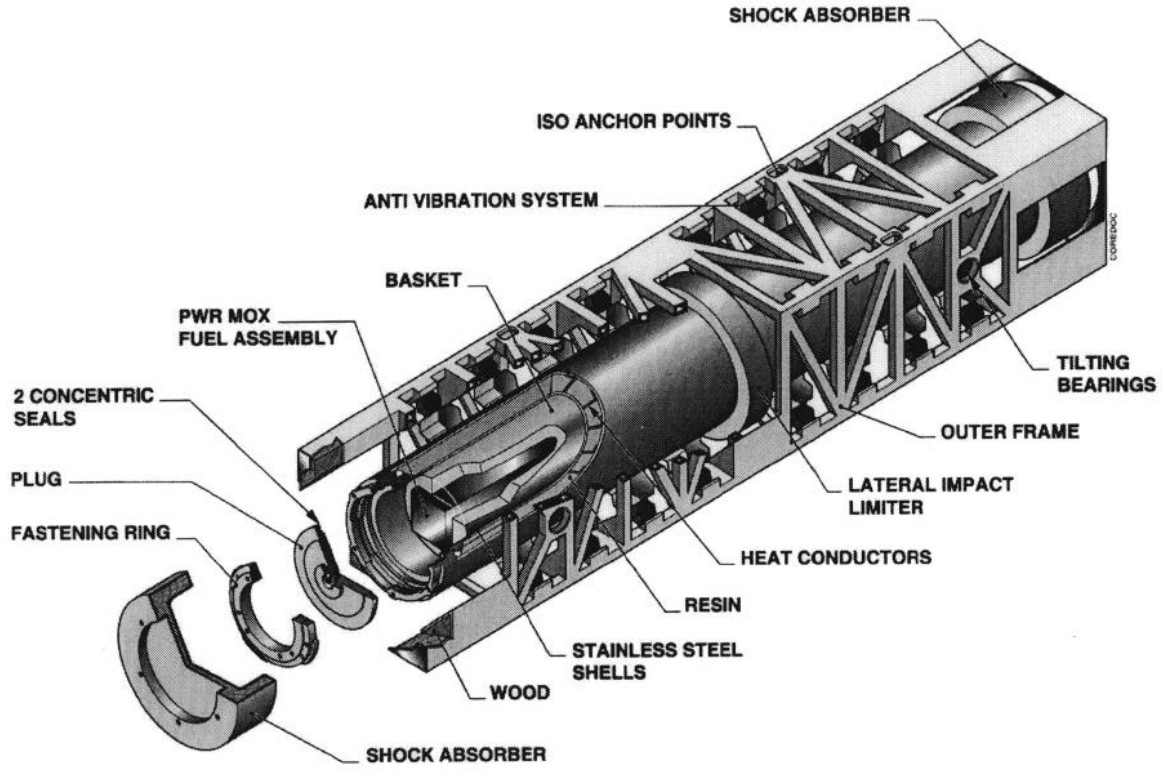

FIG. 3. FS 65 Packaging

5. FUTURE TRENDS

Although to-date plutonium and MOX transports are performed on a large scale with sometimes comfortable safety margins, further evolution is under preparation.

Firstly, to cope with the continued technical and economical optimization of the fuel cycle, higher constraints on PuOz and MOX transportation activities are induced :

Residual thermal power per kg of separated plutonium.
Gamma and neutronic radiation.
Criticality.
Higher Pu content in MOX fuel.

Secondly, new trends in ICRP recommendations and occupational and public exposure regulations are to be complied with.

COGEMA and Transnucleaire are developing new generations of transport systems :

FS 80 transport system for PuOz powder, with a 3 times larger capacity than the FS 47 transport system in order to reduce the number of transports by 3. The goal is to put it in operation in the years 2001/2002.
MX 8 transport system for MOX fuel assemblies (8 assemblies per transport). This is an optimized transport system integrating safety and security requirements on the package itself and in compliance with EdF reactors wet unloading conditions. This new transport system should be commissioned before end of 2000.

- FS 81 transport system for air transport of fresh MOX fuel assemblies. A concept study was made in accordance with the new type C packaging requirements defmed in the 1996 revision of the IAEA regulations and with the requirements set forth by the NRC in 10 CFR Part 71. This type of packaging must, in particular be demonstrated to withstand high speed impact tests of 90 m/s for the IAEA and 129 m/s for the NRC regulations.

COGEMA and Transnucleaire are now preparing large scale fuel assemblies transport from Europe to Japan. FS 65 are used for the road transport in Europe. For sea transport, dedicated heavy packagings (100 t TN 12/2 and TN17/2 packagings previously used for spent fuel) and dedicated ships are used. For that purpose, several dedicated baskets equipped with tie1 tightening systems have been developed, qualified and manufactured.

6. CONCLUSION

The transports of PuOZ and MOX fuel between the reprocessing plant, the manufacturing plants and the power plants in France are now well established on an industrial basis. Transnucléaire has solved and fully mastered the safety and security issues due to the presence of plutonium.

Further developments are underway to increase the payload of the packagings and to improve the transport conditions, safety and security remaining of course top priority.

CHEMICAL AND TECHNOLOGICAL ASPECTS OF SPENT U-Pu FUEL REPROCESSING

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Abstract

Spent U-Pu fuel reprocessing generally conforms to the ideology of the aqueous chemical «Purex» process used in the radiochemical industry. However, the specific features of this fuel, namely, the high Pu content and extended burn-up need modifications of the known processes. These modifications are most substantial as applied to the fast reactor fuel. In a more general aspect the technology of U-Pu materials processing is to be considered together with the preparation of U-Pu oxide powders from regenerates. The product stream of the «Purex» aqueous stage, i.e.,the co-product of U and Pu, is directly supplied to the co-precipitation of both the elements followed by the preparation of U-Pu oxides. The advantages of this technological solution are the high quality of oxide powders in terms of the reactor fuel production (preparation of the true U-Pu solid solutions), the higher solubility of spent fuel in the post-irradiation reprocessing cycle.

1. INTRODUCTION

In the world-wide model of the nuclear fuel cycle plutonium is an inevitable «satellite» of nuclear technologies. Its use in the fast reactors remains a part of the Russia's strategy of the future developments of nuclear power. This fact explains the reason why beginning from 60'"' so much attention is focused on the problems of the regeneration stage of the U-Pu fuel management. To-day the urgency of the investigations is also governed by the objectives of the surplus weapons grade Pu conversion; its utilization via the fast reactor fuel cycle is most efficient.

Under the conditions of the postponed fast reactor fuel cycle France, Germany and other West European countries are realizing the use of MOX fuel in thermal reactors. This model is also under study in Russia as applied to WER-1000 type reactors. The final characteristics of that fuel little differ from those of uranium fuel; it may be reprocessed without any significant changes introduced using the standard aqueous extraction process. However, an order of magnitude higher Pu content and much more extented burn-up of spent fast reactor fuel make it necessary to seriously modify the reprocessing. The main directions of the modifications are discussed below in the order of implementing the process operations.

2. DIRECTIONS OF MODIFYING AQUEOUS EXTRACTION TECHNOLOGY TO MEET OBJECTIVES OF FAST REACTOR U-Pu FUEL REGENERATION

2.1. Opening of fuel

At the RT-1 plant in Russia the process of opening the VVER-440 core fuel rods by mechanical shearing has been mastered that is carried out before the dissolution [1]. This process is also used for the BN-600 fuel. In principle, it is applicable for the resolution of the problem discussed.

However, it is advisable to discuss the feasibility of employing thermal processes at this stage; those processes have substantial R&D back-logs in Russia and the world. In particular, the process of the oxidation treatment of fuel seems to be attractive if implemented in addition to the opening operation. The oxidation treatment within 300-500°C makes it possible to remove quantitatively ^{3}H and ^{129}I from the fuel. This is already an element of the useful partitioning of waste from reprocessing.

However, the partial volatility and other components (caesium, ruthenium, technetium, crypton) «smeared» in the process streams do not promise any simple solutions in terms of their localization; further studies are needed.

2.2. **Dissolution of fuel**

Our experiments carried out with actual samples of high burn-up (up to 100 000 MW.day/t) U-Pu fuel of fast BR-10, BOR-60 (SSC VNIINM, SSC FEI, SSC NIAR) gave an evidence that the quantitative (99.9%) dissolution of U-Pu oxides in concentrated HNO_3 solutions is feasible. However, two substantial limitations are inherent in the results:

(I) need in the stage of the re-dissolution and the introduction of HF into the chemical environment to reduce the residual contents of U and Pu in the indissoluble residues;

(2) highly intensive transition of structural material (steel) components (up to 8%) in solutions at the stage of the basic dissolution and up to 35% in fluorine-ion containing media.

Two conclusions are drawn from this:

(1) special attention is to be focused on the initial nature (morphology) of fuel used in fuel rods; the true U-Pu solid solutions prepared via the co-precipitation dissolve at a higher rate and more completely without the use of chemically aggressive environments. These considerations served in favour of developing the «GRANAT» technology [2];

(2) coming back to the mechanical opening of fuel it is advisable to pre-separate (before the dissolution) the fuel and the structural materials; the technical feasibility of this procedure is undoubtless.

The high content of fissile nuclei in U-Pu fuel limits to a certain extent the capacity of the dissolution units. In this connection at SSC VNIINM a special complex of investigations was carried out on the feasibility of using homogeneous water-soluble neutron absorbers in dissolution processes [3]. The investigations are brought to the industrial level tests at the RT-1 plant in BN-350 fuel reprocessing. However, this operation might be useful only when dissolvers are used that have inadequately high coefficients of nuclear safety margins not intended for U-Pu fuel reprocessing.

In laboratory-scale experiments Pu(VI) was observed to form in the resultant solutions. This is undesirable since it leads to an abnormal build-up of Pu in a specific zone of the subsequent extraction process. It is shown that the holding of the solution or implementing the dissolution process at temperatures a little lower than the boiling temperature level-off this effect.

2.3. **Filtration of initial solutions**

The solutions that result at the dissolution stage are highly dispersed suspensions of 1 g/l. Their conditioning for chemical treatment is a responsible stage.

The principles of approaching the qualitative clarification of high activity level solutions, i.e., suspensions, were discussed by us in paper [4]. The filtration procedure developed at SSC VNIINM involves the application of corrosion resistant moving materials (steel powders etc) and formed the basis of designing virtually ((everlasting)) filters not needing any repair or replacement of the filtration element for the period of the scheduled operation. At TS «SNIIKLM» (Ekaterinburg city) the designers have developed a series of powder filled filters; one of them in the nuclear safe construction is schematically illustrated in fig. 1.

The powder filled filters became a part of the base equipment at the RT-1 plant and confirmed their reliability and efficiency.

2.4. **Studies of extraction chemistry**

A substantial scope of investigations was implemented to study the physical chemistry of the aqueous extraction systems at the concentrations of the components corresponding to the high fuel burn-up and the high Pu content. Mathematical models were designed that describe the extraction equilibria in the main process operations (a large scope of the work was implemented by A.Rozen, L.Andrutsky, V.Rubisov, M.Shapovalov and others at SSC VNIINM).

Much attention was paid to the choice of an extractant from the view point of its homogeneous resistance to the formation of the «second» organic phase at the high Pu content. It is shown that in

the 30% TBP - n-paraffine (C_{10}-C_{15}) systems the homogeneity (with the formation of the «heavy» solvate) is disturbed at the Pu(IV) content of 22-25 g/l in the organic phase. In the TBP – n-dodecane system this parameter is increased to reach 40-50 g/l. As applied to emergency events the studies were carried out of the dynamics of the secondary organic phase formation and behaviour in mixer-settlers and centrifugal contactors at various temperatures.

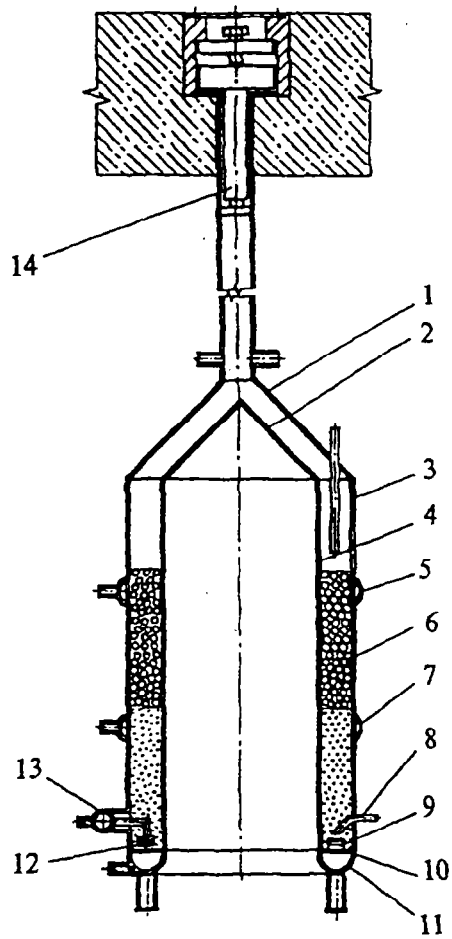

FIG. 1. Annular nuclear safe powder filled two-bed filter.
1,2 – bottom; 3,4 – shell; 5 – top distributing device; 6 – filtering charge (steel, corundum); 7 – middle distributing device; 8 – branch pipe; 9 – drain element; 10 – grid; 11 – annular bottom; 12 – distributing element; 13 – collector; 14 – device for charging filtration materials.

The search for the extractant structure resulted in finding and synthetizing compounds that conform to the conditions of the absolute homogeneous stability of extracts independent of the Pu(IV) content of initial solutions [6]. One of the specimens of the extractants of this type, namely, triisoamylphosphate (TIAP) was subjected to a long – term, commercial scale, and representative check at the RT-1 plant.

The other favourable features of TIAP comprise more than an order of magnitude lower solubility in aqueous solutions compared to that of TBP; higher hydrolytic and irradiation resistance, a little lower extractibility of some elements available in process solutions (Pd, Ru).

The investigations carried out by E.A.Filippov et al (VNIIKhT) have validated the alternative approach to the choice of an extraction system also resolving the problem of its complete homogeneous stability. In this option retaining the traditional extractant (TBP) of the «Purex» process the «light» paraffine diluent is replaced by the «heavy» hexachlorbutadien (HCBD). The favourable

TABLE I. COMPARATIVE CHARACTERISTICS OF EXTRACTANTS AND DILUENTS

Parameter	TBP	TIAP	RED[a]	HCBD
Formula	$(C_4H_9O)_3PO$	$(н\text{-}C_5H_{11}O)_3PO$	C_{10}-C_{15}	C_4H_6
Molecular mass	266,3	308,4		260,76
Density at 20°C, g/cm^3	0,975	0,952	0,75	1,68
Boiling temperature,°C	170(12mm Hg)	160(15mm Hg)	230-270	208-215
Flash point,°C	140	167	no	> 98
Ignition temperature,°C	160	167	no	> 104
Water solubility, g/l	0,39	0,02	<0,005	0,005
Maximal permissible concentration[b], mg/m^3	0,5	0,8	0,01	300

[a] fraction of saturated hydrocarbons with carbon atoms from 10 to 15 (basically)
[b] maximal permissible concentration in air of work rooms

feature of the TBP – HCBD system is its higher fire and explosion safety since HCBD has no flash point. This extraction system was also subjected to the rigid commercial check at the radiochemical plants in RF [7].

Thus, the scope of the implemented studies ensured the reliable choice of the extraction system for reprocessing high Pu content and high radioactivity materials. Some characteristics of the studied compounds are compared with the standard substances in table I.

The integral part of the work with the extractants was investigations of the radiation-chemical and hydrolytic transformations proceeding in them (M.V.Vladimirova, SSC VNIINM, G.F.Egorov IEL, RAN et al, also other RF Institutes).

For systems with macroquantities of actinides and fission products the data were acquired on the extraction kinetics, the influence exerted by the phase contact time on the purification from FP nuclides in high rate processes (SSC VNIINM, SSC FEI, NIKIMT etc). These investigations were focused on the application of centrifugal contactors.

2.5. Demonstration experiments

The important part of the investigations of the fast reactor fuel involved checking and mastering reprocessing models in the pilot facilities at SSC VNIINM, SSC FEI, SSC NIIAR using actual fuel materials of BR-10, BOR-60, BN-350. The maximul burn-up of the investigated fuel was 100 000 MW.day/ t; the minimal exposure was 7 months. The scope of the experiments covered dozens of kg of irradiated fuel. The data were acquired on the following technological options [8]: joint and individual reprocessing of core and blanket fuel; co-extraction of U and Pu and their co-stripping in the first extraction cycle followed by their separation in the second one; U-Pu separation in the first cycle. The process flow sheets were mastered using three types of the extraction equipment, namely, mixers – settlers, centrifugal contactors (fig.2) and pulsed columns. The typical coefficients of U and Pu purification from FP nuclides and the factors of the U isolation from Pu acquired in one of the experiments with the centrifugal contactors are tabulated in table II. The original papers contain information on the coefficients of purification from individual radionuclides; «the response» of their values to changes in the process conditions are considered.

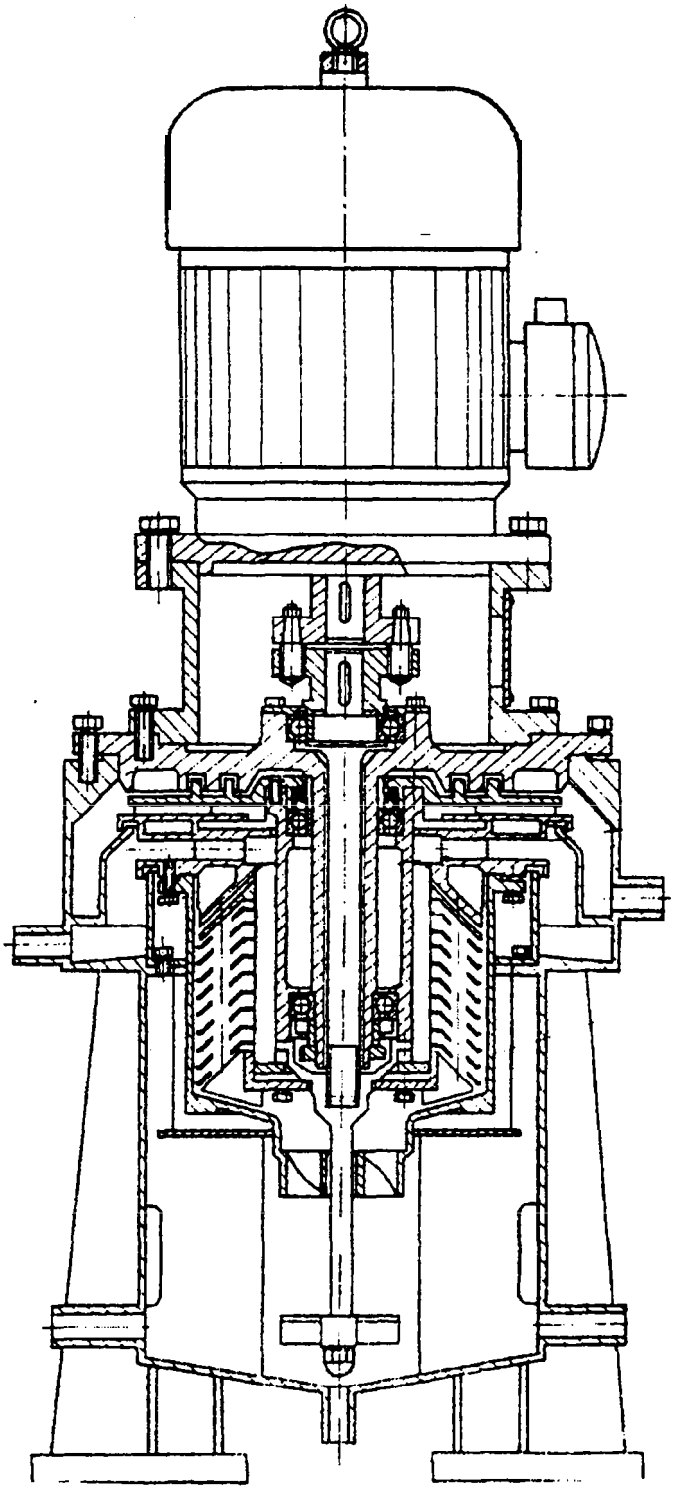

FIG. 2. *Centrifugal contactor [9].*

2.6. Final discussion

Lets put the question: what is the reasonable construction of the spent U-Pu fuel reprocessing flow sheet ?

The radiochemical plants reprocessing the nuclear power fuel currently in operation in RF, France, UK etc have automatically reproduced the ideology and general structures of the processes of the weapons Pu production. The involvement of civil Pu into the fuel cycle points to other approaches.

TABLE II. COEFFICIENTS OF U AND Pu PURIFICATION FROM FISSION PRODUCTS" (FP) AND SEPARATION[b]

Process stage	Coefficients of purification		
	U from FP	Pu from FP	U from Pu
1 St extraction cycle [c]	$1{,}2.10^5$	$1{,}2.10^5$	
2 nd extraction cycle [d]	$2{,}1.10^3$	$3{,}4.10^2$	$5{,}5.10^5$
Extraction process as a whole	$2{,}6.10^8$	$4{,}0.10^7$	$5{,}5.10^5$

[a] BN-350 fuel at the burn-up of 50 000 MW. day/ t and 2 year exposure.
[b] U-Pu separation in 2 nd extraction cycle.
[c] centrifugal contractors, phase contact time of 2 s.
[d] mixers - settlers.

The management of power generating materials requires protection of not only «the hot» part of chemical stages but also of all the subsequent stages up to the fuel shipment to NPP.

In case of the aqueous processes used to reprocess thermal reactor fuel it is not difficult to provide the coefficients of the U and Pu purification from fission products equalling $1\,0^7 - 1\,0^8$. As we have shown similar purification factors are also reached in fast reactor fuel reprocessing. The logic question will be whether those factors are superexcessive. It is to be noted that the «Purex» process by its first cycle guarantees the quantitative isolation from both fission products and stable components with the purification factors more than 10^5 for U and $1\,0^4$ for Pu. To-day no other technology is capable to compete with aqueous reprocessing in terms of those parameters (at the same time visualizing the future closed cycle we discuss the advisability of non-aqueous processes with their limited capability of purifying and isolating radionuclides).

We believe that the single – cycle «Purex» process is capable of resolving efficiently the problems related to U-Pu fuel reprocessing. This should be a separation cycle (fig.3). Its specific feature will be the formation of a U-Pu concentrate (product) at the specified ratio; it will be directly supplied to the co-precipitation and production of U and Pu oxide powders by the «GRANAT» process.

Here the problem lies in the fact that Np, the inevitable «satellite» of U and Pu, is available. It is known to be almost fully co-extracted with U and Pu. The possible resolutions are the selective stripping of Np from the organic phase after the Pu+U fraction is extracted; its co-stripping with residual (depleted) U. These options need further studies. The completeness of the separating operations will be promoted by equipping the extraction cycle with phase separators, similar in their design to powder filled filters mentioned above. The phase separators as applied to emulsions of «the oil in water)) type are capable of resolving one more problem, viz., to eliminate the non-control able migration of the extractant together with aqueous streams. This will ensure extra guarantees of nuclear and fire safety.

In our view, ,centrifugal contactors are the preferable type of the extraction equipment. With their use the amounts of materials processed in the streams are reduced (non-completed production); the degree of the radiation-chemical destruction of the extractant decreases .

Thus, with the sufficient functional abilities the single – cycle process of regeneration will allow:

a substantial reduction in the scope of aqueous waste;

the technological coordination of the initial (being reprocessed) and secondary (being produced) fuel.

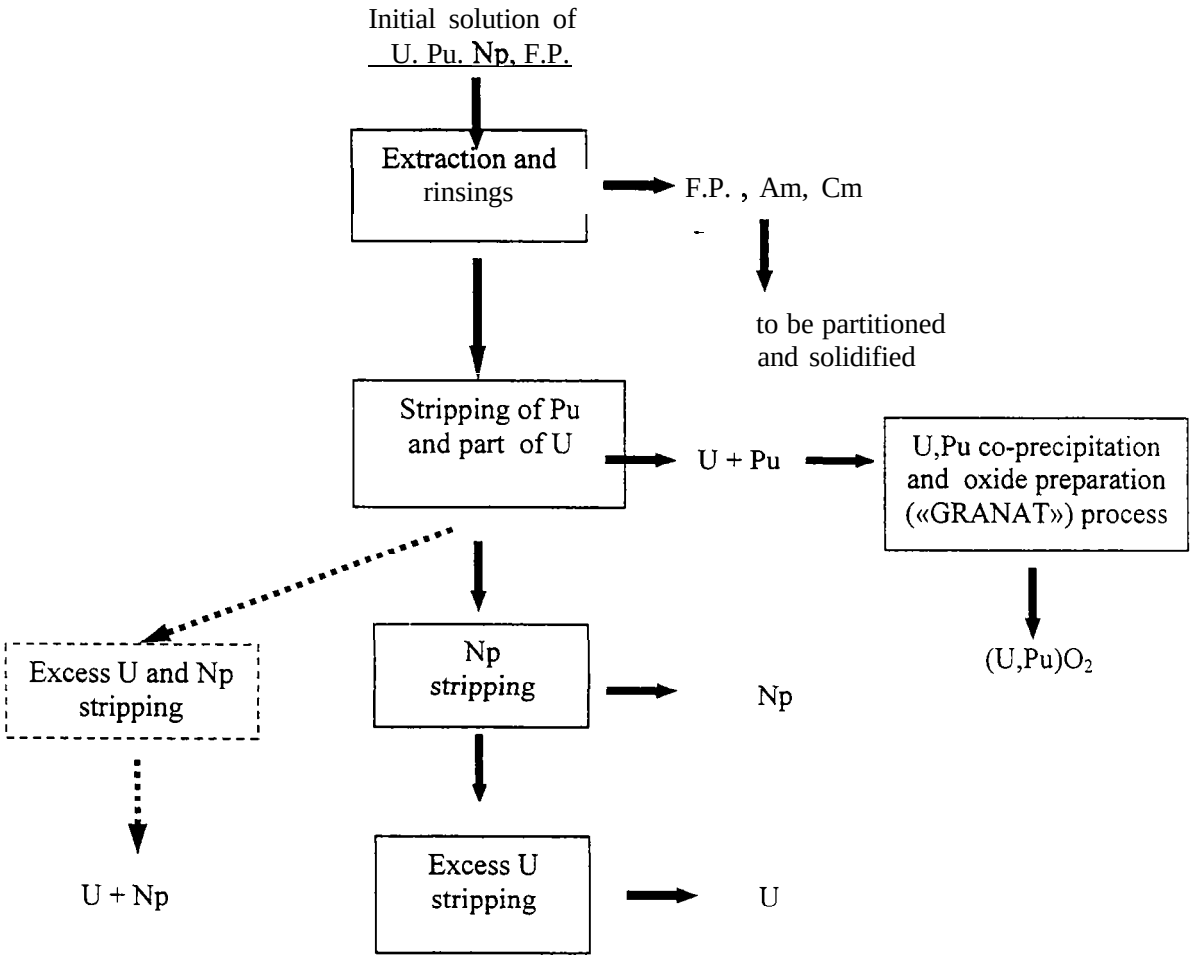

FIG.3. Single cycle U-Pufiel reprocessing outline.

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ADVANCED FUEL CYCLE ON THE BASIS OF PYROELECTROCHEMICAL PROCESS FOR IRRADIATED FUEL REPROCESSING AND VIBROPACKING TECHNOLOGY

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Abstract

For advanced nuclear fuel cycle in SSC RIAR there are developed the pyroelectrochemical process to reprocess irradiated fuel and produce granulated oxide fuel UO_2, PuO_2 or $(U,Pu)O_2$ from chloride melts. The basic technological stage is the extraction of oxides as a crystal product with the methods either of the electrolysis (UO_2 and UO_2-PuO_2) or of the precipitating crystallization (PuO_2). After treating the granulated fuel is ready for direct use to manufacture vibropacking fuel pins. Electrochemical model for $(U,Pu)O_2$ coprecipitation is described. There are being developed new processes: electroprecipitation of mixed oxides - $(U,Np)O_2$, $(U,Pu,Np)O_2$, $(U,Am)O_2$ and $(U,Pu,Am)O_2$. Pyroelectrochemical production of mixed actinide oxides is used both for reprocessing spent fuel and for producing actinide fuel. There are estimated both the efficiency of pyroelectrochemical methods application for reprocessing nuclear fuel and of vibropac technology for plutonium recovery.

1. Introduction

The current stage of nuclear power is characterized by enhanced requirements to safety and influence of all the fuel cycle stages on the environment. Involving weapon grade nuclear material in the fuel cycle results in additive requirements both to its non-proliferation and to the systems of physical shield and MC&A (Material Control and Accounting). That's why the technology, that will enable to raise considerably potential possibilities of oxide fuel and to upgrade technicoeconomic characteristic of the fuel cycle as a whole with the allowance for modem requirements are of great interest.

The following directions of the efficiency growth for nuclear power are becoming the most important:

- closed advanced fuel cycle, i.e. the internal closeness of technologic processes with the object to reducing the release of the substances, which are harmful for the environment;
- optimization-of the technologic systems, which is aimed at achieving the necessary maximum results by means of the minimum number of the process stages;
- the highest level of the inherent safety, i.e. the use of the processes, the safety of which is based not only on the engineering principles but also on the own «natural» properties of the technologic system, creating maximum grade of ecological protection.

These principles influence both general safety and economics equally.

It should be taken into consideration, that the processes, concerning nuclear fuel recovery and preparation, were formed as early as 1950s and were developing as the military technologies continuation. That is why many processes haven't been optimized yet from the viewpoint of the real public needs.

At IAEA symposium in 1997 there was emphasized, that fuel cycle facilities of the next generation should provide:

- Maximum high levels of the safety;
- Minimum costs on the fuel cycle;
- Minimum effect on the environment, including minimum waste generation;
- Minimum usage of natural resources;

- Minimum risk of proliferation and maximum guarantees;
- Public support;
- Variety and safety of energy supplies.

Nonaqueous methods are considered as possible alternative technologies for the fuel cycle. Pyroelectrochemical reprocessing, using the molten salts, is applied to oxide fuel (SSC RF RIAR, Russia) and metal fuel (Argon National Laboratory, the USA) and exhibits the following advantages:

- High chemical stability of the medium;
- High concentration in fission elements (more than 30 %);
- Unavailability of neutron moderators;
- Implementing all the chemical processes in the same apparatus independently of the kind of initial products being reprocessed (metals, oxides, nitrides and etc.);
- Minimized volume of high-active waste;
- Batchwise production from the viewpoint of control and accounting of fission components distribution.

By the moment SSC RF RlAR has formulated and substantiated experimentally chief principles of the advanced closed fuel cycle, based on mutual compatibility of the technologies for recovering uranium-plutonium fuel and manufacturing fuel pins and SA. The principles are as follows:

- using "dry" pyroelectrochemical processes with the aim of recycling irradiated fuel and, as a result of recovery, obtaining the oxide granulated fuel of polydispersive composition with the particle density, that is close to the theoretical one. The granulate is ready for vibropacking in the pin cladding;
- applying vibropacking with the object to making fuel pins and SA from granulated fuel;
- employing remote-control automated equipment in reprocessing fuel, manufacturing fuel pins and SA.

"Dry" techniques make it possible to reprocess fuel of any burnup and exposure time in few technological stage. The media, the processes proceed in , don't have moderator and therefore, the handling of fission and radioactive materials of high concentrations is possible. That is why «dry» processes are high productive, compact and result in small quantity of waste, which simplify considerably MC&A.

Remote-control processes are very important for fuel cycle, and extra charges, related to the transition from glove boxes to shielded cells, are substantiated due to the following reasons:

- there is an opportunity to optimize level of purification of FPs down to the one, meeting the requirements of reactor facility, followed by reducing charges for additional fuel purification and reprocessing of additional waste volume;
- rigorous restrictions on contents of high-toxic isotopes and transplutonium elements are removed;
- there are possibilities of working out new fuel cycle scenarios, for example:
  -it's possible to reuse fuel after partial reprocessing with aim of releasing gas and high-volatile fission products;
  -it's possible to use blanket zone fuel directly in BN Core;
  -it's possible to burn out fuel of the reactor in the other one with lower enrichment through partial fuel reprocessing;
- undeep NM purification is "less attractive", increases self-protectability level and enables to check easily NM movements, and therefore risk of their unauthorized usage decreases.

An experimental base for exercising the principles set forth above is Semi-Industrial Complex (SIC), including the Facility on granulated fuel production and the Facility of Fuel pin and SA manufacture. Tests of the fabricated fuel pins and SA take place in BOR 60 and BN 600.

2. Pyroelectrochemical technology of fuel reprocessing

Principal technologic flow-sheet of the pyroelectrochemical process of fuel reprocessing is given in Fig.2.1 .(a,b). Any kind of fuel (oxide, metal, carbide, nitride) can be an initial material for the process. At the first stage it is dissolved in the melt of salts, the most appropriate and investigated of which are chloride ones. Fuel is extracted from the melt either by electrolysis or by precipitating as crystalline oxides, for example UO_2, PuO_2 or $UPuO_2$. Upon separating the crystalline products from the salt-solvent and from other soluble impurities, the granulate of polydispersive composition, which is

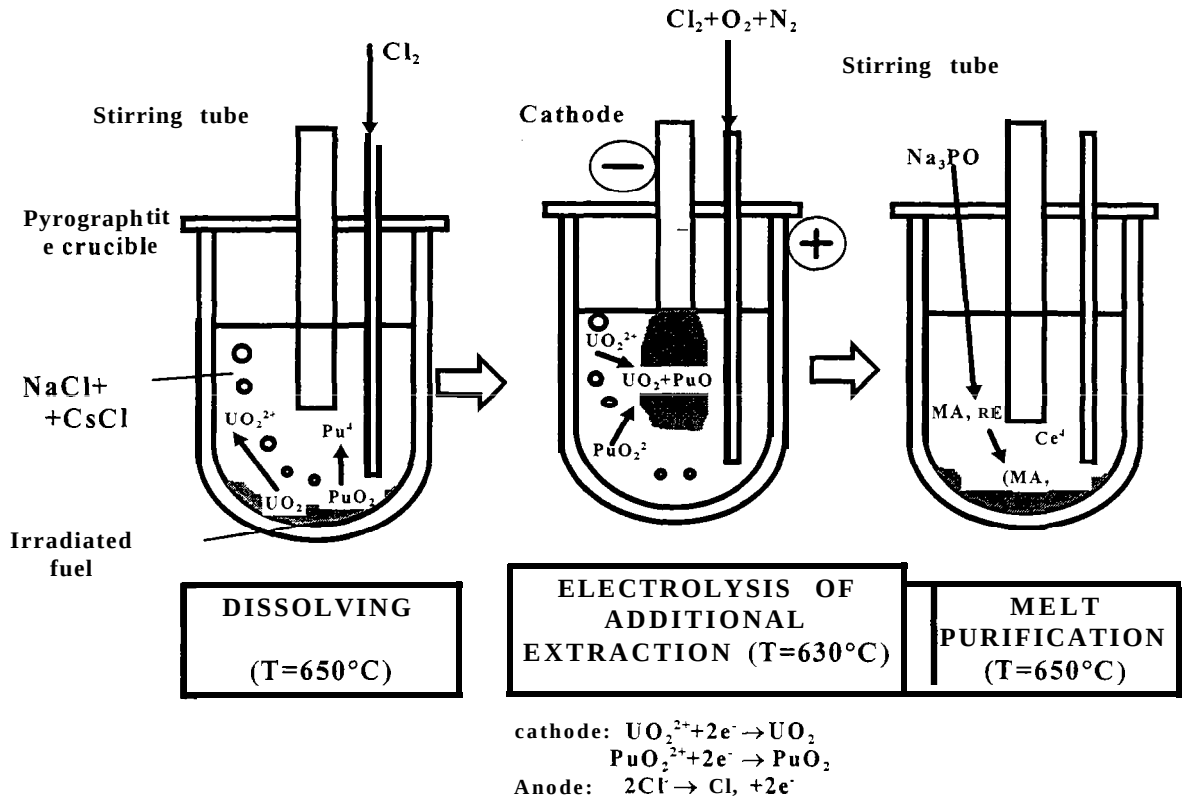

Fig.2.1.(a) Flow-sheet of pyroelectrochemical reprocessing of irradiated fuel under conditions of mixed fuel production

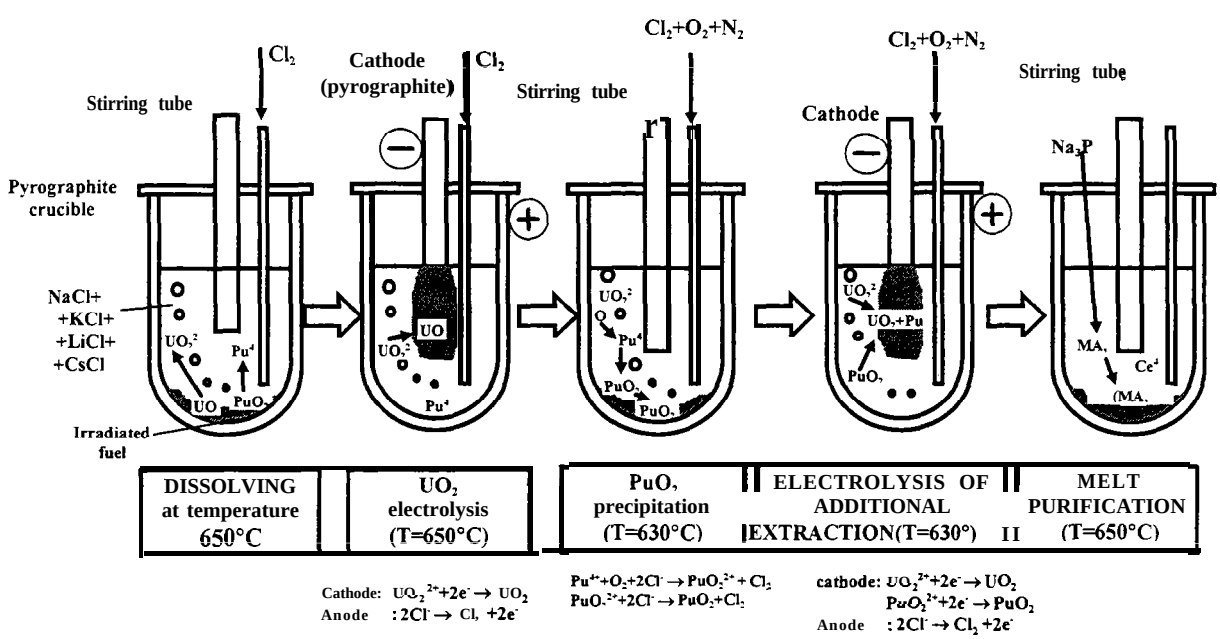

Fig.2.1 .(b) Flow-sheet of pyroelectrochemical reprocessing of irradiated fuel under conditions of plutonium dioxide production

suitable for vibropack fuel pin fabrication, is obtained. The particles have the density not less than 10,7 g&m' and their size is not more than 1,0 mm. Recovery of washing out solution is carried out by evaporating followed by returning the «dry» residue to the process beginning. Pyroelectrochemical process allows us to purify the fuel of FPs with total purification coefficient more than 100, which is sufficient from the viewpoint of reactor physics.

The waste volume is minimized, and a number of valuable elements (of ruthenium subgroup, for example) can be extracted from them. There are no rigorousrequirements to gas atmosphere for carrying out processes, so they are performed in the shielded cells under free air.

3. **Fuel pin and SA manufacture.**

Vibropacking technology is always considered as a way of fabricating the fuel column, enabling to reduce greatly the costs on the fabrication of fuel pins for nuclear reactor and to upgrade their performance. The principal merits of the vibropacking technology and vibropack fuel pins consist in

- simplicity and reliability of the production process related to the fewer number of technologic and check operations. This fact simplify automation and remote control of the process, therefore vibropacking technology can be employed to make pins of the recovered fuel in the shielded hot cells;
- the possibility of fabricating fuel column , having easily variable parameters, and on the basis of using multicomponent composition;
- the possibility of using any kind granulate: both of homogeneous composition and of mechanical mixture;
- less thermal-mechanical fuel effect on cladding in comparison with pellet fuel
- diminished requirements to inner diameter of fuel pin claddings.

The possibility of fabricating fuel pins by vibropacking is shown in two versions: hand - in the glove boxes and remote - in the shielded cells (Table2.1). Semi-industrial technology of fuel pin and SA manufacture for BOR-60, BN-3 50 and BN-600 reactors under remote conditions was implemented when working at «ORYOL» facility(1977 - 1986), SIC (1989-1997) and «Kolibry» mockup facility (1992-1997). At present there are operated two chains of glove boxes to fabricate fuel pins by vibropacking, which are used for carrying out different experimental programs and for producing fuel pins and SA of BOR-60 regular loading.

More than 25-year-old experience in manufacturing enabled to set forth the fundamental principles of the vibropack oxide fuel pin fabrication, to exercise the base technologic schedules, to choose the range of products and equipment performances. In sum there were manufactured about 30000 fuel pins, which became the base for mounting 730 fast reactor subassemblies.

During the pilot operation of the automatic line the yield of serviceable production was more than 98 %. In fabricating fuel pins manually at the technologic line, located in the shielded glove box (-1300 BN-350 fuel pins, -300 BN-600 fuel pins, -9000 BOR-60 fuel pins), the yield of serviceable fuel pins was 100 % almost in all quality parameters under check.

4.Waste handling.

Handling with waste and utilizable products, which are the result of granulated fuel, fuel pins and SA production technology can be divided into some processes:

- Reprocessing recycle products, arising from granulated fuel preparation and fuel pin fabrication.
- The processes of purifying gases being released of chlorine, high-volatile salts and radioactive aerosols.
- Purifying spent electrolyte (salt -solvant) of impurities.
- Burning and purifying combustible waste.

Besides there were developed the methods of extracting uranium and plutonium from the slime upon purifying the salt and the way of extracting U and Pu from the solutions after decontamination of high-radioactive equipment units.

All the NM content waste and utilizable products, arising from fuel and fuel pin fabrication, are placed into individual containers and sampled (if possibly) to analyze NM contents. Upon determining uranium and plutonium contents they are recycled in chlorator-electrolyzer followed by producing conditional fuel. Before starting the recovery, the waste are accumulated and stored in the shielded cell.

Evaluative data on waste amount are presented in table 4.1 and 4.2

Table 4. Il

Solid radioactive waste in fabricating MOX-fuel at SIC (estimation at annual capacity 1000 kg of fuel)

Waste title	**waste yield, kg/kg fuel**	**Waste amount, kg per year**	**Fission material content, g/kg of waste**	
			Pu	U
High-active products, being sent to Storage (in increasing effective output almost 1000 kg will be reprocessed to extract Pu)				
Concentrate upon purifying salt	0.0 13	13	0,02 %	0,08 %
Tatters, cleaning and packaging materials	0.02	20	0,033 %	0,040 %
High-active waste, being shipped to burial				
equipment made of pyrographite	0.2	200	0.4	1.6
Filters	0.13	130	0.7	1.5
Low-active solid waste, being shipped to burial				
Units of metal equipment, aids of check and remote operation (after decontamination).	0.7	700		

Table 4.2

Liquid radioactive at SIC (estimation at annual capacity 1000 kg of fuel)

Waste title	**Specific activity, Cu/l (Bk/l)**	**Quantity, l/per year**	**Product content, g/l**	
			Pu	**U**
The solution after ÓÐÏÕ*	3.1×10^{-6} (1.15×10^{5})	2×10^{5}	5×10^{-5}	1.5×10^{-4}
Dissorbent solutions	8.7×10^{-9} - 1×10^{-7} $(3.2\text{-}37) \times 10^{2}$	12×10^{5}	$1.4 \times 10^{-7} \div$ 1.6×10^{-6}	4.2×10^{-7} $t4.8 \times 10^{-6}$

*ÓÐÏÕ - facility of recirculation and chlorine absorption.

S.Analysis of safety of fuel cycle processes on the basis of «dry» pyroelectrochemical fuel reprocessing and vibropacking technology.

The perspective direction of nuclear fuel cycle safety elevation is a transition to the technologies, which possess a high level of «inherent» safety. In other words, besides engineering barriers of safety in these technologies, the properties of the systems and processes themselves are natural barriers and minimize harmful effect on the environment both under normal and under extreme conditions. The process of nuclear oxide fuel pyrochemical recycling in the chlorides melt and the technology of remote fuel pin fabrication by vibropaking can be ascribed to the enhanced safety technologies.

5.1 Radiationsafety

Vibropacking and pyrochemical technologies have a number of features, which reduce the yield of active substances in movable phase (gas or liquid):

- Fuel of pyrochemical origin exists as a crystalline product. Crystalline PuO_2 (of energetic isotope composition) generates 1.5×10^3 as less aerosols, than analogous product does after oxalate precipitation and annealing. As a result, decontamination of the facilities and shielded cells after work with granulate takes less time than it does after handling of powder.
- In reprocessing there are used the molten mixture of alkaline metal chlorides (for example NaCl-KC1 or NaCl-2CsCl- ionic liquids). While dissolving all the fuel components pass into cloride form, but the properties of their solutions in ionic liquids are so that vapors volatility decreases by several orders of magnitude lower. This reason prevents radioactive elements from going out of chemical reactions zone (except for high-volatile substances $SbCl_3$, $TeCl_3$ and etc.). In an emergency the melt solidifies and transmutes into a salt monolith, which doesn't generate aerosols, and in contacting with water the melt slowly dissolves off the surface.
- As the systems deal with concentrated materials, liquid and solid medium-active and low-active waste are practically absence. High-active waste (HAW) has FP high concentration and leads to the necessity of diluting them. The dilution can be combined with HAW introduction in the stable matrix to be buried finally.

To estimate internal safety of the technologic processes there are used the criteria, characteristic of which are given in Table 5.1. The data are the results of

- large-scale experiments on uranium-plutonium oxide (with energetic plutonium) fuel production by pyroelectrochemical way and on pilot SA/fuel pin fabrication for BN type reactor by vibropacking,
- experimental recycling of irradiated uranium and uranium-plutonium oxide fuel at the pilot facilities.

The interval of numerical criteria values for waste are due to the differences in their compositions and in their reprocessing. A high radiological danger along with both energetic plutonium and actinides (Np, Am, Cm) toxicity demanded, that two level of the personnel and environmental protection should be envisaged when realizing the recycle.

- The first level includes the sealing of technologic equipment, transport and transfer containers, the usage of pneumo-mail.
- the second level includes - the sealing of boxes and cells; two-stage purification of the air, carried off out of them; usage of unfailed electromechanical manipulators; the system of shielded cell and equipment decontamination.

Employing technologic processes with high internal safety and additional safety barriers enabled to implement large-scale (-500 kg) plutonium recycle under minimum radiation effect on the personnel and the environment. Average joint personnel dose was 0.065 person*Zv/per year; and maximum dose of man-caused effect didn't exceed 0.2 % dose of natural background. It is shown, that the content of plutonium and transplutonium elements in the environmental objects hasn't increased and is at the global background level. For example, plutonium content in soils is 0.5 - 3.0 Bk/kg, in the free air - $(0.4 - 1\,1)*10^{-7}$ Bk/m^3.

Table 5.1

Criteria of estimation of internal safety for fuel cycle technologic processes

No	Criteria	Dimensionality	Value
1	Direct plutonium yield in ready product	%	99,3 - 99,7
2	Plutonium ratio in the products, returned in technological cycle	%	0,2 - 0,6
3	Irretrievable loss	%	≤ 0,015
4	HAW yield	kg/kg of fuel	1,0
5	HAW radiation stability	W/ñm^3 gr	3,5 $3{,}5 \cdot 10^7$
6	HAW thermal stability (up to vitrification)	^{0}Ñ	500 - 900
7	HAW chemical stability (up to vitrification): • leachability in ^{137}Cs • pass in gas phase ^{137}Cs	 g/(ñm^2•days) g/(ñm^2*days)	 $1 \cdot 10^{-5}$ - $1 \cdot 10^{-4}$ $1 \cdot 10^{-11}$ - $4 \cdot 10^{-8}$
8	Activity outlet in aerosols (alpha-radiator)	% of fuel activity	$< 1 \cdot 10^{-3}$
9	Volume activity of alpha-nuclides under conditions of shielded cell and glove box atmosphere in handling energetic plutonium	Bk/l	< 37,0

These conclusions are verified with safety implementation of some experiments on the irradiated fuel reprocessing:

- with short exposure time - 6-7 months (1972 , UO_2-fuel of ÁÎÐ-60 reactor, burnup 7.7%),
- with high burnup - 21-24% (1995, ÌÎÕ-fuel of ÁÎ D60 reactor with the exposure time for 3.5 and 2.5 years).

5.2 *Nuclear safety underpyrochemical reprocessing*

Using the salt melts and unavailability of aqueous solutions excluded neutron moderators and reflectors from the technological system. The fact, that uranium and plutonium are only in two states - oxide and chloride enable to work in the same apparatus with large quantity of fission materials:

- up to 39 kg of uranium dioxide with 90% enrichment in U-235 before and after dissolving in chlorator-electrolyzer
- up to 30 kg of energetic grade plutonium dioxide before and after dissolving in the chlorator-electrolyzer.

Nuclear safety in operating is provided with the following technologic decisions:

- the solvent of fuel components - the salt melt exists in liquid state only under great apparatus heating, which excludes the possibility of solvent's random transfer to other crucibles, in which the conditions for FM accumulation and SCR (self-chain reaction) occurrence could be arisen;
- processes are carried out batch-by-batch and FM quantity control is performed by weighing without chemical analysis;
- the technology of recycling makes it possible to exclude completely water application, because the process of vacuum topping is developed to remove the salts from the products. (Sometimes the operation is carried out in the presence of limited water and spirit quantity, at that FMs are in crystalline state, they are not dissolved and dispersed, therefore there are no conditions to reduce FM critical mass.)

5.3Technical and chemical safety of reprocessing

Technical and chemical safety are due to the technical decisions, worked out in studying high-temperature processes, using chlorine and salt melts, for a long time. The following decisions are the principal ones:

- Chemical medium (the melt of salts) neither is subjected to radiolysis, nor generates hydrogen and detonating gas.
- Low fire risk is provided with unavailability of appreciable quantity of organic substances and reagent-oxidizers in the technologic processes. The equipment is located in the rooms of limited volume and faced with metal coating. Local combustion of cables, rubber hoses within the cells can be easily eliminated either by inert gas or by dry salt.
- The process doesn't use explosive gases and substances, when blending there were no generated explosive mixtures.
- Drastic poison substance - chlorine - is employed restrictedly, is delivered in balloons of 60 l capacitance. Chlorine is supplied from the reservoir of reduced down to 0,2 Mpa gas pressure by batch. There have been carried experimental investigations to provide in future sharp volume reduction of the chlorine being used by recycling with cryogen technology.
- Applying dried air in the cells under much higher temperatures than dew point minimizes equipment corrosion due to chlorine gas. The conclusion is supported by long experience in working under shielded cell condition.

5.4 Safety from the viewpoint of non-proliferation criteria

Radiation characteristic of the product, obtained in reprocessing irradiated oxide fuel by pyrochemical way, diminish the probability of its unauthorized usage owing to low factors of purification and separation and, therefore, high radiation intensity.

Fig.3.1 shows comparative experimental data on oxide fuel activity of BOR-60 reactor before and after reprocessing. It is obvious, that recovered fuel remains in the limits of irradiated fuel standard on the radiation characteristic. Due to high fuel activity any fuel movements are easy to monitore, which decrease greatly the probability of its embezzlement.

The technologic cycle is defended against unauthorized interference with some barriers:

- The first, «engineering» barrier, is the technologic equipment itself, in which FMs are stored, and the shielded cells, the equipment is located in.
- Another specific barrier is chemical stability of the pyrochemical product. The experiment proves, that pyrochemical PuO_2 is almost insoluble in aqueous acid solution. In order to separate it either in PUREX-process or in an analogous system there is required special development of the method and the equipment to solve pyrochemical PuO_2.
- The third «barrier» is the chief principle of fuel handling - discreteness, which gives us two merits:
    - in technologic line there is carried out treating fuel batch and products, so simple check by weighing and monitoring facilitate all the inspection procedures.
    - unavailability of liquid flows in the system and reservoir-storage allows us to follow FM position at any point of technologic line at each instant time.

As for vibropacking technology it should be noted, that the process has fewer operations to control FMs in comparison with the technology of pellets and fuel pins fabrication. As a whole, the technologies, developed in SSC RF RIAR, not only facilitate and simplify FM control and accounting, but also make them (MC&A) much cheaper.

Thus, in terms of comprehensive safety, pyroelectrochemical recycling and vibropacking technologies enable to implement a new elevated level of fuel cycle safety in FM recycle field.

There is posed a reasonable question: whether these investigations are sufficient for commercial introduction? It can be answered positively. And in terms of historic analogies we have to state, that the gained experimental experience agrees with the investigation scope, which preceded commer-

cial introduction of up-to-date fuel cycle technologies, such as PUREX-process, UOX-pellet technology, pellet MOX-fuel technology.

The work results of the last few years allow us to give a new look at the data massif, which is compiled in SSC RF RIAR and is realized at Semi-Industrial Complex. In our opinion, the Complex can serve as a flexible model of pilot plants on precommercial adaptation both of dry reprocessing technology and vibropacking one. One of the variants, using SIC model, is «nuclear island», including the facility of fuel recycle and 2 fast reactors. From primary estimations we can suppose, that this Complex will be competitive with VVER type reactors, and simultaneously the problem of radioactive waste will be solved.

6.CONCLUSIONS

Thus, complex of "dry" technologies and granulated fuel vibropacking in combination with unique properties of vibropack fuel pin enables to realize a new comprehensive approach to the fuel cycle problem, including the one of using different grade plutonium. Mass tests of vibropack uranium-plutonium oxide fuel pins in BOR-60 reactor, successful tests of experimental SA in BN-600, reliable operation of SIC facilities allow making a conclusion on a real possibility of developing safety, profitable uranium-plutonium fuel cycle on the basis of the set forth technologies, and recovering energetic and weapon grade plutonium in nuclear reactors under providing the reliable MC&A system as well.

EFFECTS OF HIGH BURNUP AND LONG TERM STORAGE OF LWR SPENT FUELS ON FUEL CYCLE SCENARIOS

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Abstract

We have analyzed the effects of high bum-up and long-term storage of spent fuels of light water reactors on fuel cycle scenarios from the viewpoint of mass balance, radioactivity and decay heat of spent fuels, plutonium, trans-uranium, high level waste and natural uranium savings. We have used a reactor design code coupled with the isotope generation and depletion code ORIGEN2 in order to calculate the nuclide compositions and characteristics of nuclear materials for each uranium oxide or mixed oxide fuel with discharge bum-ups of 33, 45, 55, and 70 GWd/t. The trans-uranium, radioactivity and decay heat per tonne initial heavy metal (tIHM) inventory of spent fuels increase with their bum-up in general. However, those quantities per generated energy (TWh) of spent fuels are almost independent of bum-up, because of reduction of spent fuel arising due to high bum-up. We have compared two scenarios: Sl (only storage of spent uranium fuels) and S2 (reprocessing of spent uranium fuels, recycling of plutonium in a light water reactor, and then storage of spent mixed oxide fuels, high level waste and reprocessed uranium). The scenario average quantities (TRUE inventory, radioactivity, and decay heat) per tIHM are found to increase with respect to bum-ups for both scenarios. However, quantities per TWh decrease or almost constant with respect to bum-up. The scenario S2 consumes about one third of plutonium contained within spent uranium fuels, by which natural uranium is saved by 5-10%. The rate of saving of uranium decreases as bum-up increases, If there is no delay for reprocessing, the scenario average decay heat is about 10% larger in S2 than in Sl. As the reprocessing delays, it becomes larger in S2 than in Sl for smaller (-15 years) cooling period of MOX-SF. However, the differences between Sl and S2 almost disappear, if its cooling period becomes larger (-60 years).

1. INTRODUCTION

The projected elongation of LWR era indicates that the bum-up of uranium oxide (UOX) fuels for LWRs will be extended further for reduction of spent fuel (SF) accumulation and for better fuel cycle economy. Under these circumstances in Japan, for example, some of SF will be reprocessed and the rest will be stored continuously as useful resources containing plutonium (Pu) and others. All the separated Pu recovered by reprocessing SF is assumed to be recycled immediately into LWRs as mixed oxide (MOX) fuels. Generally speaking, however, higher bum-up results in lower quality of recovered fissile materials and larger quantities of fission products (FP) and trans-uranium (TRU) isotopes, whereas prolonged periods of storage reduce ^{241}Pu and other unstable nuclides. Therefore higher bum-up could lead to lower credit for Pu and reprocessed uranium (RU) and higher reprocessing costs including high level radioactive waste (HLW) disposal. However, the reprocessing costs could be alleviated by the longer-term storage of SF.

2. OBJECTIVE

The main stream of the fuel cycle scenarios before the introduction of commercial fast breeder reactors (FBR) is the storage of LWR-SF or/and the utilization of Pu in LWRs. We analyze the effects of high burn-up and long-term storage of LWR-SF on fuel cycle scenarios before the FBR era from the viewpoint of mass balance, radioactivity and decay heat of SF, Pu, TRU and HLW and natural uranium savings. We also study the effect of burn-up of MOX fuels on these scenarios.

3. DESCRIPTION OF SCENARIOS AND CALCULATION METHODS

3.1. Definition **of two scenarios**

We assume two scenarios: Sl and S2:

- The scenario Sl corresponds to 'SF-storage option'. It uses only UOX fuels and does not reprocess spent UOX fuels (UOX-SF) which are stored indefinitely. The number of UOX loaded reactors (UOX-LWRs) is n+l . The scenario S2 defines the number 'n'. We evaluate the effects of high burn-up and the storage period dependence of SF arising, radioactivity and decay heat for LWR-UOX plant of 1000 MWe.
- The scenario S2 corresponds to 'plutonium-recycle option'. It reprocesses UOX-SF after a specified storage period (SF cooling years: T,) and separates HLW, and RU and Pu, the latter of which is recycled as MOX fuels. The scenario S2 stores RU for simplicity instead of recycling it. The capacity of LWR-MOX plant is also 1000 MWe. The preceding 'n' plants of LWR-UOX of the same capacity supply the necessary and sufficient quantity of Pu for the MOX fuels.

Fig. 1 describes the schematic relations between two scenarios. Each LWR unit generates equal electricity. Therefore, both scenarios generate also equal electricity. S2 comprises n units of uox-LWRS (UOX-LWR1, uox-LWR2, ---, and UOX-LWRn) plus a MOX-LWR whose fuels are all MOX made of Pu supplied by the preceding n units of UOX-LWRs. However, the number n needs not an integer. Quantities of the shaded boxes are averaged for each scenario. The quantity T_c is the cooling period (year) for UOX-SF until reprocessing. The quantities T_l and T_{res} are respectively, the lead time for Pu to be loaded into the LWR as MOX fuels and the fuel residence time. We start the evaluation after the discharge of MOX-SF at which the origin of time is set: t=O.

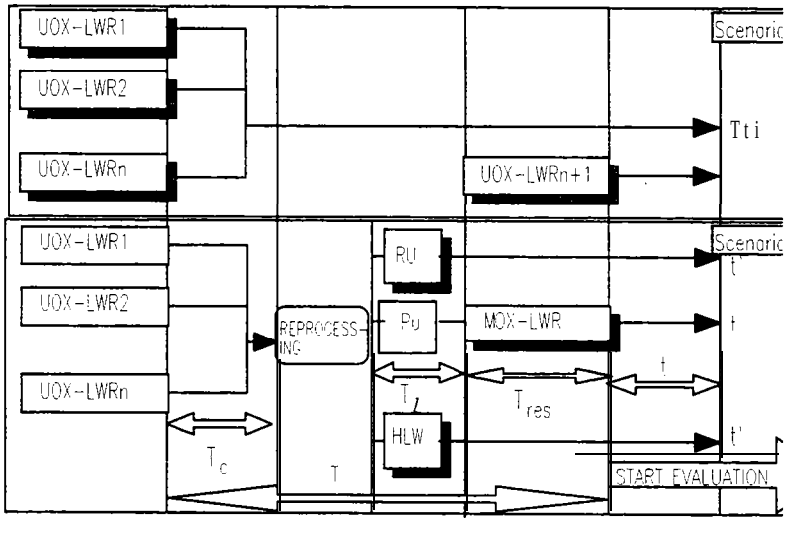

FIG. 1. Definition of two scenarios SI and S2.

3.2. Method of calculation

3.2.1. *Burn-up calculation code and fuel assembly*

Here we choose a current BWR/5 of 1100 MWe gross capacity for a representative of LWR. All the quantities such as SF arising are normalized to LWR of 1000 MWe. We have performed burn-up calculations using the neutronics calculation code [l] of a reactor design for each UOX- or MOX-LWR fuel with discharge burn-ups of 33,45, 55, and 70 GWd/t, respectively.

As the assembly design code has only limited number of nuclides which are important for the neutron balance calculation, we have also used the versatile computer code ORIGEN2 [2] coupled with it [3] for calculating the nuclide compositions and characteristics of nuclear materials. The isotope generation and depletion code 0RIGEN2 uses the effective one-group cross section library based on the assembly averaged neutron spectrum derived by the assembly design code at each time step. We have used ENDF/B-IV, V and JENDL-3 for the nuclear data library. This procedure improves the reliability of calculated fission product yields and TRU isotopes in higher burn-ups.

The BWR fuel assemblies for discharge burn-ups 33 GWd/t and 45 GWd/t are respectively 8x8 or 9x9 arrays. As the assembly design for the burn-ups of 55 GWd/t and 70 GWd/t is not yet determined at present, a 9x9 array is assumed for them. Although enrichment is different for each fuel rod in an actual BWR assembly, we assume the equal enrichment. Average void fraction is assumed to be 40 % within the channel box.

3.2.2 *Assumptions*

We explain the assumption for each fuel cycle step in Fig. 1. The relationship among UOX, HLW and MOX are summarized in Table I. The meaning of each fuel ID are given in the following.

Table I. RELATIONSHIP AMONG UOX FUEL, HLW AND MOX FUEL

UOX fuel	u3	u 4	U5	u 7
Initial enrichment (wt %)	3.0	3.8	4.6	6.0
Discharge burn-up (GWd/t)	33	45	55	70

Cooling period (years) before reprocessing: Tc	A 5	A 5	B 15	A 5	B 15	C 60	A 5	B 15	C 60
HLW	W/3A	W/4A	W/4B	W/5A	W/5B	W/5C	W/7A	W/7B	W/7C
Discharge burn-up (GWd/t) of MOX fuel 33	M3/3A	-	-	-	-	-	-	-	-
45	M4/3A	M4/4A	M4/4B	-	-	-	-	-	-
55	-	M5/4A	-	M5/5A	M5/5B	M5/5C	-	-	-
70	-	-	-	M7/5A	-	-	M7/7A	M7/7B	M7/7C

3.2.2.1. UOX fuel

The UOX fuel is made of enriched natural uranium. The enrichment is 3.0,3.8,4.6 and 6.0 wt %, respectively for the discharge burn-up 33,45, 55 and 70 GWd/t in Table I. Each fuel is named as U3, U4, U5 and U7 only for convenience. The enrichment tail is 0.2 wt % ^{235}U. The tail uranium is used as matrix for MOX fuel.

The enrichment of ^{235}U in UOX or fissile Pu in MOX fuel is determined respectively for 12, 15, 18 and 23 effective-full-power-month operation in an equilibrium cycle of a BWR/5. It is important to note that the enrichment of the U3 or U4 fuel is based on the commercial achievements. However, that of U5 or U7 is under development, and is determined so as to give the same infinite multiplication factor as the U3 or U4 at the burn-up equal to the end of equilibrium cycle of the UOX-LWR core. It is also to be noted that the initial enrichment of U7 is larger than 5%, which requires some license renewals.

3.2.2.2. Storage of UOX-SF and Tc (years of cooling UOX-SF until the reprocessing)

There are three cases for Tc. We choose (A) five years for a standard value of Tc, (B) fifteen years corresponding to the approximate half-life of ^{241}Pu, and (C) sixty years for more than 90 % decay of ^{241}Pu into ^{241}Am.

3.2.2.3. Reprocessing of UOX-SF and HLW

The reprocessing of UOX-SF is performed after Tc. Then we recover Pu and RU as resources. The HLW produced during reprocessing is the main part of the radioactive waste arising in the whole fuel cycle processes except for the SF itself. It is reported [4] that 97.6 % of the total non- volatile FP, 99.5 % of the total MA (minor actinides), 0.12 % of the total Pu and 0.15 % of the total U are contained within the vitrified HLW. Based on this report, we assume the vitrified HLW contains:

- 0.1 % of Pu and 0.2 % of U;
- 100 % of FP except for ^{3}H, ^{14}C, ^{85}Kr and ^{129}I;
- 100% of MA.

Other wastes arising during reprocessing is neglected. As radioactivity of reprocessed uranium is small enough compared with HLW, it is neglected. The relationship between UOX-SF and HLW is summarized in Table I. For example, HLW produced by reprocessing the SF of U5 after five years of cooling is denoted as `W/5A`.

3.2.2.4. Fabrication of MOX fuel and Operation of MOX plants

Table I also shows the fuel relationship between UOX and MOX. The lead time T_l from the recovery of Pu to the loading of MOX fuels into the LWR is fixed to be 2 years. We have neglected MOX fuel fabrication loss only for simplicity.

The MOX-LWR plant is also the BWR/5 of 1100 MWe class, the size of which is normalized to 1000 MWe. The MOX plant is assumed to be a full MOX core. The enrichment of MOX fuels is determined to give the same infinite multiplication factor at EOC as the corresponding UOX fuels. The MOX fuel ID is M3, M4, M5, and M7 according to the discharge burn-up 33,45, 55 and 70 GWd/t, respectively in Table I. The origin of Pu for each MOX fuel is also identified like M5/4A, whose Pu is supplied by reprocessing the spent U4 fuels after five years of cooling.

4. RESULTS OF CALCULATION

Before presenting the results for two scenarios, we show characteristics of UOX-SF and MOX-SF in the first two sections.

4.1. Effect of high burn-up on each SF

In order to analyze the effect of high burn-up of UOX fuels, we assume a fixed value for SF cooling period $T_c = 5$ years before reprocessing UOX-SF and equality of burn-ups between MOX and UOX fuels. Therefore the MOX-LWR has exactly the same operating conditions as UOX-LWR.

We calculate material balance of selected isotopes, radioactivity and decay heat of UOX-SF, HLW, and MOX-SF, respectively. These are evaluated for the initial heavy metal inventory (tlHM) or for the total energy generation (TWh).

4 . 1 . 1 . UOX fuels

We compare characteristics of UOX-SF of U3, U4, U5 and U7 given in Table I.

4.1.1.1. Material Balance

Fig. 2a shows the burn-up dependence of the TRU inventory (kg/tIHM) in the UOX-SF at five years after discharge. Generally speaking, amount of TRU of higher atomic mass increases with burn-up. However, the tendency is different for indivisual isotopes:

(1) The fraction of fissile Pu (Puf) decreases with burn-up.
(2) The fraction of Np amounts to half of the total MA. The ratio of Am to total MA has a decreasing tendency with respect to burn-up.
(3) The ratio of MA to Pu increases with burn-up.

Fig. 2b shows the same quantities as Fig. 2a. They are expressed in terms of kg/TWh. We find a tendency different from Fig. 1 a:

(1) The total heavy metal inventory decreases with burn-up. The decrease of fuel heavy metal weight per TWh implies the reduction in the SF storage and reprocessing costs.
(2) Total Pu and Puf in SF decreases with burn-up.
(3) The ratio of MA with respect to Pu increases with burn-up. Total MA increases with burn-up. The rate of its increase is about 10 % for the 50 % increment of burn-up. Especially, fractions of Np and Cm increase while those of Pu and Am are decreasing with burn-up.

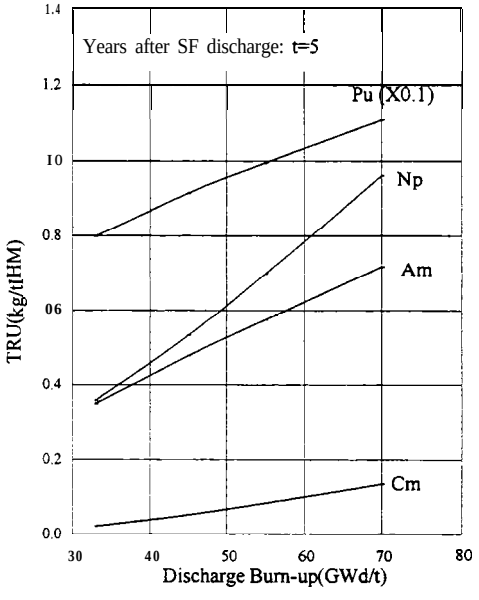

FIG. 2a. Dependence of TRU inventory/tIHM on dicharge burn-up of each UOX-SF (5 years after discharge)

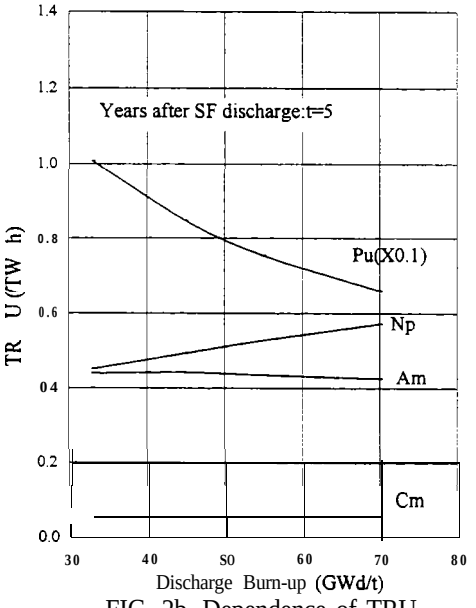

FIG. 2b. Dependence of TRU inventory/TWh on discharge burn-up of each UOX-SF (5 years after discharge)

4.1.1.2. Radioactivity and decay heat

We have calculated the radioactivity and decay heat of each spent UOX fuel as a function of the time 't' elapsed after the discharge of SF. The total radioactivity is contributed almost by FP, especially ^{90}Sr (half-life 29 years)- ^{90}Y and ^{137}Cs (half-life 30 years)- ^{137m}Ba). The half-lives of these isotopes governs the decay character of the total radioactivity. The radioactivity per TWh decreases as the burn-up increases.

Fig. 3a and 3b show the burn-up dependence of decay heat of UOX-SF per tIHM or per TWh, respectively. The contribution to the total decay heat is mainly by FP. However, the contribution due to actinides becomes larger for longer years after SF discharge. Decay heat per TWh is almost constant with respect to burn-up.

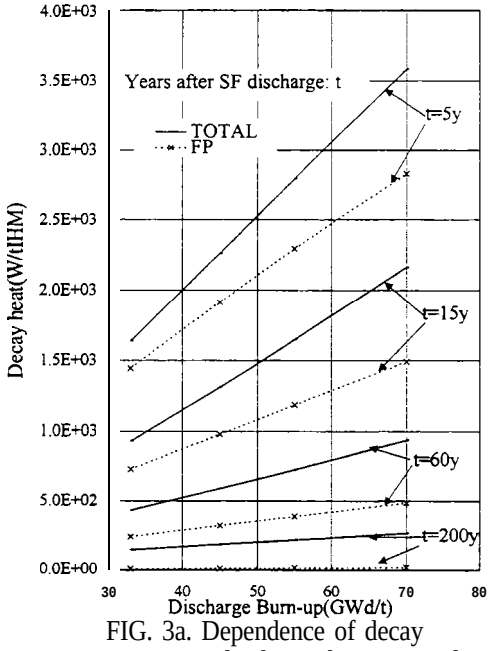

FIG. 3a. Dependence of decay heat/tIHM on discharge burn-up and UOX-SF cooling period t.

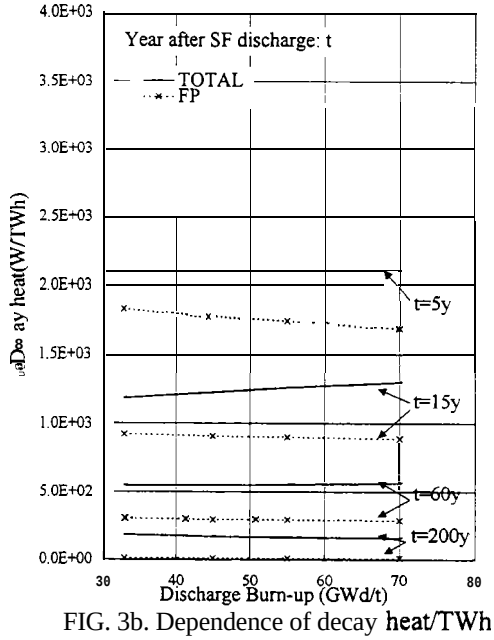

FIG. 3b. Dependence of decay heat/TWh on discharge burn-up and UOX-SF cooling period t.

4.1.2. *HLW and MOX fuels*

We have calculated material balance, radioactivity and decay heat of HLW: W/3A, W/4A, W/5A and W/7A given in Table I. The total radioactivity and decay heat of HLW are mainly contributed by FP just like the UOX-SF. The burn-up dependence of the radioactivity per TWh decreases. Decay heat per TWh is almost constant with respect to burn-up.

The burn-up dependence of material balance, radioactivity and decay heat of MOX fuels: M3/3A, M4/4A, M5/5A and M7/7A are also calculated. Figures 4a and 4b show the burn-up dependence of the TRU inventory (kg&MM) in the MOX-SF at five years after discharge.

As far as we want to design a MOX fuel with burn-up equal to UOX fuels which supply Pu to the MOX, it is necessary to enrich more Pu in higher burn-up MOX fuel because of less Puf. This fact is reflected in Fig. 4a. However, Fig. 4b indicates that Pu inventory is almost independent of the discharge burn-up, when it is measured in terms of energy generation (TWh).

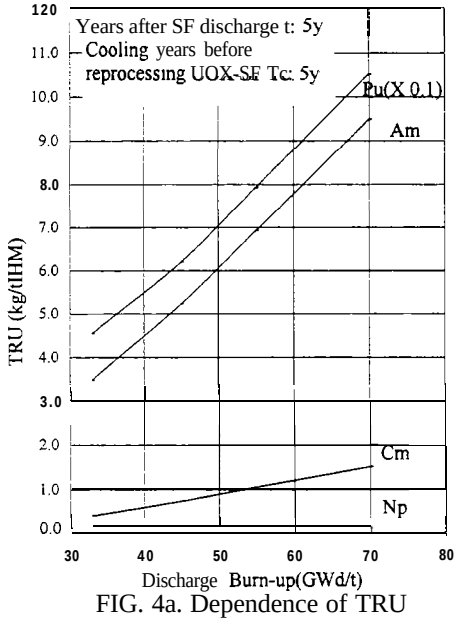

FIG. 4a. Dependence of TRU inventory/tIHM on dicharge burn-up of each MOX-SF (5 years after discharge)

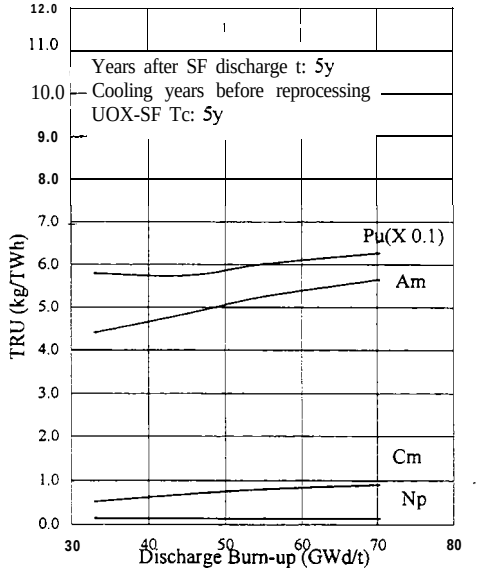

FIG. 4b. Dependence of TRU inventory/TWh on dicharge burn-up of each MOX-SF (5 years after discharge)

Figures 5a shows the decay heat of MOX-SF, expressed in terms of tIHM. Total decay heat is mainly due to actinides. The contribution of FP is rather small compared with them. It is important to note the present assumption that Pu for high burn-up MOX fuel is supplied by the same high burn-up UOX-SF. Fig. 5b shows that the dependence of decay heat per TWh on burn-up behaves moderately.

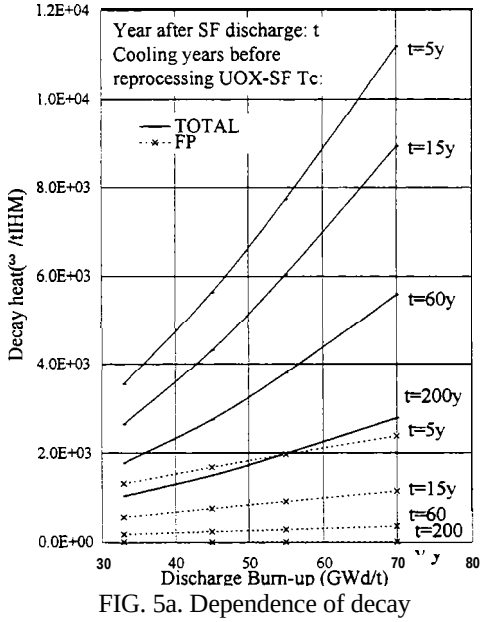

FIG. 5a. Dependence of decay heat/tIHM on dischargeburn-up and MOX-SF cooling period t.

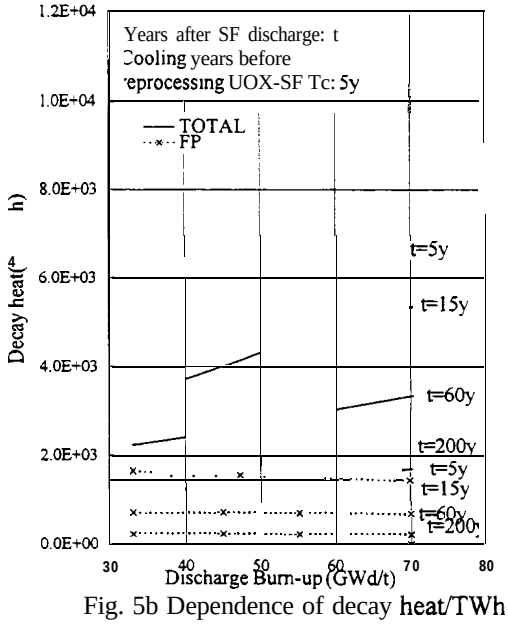

Fig. 5b Dependence of decay heat/TWh on discharge burn-up and MOX-SF cooling period t.

4.2. Effect of long-term storage on each SF

In order to study the effect of long-term storage of UOX-SF, we vary Tc (UOX-SF cooling years before its reprocessing). We compare characteristics of MOX-SF for M4/4B, M5/5B and M7/7B (Tc=15 years) and M5/5C and M7/7C (Tc=60 years) with MOX-SF of Tc=5 years. Fig. 6 shows the TRU inventory of each MOX-SF. As the storage period of UOX-SF becomes longer, more decays of ^{241}Pu require more enrichment of Pu in MOX fuel. On the other hand, the inventory of ^{241}Am in MOX-SF becomes less, because it is generated by the beta-decay of ^{241}Pu during irradiation. Fig. 7 shows the decay heat of each MOX-SF. The longer storage period of UOX-SF results also in more decays of ^{238}Pu, the half-life of which is 87.7 years. Therefore, the inventory of ^{238}Pu in MOX fuel becomes less.

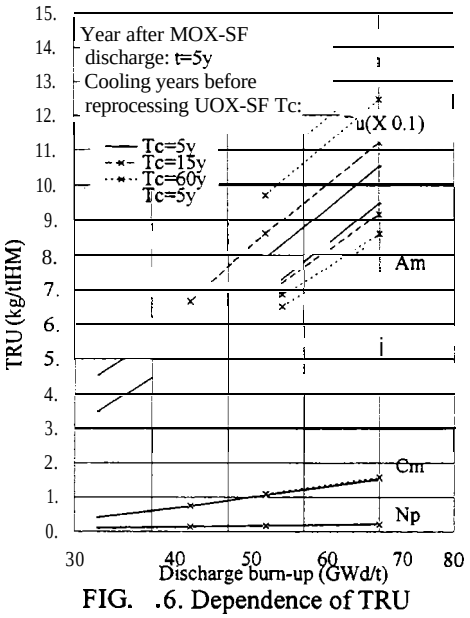

FIG. .6. Dependence of TRU inventory/tIHM of MOX-SF on discharge burn-up and Tc of UOX-SF

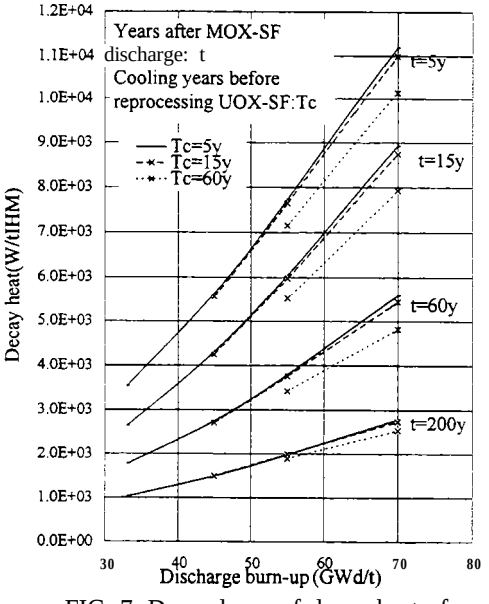

FIG. 7. Dependence of decay heat of MOX-SF on discharge burn-up and Tc of UOX-SF

4.3. Evaluation of scenario average quantities

In the practical point of view, MOX fuels can replace only limited fraction of all the fuels for LWRs, even if reprocessing all the UOX-SF is performed. In order to understand this situation, we calculate the scenario average quantities for isotopic inventories, radioactivity, and decay heats for each scenario S 1 and S2 shown in Fig. 1. The scenario average quantities Zl for scenario Sl are only those of UOX-SF. They are defined by Zl = (n*Ul + U2)/(n+l). Here, Ul is corresponding quantities of UOX-SF of the preceding UOX-LWR, and U2 is those of the n+l st UOX-LWR which is operated in the same period as MOX-LWR. The similar quantity Z2 is defined for the scenario S2 by Z2 = (n*Wl + M2)/(n+l). Here, Wl is those of HLW and RU, and M2 is those of MOX-SF. The scenario average quantities Zl and Z2 are expressed either for initial heavy metal inventory (per tHM) or for power generation (per TWh). The time scale t is defined in Fig. 1. It is the elapsed time since the discharge of MOX-SF (or UOX-SF of the n+lst UOX-LWR). The relations between t and others are also defined in Fig. 1.

4.3. I. Number of UOX plants for recycling of Pu for a single MOX plant

The number 'n' of the preceding UOX-LWRs to make up a single MOX-LWR of the equal electricity generation capacity is an increasing function of burn-up and cooling period Tc. It is 7.96, 9.82, 11.67 and 14.10, respectively for M3/3A, M4/4A, M5/5A and M7/7A at Tc = 5 years. It becomes 12.92 (M5/5B) and 15.39 (M7/7B) at Tc = 15 years, or 15.14 (M5/5C) and 17.86 (M7/7C) at Tc = 60 years. The degree of reduction in UOX plant in the scenario S2 is measured by l/(n+l), which is of the order of 5-10 %. This is equivalent to the natural uranium saving by Pu recycling.

4.3.2. *Scenario average TR U inventory and decay heat*

Fig. 8 shows the burn-up dependence of the scenario average TRU inventory per TWh at Tc = 5 years. It indicates that about one third of Pu is consumed by recycling.

We compare the scenario average decay heat per TWh for each scenario in Fig. 9 also at Tc = 5 years. Comparison of Fig. 3a with Fig. 5a indicates that the decay heat of MOX-SF is about three times larger than that of UOX-SF as far as the cooling period is rather short. However, the scenario average value shows at most 10 % difference at t = 5 years. They are almost independent of discharge burn-ups. For cooling period smaller than about 15 years, the main contributors to the decay heat of MOX-SF are ^{244}Cm (half-life 18.1 years) and ^{238}Pu, while ^{137}Cs and ^{90}Sr are predominant in UOX-SF. The difference between S 1 and S2 disappears after t=60 years.

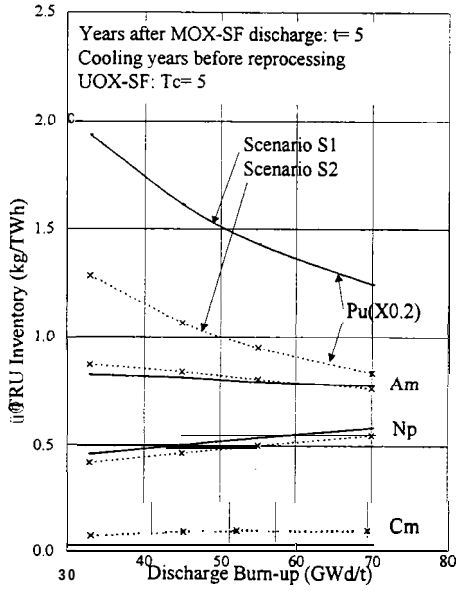

Fig. 8 Scenario average TRU inventory per TWh (Tc=5y,t=5y)

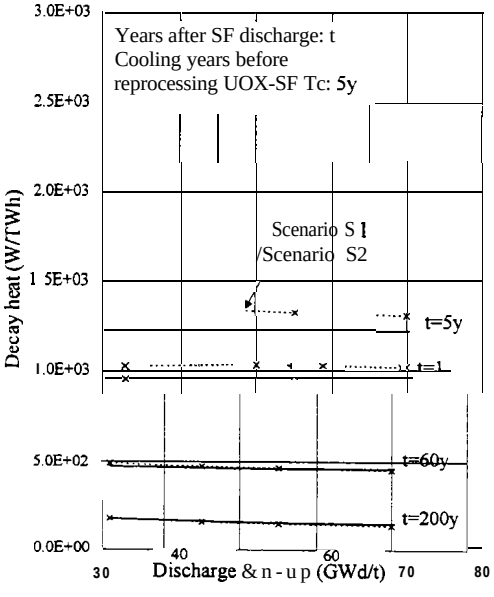

FIG. 9. Scenario average decay heat per TWh (Tc= 5 years).

5. SUMMARY AND CONCLUSIONS

We summarize qualitatively the effects of high burn-up and long-term storage of UOX-SF on nuclide compositions and characteristics of nuclear materials in TABLE II. Individual character and quantity of each material affect the detailed processes at each step of fuel cycle scenarios including their economy and environmental impact.

The results and conclusions are summarized as follows:

1) We have used the versatile computer code ORIGEN2 coupled with a reactor fuel design code for calculating the nuclide compositions and characteristics of nuclear materials in high burn-up fuels.

TABLE II. EFFECTS OF HIGH BURN-UP AND LONG-TERM STORAGE ON NUCLIDES

	High burn-up of UOX fuel	Long-term storage of SF
Fresh UOX	^{235}U >5 wt % is required	
UOX-SF	^{235}U < 0.711 wt % ^{236}U = or > ^{235}U; ^{232}U increases Pu increases but Puf/Pu decreases ^{237}Np increases, then ^{238}Pu increases Am and Cm increase FP increases	Decay of ^{241}Pu to ^{241}Am Decay of ^{238}Pu Decay of FP
HLW	Increase of Np, Am and Cm	More ^{241}Am transfers mto HLW
Fresh MOX	Contents of Pu need to be increased Contents of ^{238}Pu increase	Less ^{241}Pu results in less ^{241}Am Contents of Pu need to be increased Contents of ^{238}Pu decrease
MOX-SF	Puf/Pu decreases ^{238}Pu increases	^{241}Am and ^{238}Pu decreases

2) The TRU inventory, radioactivity and decay heat per tIHM of each SF increase with discharge burn-up in general. However, those quantities per TWh of each SF are almost independent of it, because of reduction of SF arising due to high burn-up.

3) As the storage period of UOX-SF becomes longer, more decays of ^{241}Pu require more enrichment of Pu in MOX fuel. On the other hand, the inventory of ^{241}Am in MOX-SF becomes less, because it is generated by the beta-decay of ^{241}Pu during irradiation. The longer storage period of UOX-SF results also in more decays of ^{238}Pu, the half-life of which is 87.7 years. Therefore, the inventory of ^{238}Pu in MOX fuel becomes less.

4) We have compared two scenarios: S 1 (only storage of UOX-SF) and S2 (reprocessing of UOX-SF, recycling of Pu in an LWR, and then storage of MOX-SF, I-ILW and RU). The scenario average quantities (TRU inventory, radioactivity, and decay heat) per tIHM are found to increase with respect to burn-ups for both scenarios. However, quantities per TWh decrease or almost constant with respect to burn-up.

5) The scenario S2 consumes about one third of Pu contained within UOX-SF, by which natural uranium is saved by 5~10%. The rate of saving of uranium decreases as burn-up increases.

6) If there is no delay for reprocessing, the scenario average decay heat is about 10% larger in S2 than in S 1. As the reprocessing delays, both quantities become larger in S2 than in S 1 for smaller (- 15 years) cooling period of MOX-SF. However, the differences between S 1 and S2 almost disappear, if its cooling period becomes larger (-60 years).

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[3] NUCLEAR DATA NEWS (ISSN 0385-4876: an internal informal newsletter written in Japanese) "SPINOZA: an ORIGEN 2 scenario coupled with TGBLA to correct neutron-spectrum", JAERI, February 1996, 1 l-20.

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REACTOR-BASED PLUTONIUM DISPOSITION: OPPORTUNITIES, OPTIONS, AND ISSUES

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Abstract

The end of the Cold War has created a legacy of surplus fissile materials (plutonium and highly enriched uranium) in the United States (U. S.) and the former Soviet Union. These materials pose a danger to national and international security. During the past few years, the U. S. and Russia have engaged in an ongoing dialog concerning the safe storage and disposition of surplus fissile material stockpiles. In January 1997, the Department of Energy (DOE) announced the U. S. would pursue a dual track approach to rendering approximately 50 metric tons of plutonium inaccessible for use in nuclear weapons. One track involves immobilizing the plutonium by combining it with high-level radioactive waste in glass or ceramic "logs". The other method, referred to as reactor-based disposition, converts plutonium into mixed oxide (MOX) fuel for nuclear reactors. The U. S. and Russia are moving ahead rapidly to develop and demonstrate the technology required to implement the MOX option in their respective countries.

U. S. MOX fuel research and development activities were started in the 1950s, with irradiation of MOX fuel rods in commercial light water reactors (LWR) from the 1960s – 1980s. In all, a few thousand MOX fuel rods were successfully irradiated. Though much of this work was performed with weapons-grade or "near" weapons-grade plutonium – and favorable fuel performance was observed – the applicability of this data for licensing and use of weapons-grade MOX fuel manufactured with modern fuel fabrication processes is somewhat limited.

The U. S. and Russia are currently engaged in an intensive research, development, and demonstration program to support implementation of the MOX option in our two countries. This paper focuses on work performed in the U. S. and provides a brief summary of joint U. S./Russian work currently underway.

1. INTRODUCTION

Over the last several years, the U. S. and Russia have engaged in an intensive dialog relating to the management and storage of surplus fissile materials in the two countries. In September 1993, the U. S. committed to eliminate surplus fissile materials. The following January, President Yeltsin and President Clinton issued a joint statement limiting the use of fissile materials and initiating joint studies regarding long-term disposition of plutonium. Several meetings and agreements have been made since that time with the latest, "Joint Statement on Principles for Management and Disposition of Plutonium", issued in September 1998. This statement declared the intent of Russia and the U. S. to each disposition 50 MT of military plutonium.

A decision has been made in the U. S. to pursue a dual approach to plutonium disposition, which utilizes immobilization and irradiating plutonium as mixed oxide (MOX) fuel in commercial nuclear power reactors. Several factors contributed to this dual-approach decision:

Two technologies provide some assurance that the mission will continue if any unforeseen technical, schedule, cost, or institutional obstacles are encountered. No domestic or international consensus exists concerning the best technical approach. Russia has expressed a concern that immobilization does not destroy plutonium and leaves it as weapons grade material that could be used for possible weapons reuse. Plutonium disposition as MOX fuel provides the best opportunity for the U. S. to work with Russia and other countries to reduce Russia's excess plutonium.

The goal of the U. S. plutonium disposition effort is to achieve the "Spent Fuel Standard'. The Spent Fuel Standard states that surplus plutonium will be made as inaccessible and unattractive for retrieval and weapons use as the residual plutonium in spent fuel from commercial reactors. The primary characteristics associated with the Spent Fuel Standard are the material's radiation field, plutonium concentration, chemical form, size and weight, and storage location.

Currently, the U. S. plans to complete the 50 MT plutonium disposition campaign by the year 2022. Achievement of this goal will require the construction of a Pit Disassembly and Conversion Facility (to extract the plutonium from weapons components and convert it to plutonium oxide feed material), and a MOX Fuel Fabrication Facility. The U. S. Department of Energy (DOE) is currently negotiating with Raytheon Engineers & Constructor, Inc. for the preliminary and detailed design of the Pit Disassembly and Conversion Facility. Construction of the facility is expected to begin in 2001 with startup in 2006.

The Department has contracted with Duke Engineering & Services, COGEMA, Inc., and Stone & Webster (collectively known as "DCS") to provide mixed oxide fuel fabrication and reactor irradiation services in support of the department's mission to dispose of surplus weapons plutonium. DCS will design, provide construction management services, operate and deactivate a weapons-grade mixed oxide fuel fabrication facility in the U. S. The team will also modify six existing U. S. commercial light water reactors at three sites to irradiate mixed oxide fuel assemblies. These reactors sites are Catawba in York, South Carolina; McGuire in Huntersville, North Carolina; and North Anna in Mineral, Virginia. The consortium will be responsible for obtaining a license to operate the fuel fabrication facility and the license modifications for the reactors from the Nuclear Regulatory Commission (NRC). DOE is selecting a contractor to provide these fabrication and irradiation services in parallel with determining the location for the fuel fabrication facility.

The department is preparing a Surplus Plutonium Disposition Environmental Impact Statement that analyzes the potential environmental impacts associated with establishing plutonium disposition facilities at DOE sites. Those sites are: the Hanford Reservation near Richland, WA; the Idaho National Engineering and Environmental Laboratory near Idaho Falls, ID; the Pantex Plant near Amarillo, TX; and the Savannah River Site near Aiken, SC. In June 1998, DOE announced that the Savannah River Site was the preferred site for the mixed oxide fuel fabrication facility. The Record of Decision on this environmental review is expected this summer.

2. UNIQUE CHARACTERISTICS OF WEAPONS-GRADE PLUTONIUM

The technical issues relating to reactor-based plutonium disposition stem directly from the differences between reactor-grade (RG-) plutonium and weapons-grade (WG-) plutonium. Weapons-grade plutonium contains more Pu-239, less Pu-240, and less Am-241 than reactor-grade plutonium. While neither the U. S. or Russia has made a final decision regarding the weapons disassembly and plutonium conversion and purification processes to be employed in the disposition mission, the U. S. has not ruled out the use of new dry Pu conversion and purification processes. These processes could lead to differences in weapons-grade plutonium oxide powder morphology and impurities (relative to commercial reactor-grade plutonium). All the conversion processes under consideration in the U. S. are dry processes. The U. S. will employ a hydride-dehydride process for disassembly of the weapons components and extraction of the plutonium metal.

Metal-to-oxide conversion processes under consideration at the present time include both direct metal oxidation and a multi-step process in which the plutonium metal is first converted to a nitride and then to an oxide. The U. S. has evaluated the use of both dry thermal plutonium purification processes, and traditional aqueous polishing techniques for removal of trace elements such as gallium. The best available data in the U. S. suggests that the use of dry processes for conversion and purification will yield gallium concentrations in the fabricated MOX fuel of approximately 0. l-l .O parts per million. (For purposes of_comparison, the fission yield of gallium in existing LEU and RG- MOX fuel is on the order of **0 . 1** parts per billion.)

3. U. S. LWR MOX FUEL EXPERIENCE

U. S. MOX fuel experience is substantial but dated, because LWR MOX fuel development work was halted in the late 1970s following a Presidential Executive Order banning fuel recycle. U. S. MOX fuel research and development activities were started in the 1950s with irradiation of MOX fuel rods in commercial reactors from the 1960s – 1980s. Table 1 provides details of the irradiations that were conducted on the MOX rods.

Table 1. U. S. LWR MOX Irradiation History

Reactor	**Dates of Irradiation**	No. of **MOX Assays (rods)**	**Burnup (MWd/Mt) Max Avg. Assay (Peak MOX Pellet)**	**Examin-ations**	**Comments**	**Data Utilization**
Ginna (PWR)	1980-1985	4 (716)	39,800 (?)	None	Assemblies intact (82% fissile Pu)	FY99 spent fuel exam plan
Quad Cities-l (BWR)	1975-1980s	5 **(48)**	39,900 **(57,000)**	D & ND core phys	Well documented EPRI (80 & 90% fissile Pu)	Neu benchmarks constructed and analyzed
Big Rock Point (BWR)	1969-late 1970s	53 (1248)	-20,000 est (30,200)	D & ND	Little documentation	No current plans
San Onofre–1 (PWR)	1970-1972	4 (720)	19,000 (23,500)	Some D	PIE documents found	FY99 neu analysis plan
Dresden-l (BWR)	1968-early 1970s	15 (103)	-19,000 (-14,000)	–	Little documentation	No current plans
Saxton (PWR research reactor)	1965-1972	9 (638)	Many reconstitu-tions (51,000)	Fuel perf D and physics tests	Relatively well documented fuel perf data (9 1.4% fissile Pu)	Critical experiments analyzed and reported
Misc. Test Rx (Exp BWRs) (Pu Re Test Rx) (Mat Test Rx) (Eng Test Rx)	1960s-1970s	1000s of rods	?	Variety of D	Capsules and rods irradiated. Little historical research conducted	No current plans

Abbreviations:
D – destructive, ND – non-destructive, Neu – neutronics, est – estimated, perf – performance, Rx – reactor, Exp – experimental, Re – recycle, Mat – material, Eng, – engineering

A few thousand MOX fuel rods were successfully irradiated in the U. S. Plutonium isotopics used for the tests included near weapons-grade compositions. The fuel performance was found to be equivalent to contemporary LEU fuel performance. Reviews conducted by the NRC, and documented

in NUREG-0002, concluded there were no significant health and safety impacts to the public of MOX fuel fabrication or reactor operations.

Critical experiments were conducted at the Critical Reactor Experiment facility at the Westinghouse Reactor Evaluation Center. These experiments included 44 lattice configurations of

MOX and LEU rods involving both single and multiple regions. MOX lattice pitches ranged from 1.32 to 2.64. The MOX rods contained 6.6 wt % PuO_2 with 90% ^{239}Pu in the plutonium. The plutonium rods were then irradiated in the Saxton reactor. As an extension of the Saxton critical experiments, critical experiments were conducted at ESADA to consider variations in ^{240}Pu. Variations included 2 wt % PuO_2 in natural uranium with 8% ^{240}Pu and 24% ^{240}Pu. Single region experiments were conducted to evaluate buckling, reactivity worth, and power distribution. Multi-region experiments were conducted to evaluate reactivity worth, power distribution, and lattice pitches. In addition concentric regions and "salt and pepper" arrangements were evaluated.

Additional critical experiments were conducted at Pacific Northwest National Laboratory (Battelle). Six lattices of MOX rods with 2 wt % PuO_2 with 8% ^{240}Pu in water, and six lattices of UO_2 were constructed. Results of the study are documented by EPRI in *Clean Critical Experiment Benchmarks for Plutonium Recycle in LWRs*, report number EPRI NP-196. The ANS Cross Section Evaluation Working Group has adopted this work as a benchmark.

4. U. S. MOX LICENSING CONSIDERATIONS

The U. S. regulatory review focus and process is dictated by 10CFR50, and is facilitated by U. S. NRC Regulatory Guidelines, NUREG-0800 (NRC's Standard Review Plan), and NRC Generic Letter 88-20 (which relates to the execution of Probabilistic Risk Assessments and Individual Plant Risk Examinations). The focus of the U. S. regulatory review process is to ensure:

1. no fuel damage can occur during normal operations and anticipated transients;
2. fuel damage during a design basis accident cannot proceed to the point of preventing control rod insertion;
3. a coolable core geometry is maintained during design basis accidents; and
4. the radioactivity release during a design basis accident is not underestimated.

Three basic questions will need to be answered as a part of the U. S. regulatory review of WG-MOX use in commercial reactors:

- Are safety margins significantly reduced?
- Is the probability or consequence of any previously analyzed accident increased?
- Does the use of MOX fuel create the possibility of new types of accidents?

In order to address these questions, the U. S. licensee will need to demonstrate that the thermal, mechanical, and physics performance of weapons-grade MOX fuel is equivalent to LEU fuel, and that analytical methods used to demonstrate fuel performance are as accurate as current methods used for LEU fuel.

The U. S. is currently transitioning to "risk-informed regulation." In July 1998, the U. S. Nuclear Regulatory Commission issued Regulatory Guide 1.174, "An Approach for Using Probabilistic Risk Assessment in Risk-Informed Decision on Plant-Specific Changes to the Licensing Bases." As recently stated by Shirley Ann Jackson, Chairman of the Commission [Nuclear News, January 1999],

> "This Regulatory Guide provides a general framework for plant-specific NRC decisions that have been requested and initiated by licensees. It sets forth the Commission-approved principles for NRC staff evaluation of such proposals, including expectations for application of the Commission Safety Goal Policy,reliance on traditional defense-in-depth approaches, and maintenance of sufficient safety margins when initiating changes to the licensing bases. In addition it provides criteria for the scope, level of detail, and quality the PRA supporting the licensee submittal."

In addition to the traditional deterministic safety case reflected by lOCFR50, it is clear that reactor licensees who intend to employ WG-MOX fuel will need to demonstrate that the impacts of WG-MOX fuel use are acceptable from the overall risk standpoint, including beyond design basis accidents.

Thus the U. S. reactor owner will be required to demonstrate a thorough understanding of the impacts of WG-MOX substitution on overall plant performance and safety. Demonstration of the safety case will rest upon the availability of data and validated computational tools. Where reliance is made on commercial (reactor-grade) MOX fuel data, it will, of course, be necessary to show that the data is both applicable to weapons-grade MOX fuel and sufficient to support the safety case. However, based upon the established track record of favorable fuel performance in the commercial MOX industry, there is high confidence that WG-MOX fuel can be successfully licensed in the U. S.

5. U. S. R&D

Given the abundance of favorable international commercial MOX experience, the U. S. reactor-based plutonium disposition research and development program has focused on issues that derive from the unique characteristics of weapons-grade MOX: (1) isotopics, (2) dry-processed oxide powder morphology, and (3) impurities.

Reactor physics studies have provided a better understanding of the impact of weapons-grade plutonium on MOX fuel performance. An integral part of the current work is the international ARIANE project, which includes irradiation of RG-MOX fuel samples at Dodewaard in Holland, and at Beznau and Goesgen in Switzerland; with examination of the irradiated samples at CEN in Belgium, PSI in Switzerland, and TUI in Germany. Both BWR and PWR LEU and MOX data are being obtained at multiple burnups, with the intent of expanding the database of on fission products and actinides yields during irradiation. Potential applications of this data include reactor physics, accident analysis, reactor operations, spent fuel management, and spent fuel shipping.

Figure 1 provides a comparison of the predicted actinide inventories (as calculated by SCALE and HELIOS-2) of the Dodewaard BWR RG-MOX fuel. The uncertainty in the burnup of the test fuel is estimated to be up to 4%. The sensitivity of the U-235 concentration to burnup is 2.5; (i.e. an uncertainty of 1% in burnup translates to an uncertainty of 2.5% in the U-235 concentration). For Pu-239 the sensitivity to burnup is about 2.0. Pu-239 is also sensitive to the large concentration of U-238 and thus any uncertainty in the U-238 capture cross section is magnified. Am-241 and Am-242m agreements are poor. The latter nuclide, however, has a very low concentration (high measurement uncertainties) and the former is complicated by the Pu-241/Am-241 ratio in the fresh fuel. The calculated-to-measured agreements for the curium isotopes are considered reasonable.

While the HELIOS results compare much more favorably than the SCALE calculations, the *trends within* the HELIOS results are similar to those in SCALE and suggest that uncertainties in cross section data are driving the major observed discrepancies between computed and measured data. It is not surprising that the HELIOS results compare more favorably with the data than the SCALE results. SCALE is designed to provide assembly-average information. However, the MOX fuel samples analyzed in this program were extracted from fuel assemblies in which a few MOX fuel

rods were surrounded by LEU rods. Additional data on PWR MOX fuel will be forthcoming from the ARIANE project in the near future.

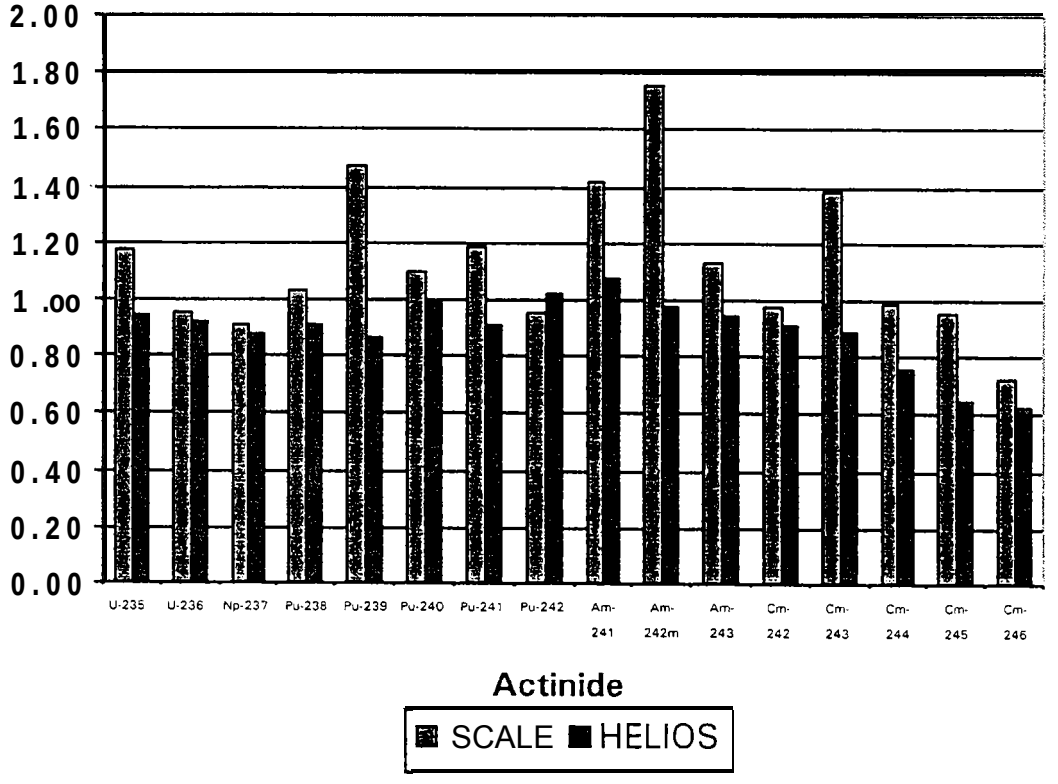

Figure 1. Comparison of SCALE and HELIOS predictions for actinide inventories to ARIANE Dodewaard BWR RG-MOX fuel data.

Figure 2 is a pictorial representation of comparison between measured data and HELIOS-2 calculations for Quad Cities-l BWR MOX fuel lead assembly described in Section 3. The results of these analyses indicate that the maximum uncertainty in calculated pin powers is similar for LEU and MOX fuel pins. The results suggest that the host reactors should not have to operate at derated conditions in order to utilize the WG-MOX fuel.

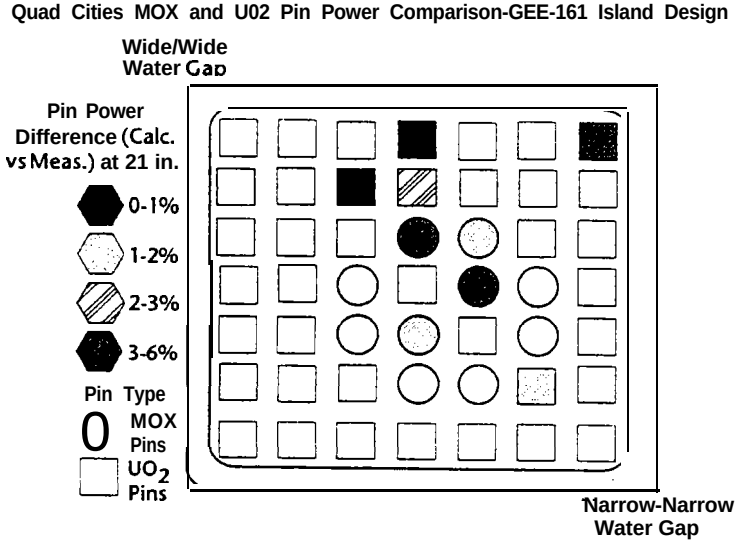

Figure 2. Comparison of HELIOS-2 calculations for Quad Cities BWR MOX fuel bundle pin powers to measured data.

A coordinated test program is underway to characterize the impact of residual gallium in MOX fuel. The intent is to gain an understanding of the gallium transport mechanisms, reactions, kinetics, and damage mechanisms. A two-phase out-of-reactor test program was conducted to determine general and separate effects. Table 2 summarizes the test parameters for the study. The results of the limited testing performed to date indicate that intermetallic compound formation is the only potential gallium/cladding corrosion mechanism. No evidence of either grain boundary corrosion or liquid metal embrittlement has been observed. This intermetallic compound formation has been observed to occur at temperatures above 300 C in liquid gallium and at temperatures above 500 C in gallium oxide. No structural deformation of cladding specimens has been observed to occur at gallium concentrations 10000 – 100000 times greater than that expected in WG-MOX fuel.

-Table 2. Gut-of-reactor gallium/clad compatibility tests for weapons-derived MOX fuel.

Variable	Phase I Static	Stressed	Phase II Static	Stressed
Gallium form	Metal	Metal	Ga oxide Ga/Ce oxide	Ga oxide Ga/Ce oxide
Concentration (%)	100	100	100, 1, 0.2, 0.1	100, 1, 0.2, 0.1
Cladding	Zr, Zircaloy-2, Zircaloy-4, Zirlo	Zr, Zircaloy-2, Zircaloy-4, Zirlo	Zr, Zircaloy-2, Zircaloy-4, Zirlo	Zr, Zircaloy-2, Zircaloy-4, Zirlo
Temperature (°C)	30, 300, 500[a,b]	30, 300, 500[a,b]	300, 500, 700	300, 500, 700
Time	200 h, 700 h	200 h, TTF	6 wk, 12 wk	6 wk
a - without cadmium, b – with cadmium, TTF – time to failure				

A complementary materials irradiation testing program has been initiated to extend the out-of-reactor tests to in-reactor conditions. These capsule tests employ two different WG-MOX test fuels fabricated at Los Alamos National Laboratory. Both fuels were fabricated with dry-processed 5% WG-Pu. One of the fuels was manufactured with plutonium that had received no additional purification (yielding a final gallium concentration of approximately 2 parts per million in the fuel), while the other fuel was manufactured from plutonium that had been thermally treated to remove gallium (resulting in a final gallium concentration of approximately 0.7 parts per million in the fuel). The fuel is being irradiated in the Advanced Test Reactor at Idaho National Engineering and Environmental Laboratory at liner heat generation rates of approximately 18-30 kW/m, and will be burned to approximately 30 GWd/MT burnup. The goal of this test is to develop a better understanding of gallium migration mechanisms within the fuel pellet and gallium/clad reaction mechanisms at the pellet/clad interface. The first irradiated fuel specimens (irradiated to about 8 GWd/MT) are currently undergoing post-irradiation examination at Oak Ridge National Laboratory.

6. COOPERATIVE R&D WITH RUSSIA

In keeping with the declarations made as a part of the "Joint Statement on Principles for Management and Disposition of Plutonium" in 1998, the U. S. and Russia are cooperating to accelerate the pace of the plutonium disposition activities. The focus of current cooperation is on the use of existing Russian reactors (WER-1000s and the BN-600) for plutonium disposition activities. The U. S. is working with Russia to develop and test VVER-1000, BN-600 (fast reactor) WG-MOX fuels, and to validate reactor physics, fuel cycle criticality, and safety codes as they apply to Russian reactors, Pu conversion, and fuel fabrication facilities. Based on analyses performed to date, it appears that the WER-1000 reactors would be capable of disposition approximately 425 Kg of plutonium per year. The BN-600 could burn approximately 300 Kg of Pu per year with a "hybrid core" option in which the reactor's current radial breeding blankets are removed, and up to 1.3 MT of plutonium per year if fully converted (by removal of the axial breeding blankets) for the plutonium consumption mission.

U. S. LWRs employ solid fuel pellets very similar in thermomechanical/physical design to European reactor-grade MOX fuel pellets. However, VVER-1000 reactors employ annular pelletized fuel with a central hole in the fuel pellet and a Zr-1%Nb alloy for cladding. Thus there is more uncertainty regarding the applicability of European reactor-grade MOX fuel performance data to the VVERs than there is with regard to the applicability of this data to the

U. S. LWRs. Plans are being made to conduct VVER WG-MOX irradiation tests in the MIR test reactor at the Research Institute of Atomic Reactors at Dimitrovgrad, and for irradiation of lead assemblies in a commercial VVER. The most likely candidate for irradiation of lead assemblies appears to be the Balakovo-4 plant. These tests should provide a good basis for judging the applicability of European commercial MOX fuel performance data to the VVERs.

Work is also underway to plan and implement conversion of Russia's BN-600 fast reactor to a plutonium-burning reactor. Russia has proposed to use their one BN-600 reactor at Beloyarsk to disposition 20 Mt of plutonium from the year 2005 to 2020. Reviews are currently being conducted of the safety and licensing requirements and economics.

As stated above, the U. S. is also collaborating with Russia to benchmark and validate the computational physics and criticality safety computer codes required for analysis and licensing of reactor and fuel cycle facilities. These codes include VENTURE, SCALE, HELIOS, MCNP, MCU, TVS-M, and TRIANG-PIN. A suite of over 100 benchmark calculations have been performed with aqueous WG-Pu solutions, Pu and MOX powders, fuel assemblies, and reactor lattices. The results of these analysis indicated that calculated k-effectives agree with experimental values to within 1%. Calculated power distributions within lattice experiments generally agree with measurements to within 2% although values as high as 6.5% were seen. These variations are larger than those found for LEU calculation-to-experiment comparisons. However, since MOX assemblies are expected to be placed in non-limiting positions in the reactor core host reactors should be able to operate at currently rated powers. Based on these results, it would appear that no additional critical experiments are required prior to insertion of the lead test assemblies into a VVER-1000. Furthermore, our Russian counterparts have judged that no additional critical experiments are necessary to support nuclear safety analyses for the out-of-reactor portion of the MOX fuel cycle.

In addition to these activities, the U. S. is cooperating with Russian to evaluate the feasibility of using reactors outside of Russia (CANDU reactors in Canada) or new/advanced reactors within Russia (gas turbine modular helium reactors) to disposition plutonium to supplement the disposition capacity provided by the current VVER-1000s and the BN-600. One CANDU test fuel bundle, comprised of fuel manufactured in the U. S. at Los Alamos National Laboratory and in Russian at the A. A. Bochvar All-Russian Research Institute of Inorganic Materials is scheduled to be loaded into the NRU reactor at Chalk River, Canada later this year. This test program will provide valuable information on the performance of WG-MOX fuel in CANDU reactor environments. Preliminary design work is just beginning for a plutonium consumption GT-MHR that could disposition 250 Kg of plutonium per year per module.

7. SUMMARY

The end of the Cold War has created a legacy of surplus fissile materials (plutonium and highly enriched uranium) in the U. S. and the former Soviet Union. During the past five years, the U. S. has devoted significant effort to the development of a practical, safe, and robust plutonium disposition option based on the use of existing commercial light water reactors and immobilization. The work conducted to date, along with international commercial MOX experience provides a strong technical basis for optimism that the use of weapons-grade MOX fuel can be successfully licensed and implemented in the U. S. The U. S. and Russia are cooperating to develop and demonstrate the technologies required for successful implementation of the reactor-based plutonium disposition option in Russia.

POSTER SESSION

MAIN TRENDS AND CONTENT OF WORKS ON FABRICATION OF FUEL RODS WITH MOX FUEL FOR THE WWER-1000 REACTOR

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Abstract

The main trends of production of pellet MOX-fuel for the VVER reactors using the trial-experimental equipment at SSC RF RIAR are set forth. The main realized parameters of fabrication of MOX-fuel pellets are presented. The content of the reactor tests program is considered with allowance for their licensing requirements for the VVER reactors.

1 INTRODUCTION

An important part of the stage of utilization of Russian weapon Pu excess is fabrication of 3 FA with MOX-fuel for the 4-th unit of the Balakov NPP, experimental substantiation of their serviceability during full-scale tests and obtaining the license for operation of MOX-fuel in the VVER- 1000 reactors.

Realization of such a decision supposes implementation of a complex of scientific-technological, test and research works to substantiate the creation of a new type of nuclear fuel as well as organizational and technical works to substantiate its commissioning. SSC RF RIAR possesses a definite technological base, modem research equipment and qualified personnel to fulfil such work. An advantage of RIAR choice to conduct this class of works is the fact that on one site there are technological sections, research reactors and a complex of hot cells for post-reactor material science investigations that allow license tests of fuel in the full volume and at the earliest possible date.

2.MAIN TRENDS OF WORKS ON FUEL TECHNOLOGY AT SSC RF RIAR

SSC RF RIAR has accumulated considerable experience on development and fabrication of various fuel rods, their pre-reactor and reactor tests, further material science investigations.

A trial-industrial complex is created at the Institute, at which fuel rods and FA with vibropac oxide uranium and MOX-fuel for fast reactors have been fabricated for more than 20 years. Up to now: in the BOR-60 reactor there are 500 spent FA with oxide uranium fuel and more than 500 FA with MOX-fuel; in the BN-600 - 6 FA **with** MOX and 4 FA with uranium fuel, in the BN-300 - 2 FA with MOX and 7 FA with uranium oxide fuel.

From other types of fuel rods and FA that were developed and fabricated in the Institute the following can be divided:

- fuel rods based on pellet oxide uranium fuel;
- fuel rods of the BOR-60 reactor with pellet MOX-fuel;
- fuel rods with dispersive fuel compositions;

- fuel rods of the BOR-60 reactor with metallic non-alloyed U and U-Pu fuel, with metallic alloyed U-Zr and U-Pu-Zr fuel;
- fuel rods of the BOR-60 reactor with cermet U-Pu fuel.

The technological sections for fuel production and fuel rods fabrication are maintained in the working state. The maintenance personnel-have sufficiently high qualification.

3. CONTENT OF WORKS ON PELLET MOX-FUEL AT SSC RF RIAR

The first stage of mastering the conversion process of Russian weapon Pu plans fabrication and testing of 3 trial FA (1000 fuel rods) with MOX-fuel in the 4-th unit of the Balakov NPP. Taking into account the short time of the test program, up to now the greatest preparedness of Russia is seen to produce fuel at the BTU glove box section of SSC RF RIAR.

The BTU production section has a license for the right of handling fissile materials, according to which 2 kg of Pu-239 or 5 kg of U with 90% enrichment are allowed at the working place.

The fabricated and controlled pellets are sent to the technological section of assembling, control and packing of fuel rods based on the production room with large-sized shielded boxes.

The following scheme of interaction of the project participants must be provided during fabrication of fuel rods with MOX-fuel for 3 FA:

- fabrication and delivery of initial U and Pu dioxides powders to SSC RF RIAR;
- fabrication and delivery of the relevant set of fuel rod claddings to SSC RF RIAR;
- fabrication of MOX-fuel pellets at the SSC RF RIAR technological section;
- fuel rods assembling, their sealing and certification;
- shipment of prepared fuel rods to the FA plant-manufacturer;
- assembling of 3 FA, their transportation to the Balakov NPP and testing in the 4-th unit.

In this case fabrication of the WER-1000 fuel rods is preceded by fabrication of a batch of trial fuel rods to conduct license tests at the MIR reactor.

The design of fuel rods with the pellet MOX-fuel for the WER-1000 reactor in terms of its characteristics must be identical to the regular fuel rod with pellet fuel based on U oxide with the average Pu content in FA - 3.4%.

The indispensable condition of fuel rods fabrication for 3 FA is the maximum compliance with their fabrication technology under plant conditions.

The basis of the requirements for the delivered U and Pu dioxides powders can be the characteristics of powders used in industrial fabrication of fuel rods for the WER reactors.

4. SCHEME AND POSSIBILITIES OF PELLETS AND FUEL RODS FABRICATION SECTIONS

The MIMAS technology spent under industrial conditions and that underwent comprehensive checking was taken as the basis for MOX-fuel pellets fabrication.

The BTU glove box section consists of 13 heavy glove boxes (BTU l-130 and 2 boxes (BTU-A and BTU-B) with enhanced protection served using tight manipulators. The boxes

Milling, granulation, sieving
(*) Mill, 2 kg/h ; Granulation facility, Grinding of pellets
(*) Grinding machine

BHU-À

BHU-Á

Storage of special products
Storage (U5% enrichment up to 35 kg; Pu up to 2 kg)

BHU-01

Preparation of powders, control of pellets
VLKT-500 scales
Mixing of powders, pressing of pellets
Mixer, press 10 t

BHU-02

(*) Pellet pressing facility, 20 kg/shift
pressing of briquettes and pellets, drying,
sintering
Calcination apparatus, 1000°C
Press 63 t; furnace 1700°C, loading of 2 kg

BHU-03

BHU-04

Drying of briquettes and pellets
Induction furnace 1500°C, VLKT-2000 scales

BHU-05

Pressing of pellets
Press 63 t,
Pellet pressing facility, 5 kg/shift

BHU-06

BHU-07

Drying and sintering of pellets
Furnace 1700°C, loading of 2 kg

BHU-08

Control of pellets (geometrical characteristics)
Pellet diameter measurement facility

BHU-09

Assembling of columns, fuel rods

BHU-10

Welding of fuel rods (up to 1100 mm)

BHU-11

Control of fuel rods tightness

Loading of sets of pellets to transport canisters

BHU-12

Decontamination and loading of canisters to transport
container. Control of Pu distribution.

BHU-13

FIG.1. *Placement of the equipment for pellets fabrication for 1000 fuel rods*

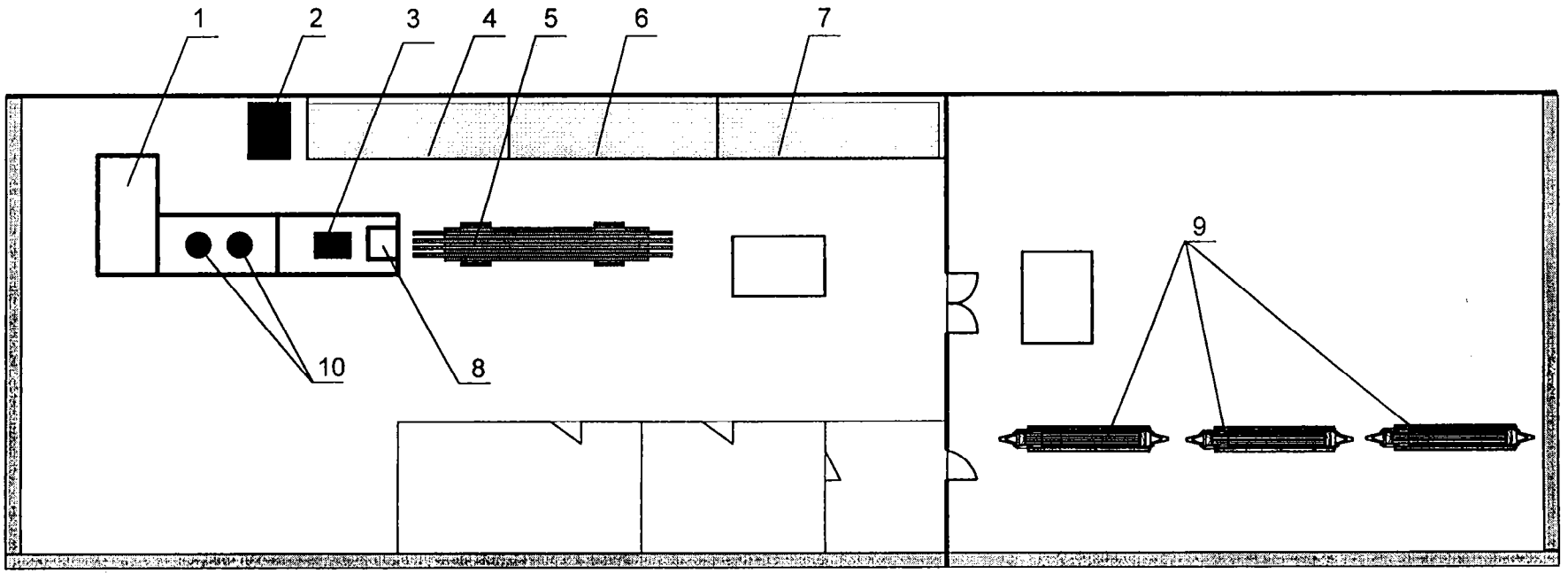


Fig.2. Scheme of layout

1 - box (1800õ700) for recieving, weighing and drying MOX-fuel; 2 - the stand of ultrasound check; 3 - the stand of butt-resistance welding (BRW); 4 - the stand of X-ray check; 5 - freight truck; 6 - the stand for checking integrity of fuel pin cladding; 7 - ACORT installation; 8 - technologic prechamber; 9 – TK-C2 container; 10 - furnace for pellet drying

are equipped with all engineer-technological systems necessary for normal work. All boxes are connected by the common transporter line. Each box is a box design of thin-sheet stainless steel. The box vessel is lined with steel sheet of biological protection with the front wall, up to 50 mm thick. Under each box there is a room for equipment and auxiliary systems (vacuum, electrical, testing instruments). The chain equipment can be quite easily re-adjusted to fulfil operations with fuel rods of various designs and sizes.
The purpose of the boxes and equipment placed in them is shown in Fig. 1. To fabricate the MOX-fuel pellets to assemble 1000 fuel rods of the VVER-1000 type, the section is supplied with the following:

- ball mill/mixture loading of 1.5-2 kg/;
- automatic press /up to 15-20 kg/shift/;
- mixer-homogenizer /loading of 2 kg powder/;
- sintering furnace /10 kg pellets per cycle/.

Meeting the requirements of high quality of the products assumes organization and control at all stages of the process, including:

- input control of the initial powder and documents;
- operational control of the technological process;
- control of the prepared pellets.

Besides, the progress of the technological process is under selective periodical control by the available techniques: porosity and structure of pellets, Pu distribution, oxygen/metal ratio, chemical composition, residual gases, etc.

All operations of assembling, fabrication and control of fuel rods with MOX-fuel are typical for the technological fabrication process of fuel rods with pellet uranium fuel and are conducted in the following order:

- input control of assembling parts and fuel;
- drying of pellets;
- making up of fuel columns;
- assembling of fuel rods and filling with helium;
- sealing with welding;
- decontamination;
- control operations;
  packing and transportation to the FA plant-manufacturer.

Trial fuel rods with MOX-fuel for irradiation in the MIR reactor are fabricated at the existing technological section.

Realization of the program of fabricating 1000 experimental fuel rods of the VVER reactor - 1000 fuel rods with pellet MOX-fuel requires the creation of a new section of fuel rods fabrication on the existing production areas using the large-sized shielded boxes and providing them with the pipelines of engineer systems.

The layout of the equipment at the fuel rods fabrication section is presented in Fig.2.

The obligatory types of equipment include the contact-butt welding and ultrasound control stand for fuel rods.

5. FUEL LICENSING

Licensing of a new fuel type assumes the availability of the experimental and calculated information on its behavior in all design states of the reactor facility.

5.1. **The initial position to form the program of works to substantiate licensing is, thus, determined by the following information:**

license requirements for fuel characteristics (including the requirements for FA and fuel rods) identified as fuel with serviceability. The main normalized parameters:

- pressure of helium and GFP under the cladding by the end of campaign;
- maximum fuel temperature;
- plastic deformation of the cladding due to its interaction with fuel;
- content of hydrogen in the cladding material;
- crack-formation in the fuel rod cladding (pressurized corrosion cracking);
- mechanical strength of the cladding in the axial direction;
- temperature of the fuel rod cladding at maximum design accident;
- oxidation depth of the 'fuel rod cladding (as part of the wall thickness);
- fuel enthalpy during the RIA type accident;
- power margin before fuel melting;
- ultimate number of untight fuel rods (for gas and fuel in the core);
- probability of fuel rod failure during operation.

main types of operating conditions of the WER-1000 reactor typical for normal operation (due to technical characteristics of the reactor) and emergency design situations determined by the Chief designer, namely:

- long operation at nominal power modes;
- bringing the reactor to power and its shutdown;
- change of the reactor power (FA while changing its placement in the core);
- after-loading modes (power ramp);
- emergency situation (event) related to fast unauthorized introduction of positive reactivity;
- break of the heat removal from the fuel rods surface (partial drying of the surface, heat removal crisis, type 1);
- operation of untight fuel rods.

license requirements for fuel must be met at all the design operation modes of the reactor.

5.2. **Test and investigation types:**

A set of tests and investigations is aimed at determining the values of fuel license parameters during realization of all design situations typical for the power reactor. The basic criterion of the fuel initial state is its burnup achieved at the regular irradiation parameters (except the tests of untight fuel rods). Therefore, the experimental information to substantiate the fuel licensing is obtained in 2 stages - implementation of reactor experiments modeling the loading modes typical for the reactor design states and post-reactor (nondestructive and destructive) investigations indicating the relevant fuel state (with determination of the license characteristics).

A set of tests includes:

- service-life tests;
  tests in transient modes (including: RAMP tests, manoeuvering experiments, tests of instrumented fuel rods);
  heat removal damage tests (including: tests simulating loss of coolant (LOCA tests), heat removal crisis tests);
- power pulse tests;
- tests of untight fuel rods (including: power pulse tests and RAMP type tests, heat removal damage tests);
  capsule experiments.

A set of post-reactor investigations includes:

- y-scanning;
- profilometry;
- cladding puncture with analysis of amount and composition of intra-fuel rod gas;
- fuel rods dismantling to samples, their preparation and photography;
- a-autoradiography of samples;
- mathematical processing of the results and building of models.

5.3. **Tests of FA mock-up in the MIR reactor:**

Start of the trial operation of new fuel in the power reactor can be provided prior to obtaining the full set of license information by parallel (with license experiments, which cannot be finished before 2006) testing of FA mock-up in the research reactor. In this case the mock-ups in the research reactor are irradiated ahead of schedule as compared to tests of first FA in the VVER- 1000 reactor. Such irradiation is an additional confirmation of fuel serviceability. The test schedule is built on the basis of parameters of standard 3-year utility, the advance of the burnup gained in fuel of the mock-up assembly (the MIR reactor) makes up 1.5 years. Test modes of the mock-up will be deliberately more rigid as compared to the trial operation modes of FA in the VVER- 1000 reactor, which is stipulated by more intense power manoeuvering typical for the research reactor. Rigidity of the irradiation modes of the mock-up in its turn will provide some reliability margin of trial FA.

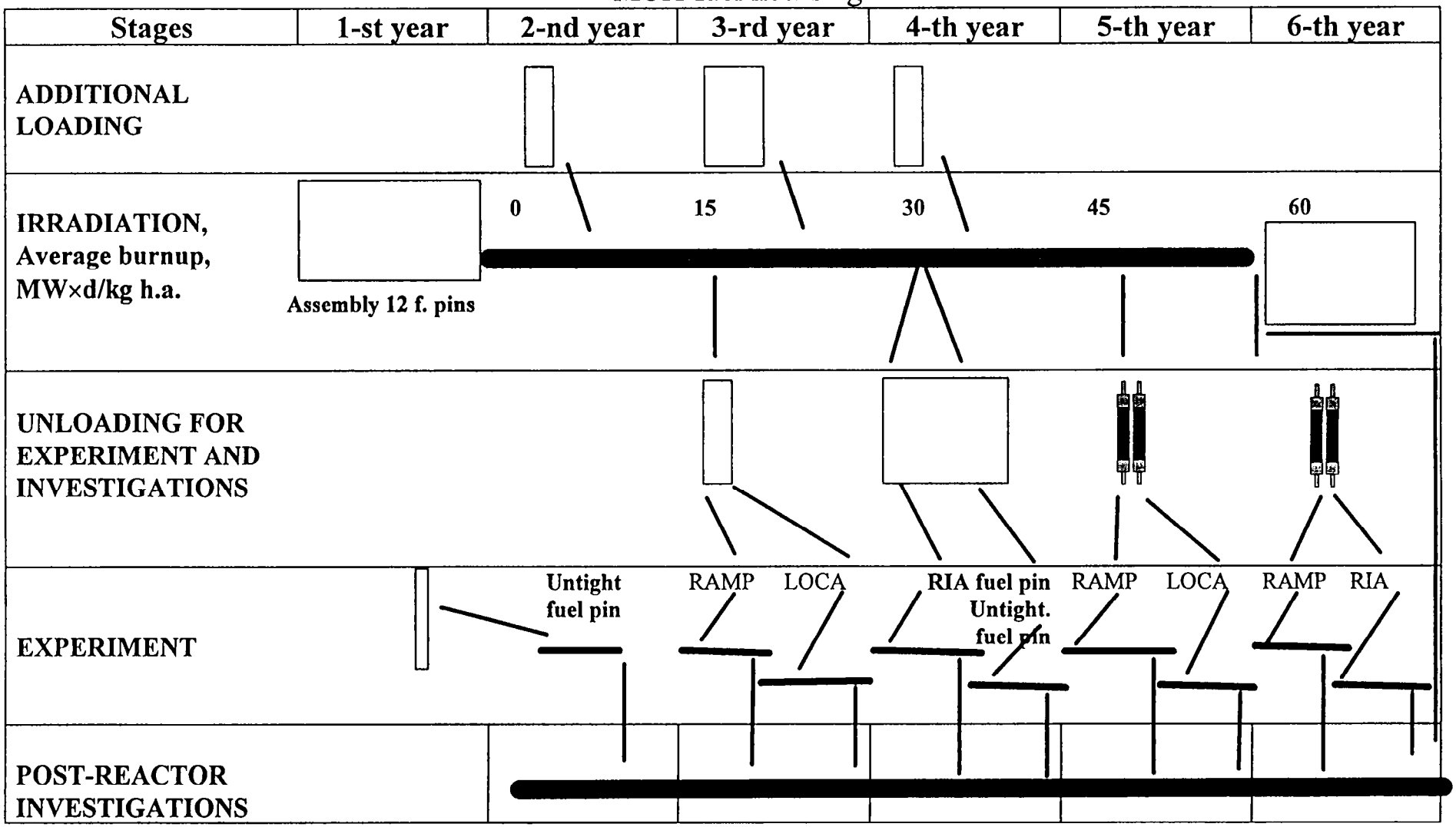
6. CONCLUSION

SSC RF RIAR possesses the necessary technological and test equipment to fulfil the following main works on substantiation of MOX-fuel usage in the VVER reactor:

- fabrication of pellet MOX-fuel for fuel rods in the MIR and VVER reactors;
- fabrication of the VVER-1000 type fuel rods (up to 1000) for tests in the WER reactor;
- fabrication and tests of experimental fuel rods in the MIR reactor within the frames of the licensing program of MOX-FA delivery for irradiation to the WER- 1000 reactor.

Test and investigation schedule to substantiate the vibropac

MOX-fuel licensing

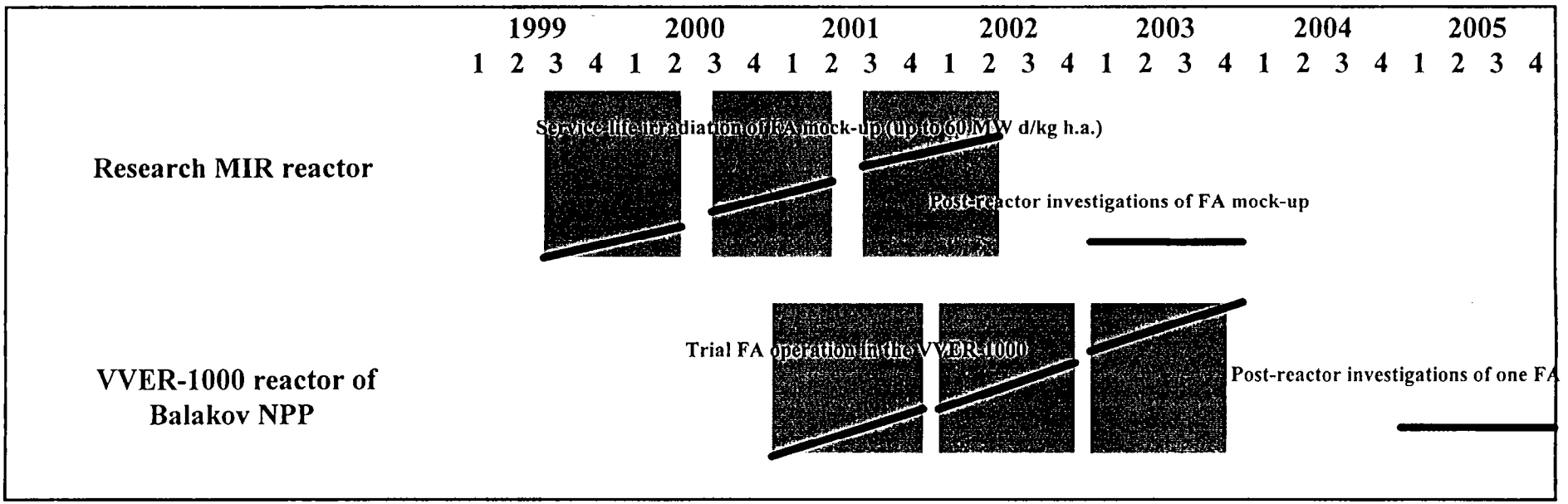


Reactor tests to substantiate MOX-fuel

1	Name	Purpose and content	Initial fuel state	Test results, investigated processes, mechanisms and parameters
1	**Service-life tests**	**Determining the state of fuel rods (fuel and cladding) in the range of design burnup of the VVER fuel, preliminary irradiation for further experiments (RAMP, LOCA, RIA).**	**Fresh fuel**	**Depend on burnup - medium pressure in the fuel rod cavity, deformation parameters, hydrogen saturation and oxidation depth of cladding, fuel structure and its characteristics**
2	**Tests in transient modes**	**Determining the state and kinetics of change of state parameters for fuel rods under conditions modeling regular transient modes corresponding to the reactor manoeuvering operations**	**Fresh fuel, irradiated fuel**	**Depend on parameters of power change and initial state of fuel rod - gas release, mechanical characteristics of fuel and cladding, critical linear thermal loading**
2.1.	RAMP tests	Determining the state (change of state) of fuel rods under power ramp conditions	Fresh fuel, irradiated fuel	Depend on the rate, power ramp, initial and final power level, burnup - gas release, relaxation thermo-physical characteristics, threshold thermal loadings
2.2.	Manoeuvering experiments	Investigation of state change of fuel rods under conditions modeling the reactor manoeuvering modes, including cyclic ones.	Irradiated fuel	Depend on frequency and parameters of power manoeuvers (regular scenarios), burnup - gas release and relaxation thermo-physical parameters
2.3.	Tests of instrumented fuel rods	Direct (during operation) and constant change of parameters in the fuel rod cavity during modeling of transient modes (step-by-step change of power)	Irradiated fuel	Depend on parameters of power and burnup change - gas pressure in fuel rod cavity, fuel temperature, change of fuel column geometry; revealing the change extremums of the registered parameters
3.	**Heat removal damage tests**	**Determining the characteristics of state change of fuel rods under reduced (as compared to nominal modes) heat removal intensity conditions**	**Fresh fuel, irradiated fuel**	**Depend on parameters of heat removal - changes of state**
3.1.	Tests simulating loss of coolant (LOCA)	Integral experiment on study of behavior of fuel rods included in the bundle (FA) under cladding surface drying conditions and fuel overheating	Fresh fuel, irradiated fuel	Depend on temperature, flow rate and modular state of coolant, burnup and linear power - time of preserving serviceability of fuel rod, character of cladding damage, change of fuel structure, reta of oxidation and corrosion processes (including steam-zirconium reaction), FP yield
3.2	Heat removal	Experiment to study the fuel properties after	Fresh fuel,	The same with more rigid conditions up to maximum design

1	Name	Purpose and content	Initial fuel state	Test results, investigated processes, mechanisms and parameters
.	crisis tests	realization of fuel rod overheating modes (heat removal crisis, type 1)	irradiated fuel	accident conditions
4.	**Power pulse tests**	**Determining the state of fuel rod after power pulse (a series of pulses)**	**Fresh fuel, burnt-out fuel**	**Depend on shape, duration, amplitude, quantity and periodicity of power pulses - fuel enthalpy, destruction threshold, character of cladding destruction and change of fuel structure, FP yield.**
5.	**Tests of untight fuel rods**	**Investigation of peculiar behavior of untight fuel rods in regular and emergency modes**	**Fresh fuel, burnt-out fuel**	**Depend on type, shape, placement and size of cladding defect, burnup, irradiation mode - fuel rod parameters and degree of serviceability**
5.1.	Long tests of fuel rods	Investigation of fuel rod serviceability and estimation of qualitative indices of its operation	Fresh fuel, burnt-out fuel	Depend on irradiation parameters and initial characteristics of defects, burnup - defect development kinetics, time of formation of secondary damages, FP and fuel yield to the coolant, permissible operation time, parameters of cladding degradation and changes of fuel structure.
5.2.	Power pulse tests and RAMP type tests	Investigation of state change of fuel rods in transient (power) modes and at impact thermal loadings	Fresh fuel, burnt-out fuel	Depend on initial defect state, burnup and parameters of power change - enthalpy destruction threshold and linear power, character of cladding destruction, change of fuel structure, FP and fuel yield to the coolant.
5.3.	Heat removal damage tests	Investigation of state change of defect fuel rod with its overheating	Fresh fuel, burnt-out fuel	Depend on temperature, flow rate and coolant modular state, burnup, linear power, initial characteristics of defect - time of preserving serviceability of fuel rod, character of cladding damage, change of fuel structure, rate of oxidation and corrosion processes (including steam-zirconium reaction), FP and fuel yield to the coolant.
6.	**Capsule experiments**	**Investigation of separate ageing and destruction mechanisms of fuel rods accompanying their operation to verify the calculated programs and codes.**	**Fresh fuel, burnt-out fuel**	**A set and parameters of experiments are determined by algorithm and data base of the verified program.**

Schedule of test works with FA mock-ups in the MIR reactor and trial operation of FA in the VVER-1000 reactor.



Note: average annual rate of burnup gain in the FA mock-up fuel in the MIR reactor is specified by the relevant characteristics of the VVER-1000 reactor.

A FISSION GAS RELEASE MODEL FOR MOX FUEL AND ITS VERIFICATION

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Abstract

A fission gas release model for MOX fuel has been developed based on a model for UO_2 fuel. Using the concept of equivalent cell, the model considers the uneven distribution of Pu within the fuel matrix and a number of Pu-rich particles that could lead to a non-uniform fission rate and fission gas distribution across the fuel pellet. The model has been incorporated into a code, COSMOS, and some parametric studies were made to analyze the effect of the size and Pu content of Pu-rich agglomerates. The model was then applied to the experimental data obtained from the FIGARO program, which consisted of the base irradiation of MOX fuels in the BEZNAU-1 PWR and the subsequent irradiation of four refabricated fuel segments in the Halden reactor. The calculated gas releases show good agreement with the measured ones. In addition, the present analysis indicates that the microstructure of the MOX fuel used in the FIGARO program is such that it has produced little difference in terms of gas release compared with UO_2 fuel.

1. INTRODUCTION

It is generally believed that fission gas release can be enhanced in MOX fuel compared with conventional UO_2 fuel under similar operating conditions. This enhancement in MOX fuel may be attributed to slightly lower thermal conductivity, higher reactivity later in life and/or some amount of Pu concentrated in a certain number of agglomerates. Since the first two factors can be considered in terms of fuel temperature, an attempt has been made to evaluate the effect of the third factor on fission gas release in MOX fuel.

The model developed in the present paper uses the concept of equivalent spherical cell based on a gas release model for UO_2 fuel. The model considers the uneven distribution of Pu within the fuel matrix and a number of Pu-rich agglomerates that could lead to a non-uniform fission rate and fission gas distribution across the fuel pellet.

The model has been incorporated into a code, COSMOS [1], and some parametric studies were made to analyze the effect of the size and Pu content of the agglomerates. Then the model was applied to both MOX and UO_2 fuel to show the different behavior of gas release depending on the microstructure of the MOX fuel. Finally, the calculation results of the model were compared with the experimental data obtained from the FIGARO program [2] and were analyzed in terms of the microstructure of MOX fuel.

2. DESCRIPTION OF THE MODEL

A basic unit used for the development a fission gas release model for MOX fuel is an equivalent spherical cell shown in Fig. 1. An equivalent spherical cell consists of an equivalent spherical particle with the diameter of $D_{eq} = D_{agg} + 2 \cdot L_{rec}$ and the matrix surrounding it. Here, L_{rec} is the recoil length of the fission products of about 6 μm. Although about half of the gas atoms

generated in the band within L_{rec} from the periphery of the agglomerate are deposited in the region between D_{agg} and D_{eq}, the production rate of fission gas in the equivalent spherical particle is assumed to be uniform.

The diameter of an equivalent cell, D_{cell}, which is determined in such a way that the total Pu mass in each cell is equal to the sum of the Pu mass in an agglomerate and that in the matrix around it, is as follows:

$$D_{cell} = \frac{D_{agg}}{\sqrt[3]{\frac{e_p - e_m}{e_a - e_m}}} \tag{1}$$

where

D_{agg} is the diameter of the Pu-rich agglomerate (μm),

e_p is the average Pu content in the pellet (Pu/HM, w/o),

e_a is the Pu content in the agglomerate (Pu/HM, w/o),

e_m is the Pu content in the matrix between D_{eq} and D_{cell} (Pu/HM, w/o).

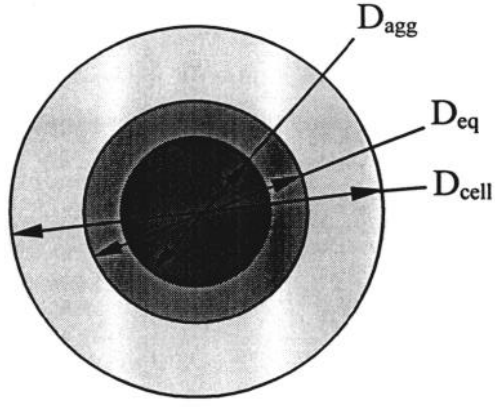

FIG. 1. An equivalent spherical cell for heterogeneous MOX fuel

If the manufacturing parameters of MOX fuel are such that D_{eq} is equal to or greater than D_{cell} calculated by Eq. (1), the MOX fuel can be treated as homogeneous, because in this case the distribution of generated fission gas atoms in the equivalent cell would be uniform.

Using the average local fission rate F_{av} that is derived from the local heat generation rate, the fission rate for the matrix between D_{eq} and D_{cell} is calculated as follows:

$$F_m = \left(\frac{e_m}{e_p}\right) \cdot F_{av} \tag{2}$$

The fission rate in the equivalent particle is then obtained in such a way that the total fission rate in an equivalent cell is equal to the sum of the fission rate in the equivalent particle and that in the matrix between D_{eq} and D_{cell}:

$$F_{eq} = \left[\frac{e_m}{e_p} + \left(1 - \frac{e_m}{e_p} \right) \frac{D_{cell}^{\ 3}}{D_{eq}^{\ 3}} \right] \cdot F_{av} \tag{3}$$

where D_{eq} is the diameter of the equivalent spherical particle defined by $D_{agg} + 2 \cdot L_{rec}$ as above.

Then, a gas release model for UO_2 fuel [3] is applied separately to the equivalent spherical particle and the matrix zone, and then the fission gas released from these two zones are added up to get the total release from one equivalent spherical cell. In this model, once the number of gas atoms on the grain boundary reaches $8 \cdot 10^{19}$ atoms/m2 [4], the grain boundary is assumed to be saturated with gas bubbles and additional gas atoms arriving here by diffusion are considered released to the fuel outside. In addition, the microstructure of the Pu-rich agglomerate is assumed to be the same as that of the matrix except for the fissile content. This means that the gas release mechanisms in the grains located in the agglomerate and in the matrix are the same. Fig. 2 shows the schematic calculation procedure for gas release in MOX fuel.

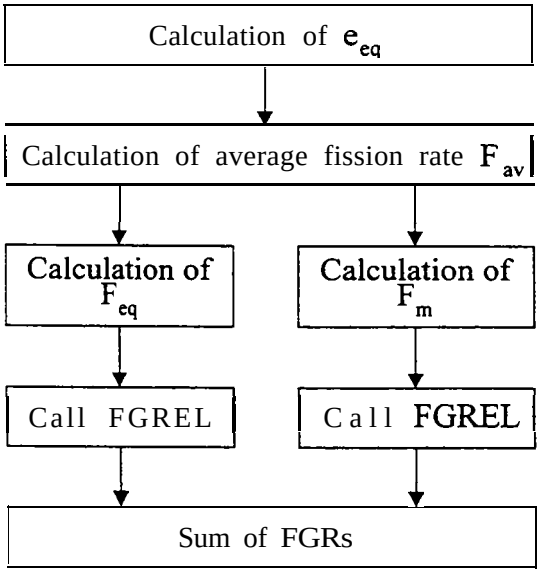

FIG.2. Calculation procedure for gas release in MOX fuel

3. PARAMETRIC STUDY

Parametric study was made to investigate how manufacturing parameters of MOX fuel such as the size of Pu-rich agglomerates and the average Pu content in the matrix affect gas release for a constant fuel temperature of 1000°C and a linear power of 250 W/cm. Fig. 3(a) shows the calculation result for the ideal case that the agglomerates with the same size containing a Pu content of 23% are uniformly distributed in the matrix with an average Pu content of 3%. In the case that the agglomerate size is 10 μm, which is the same as the typical grain size of the matrix, D_{eq} and D_{cell} are calculated to be 22 μm and 19 pm, respectively. Since D_{eq} is larger than D_{cell}, MOX fuel can be considered homogeneous and therefore, the gas release behavior is the same as for U02 fuel.

The larger the size of the agglomerates, the more gas is released due to more generation of gas atoms in the agglomerates and the earlier formation of release paths in the grain boundaries of the agglomerates. According to Eqs. (1) and (2), for the extreme case of an agglomerate size of 50 pm, the volume fraction of the matrix region between D_{eq} and D_{cell} is 71%, where 36% of the total fissions occurs. Therefore, the heterogeneity of the cases that are being considered here is not so remarkable as the manufacturing parameters of MOX fuel might suggest. This is why the difference in gas releases between the two extreme cases, that is, between a homogeneous microstructure and a heterogeneous one, is not so large compared to their absolute magnitudes. This also implies that, although each parameter of the size, Pu content and number density of the agglomerates influences gas release in MOX fuel, it is not just one parameter but the combination of the above three parameters that ultimately determines the overall gas release behavior in MOX fuel compared with U02 fuel.

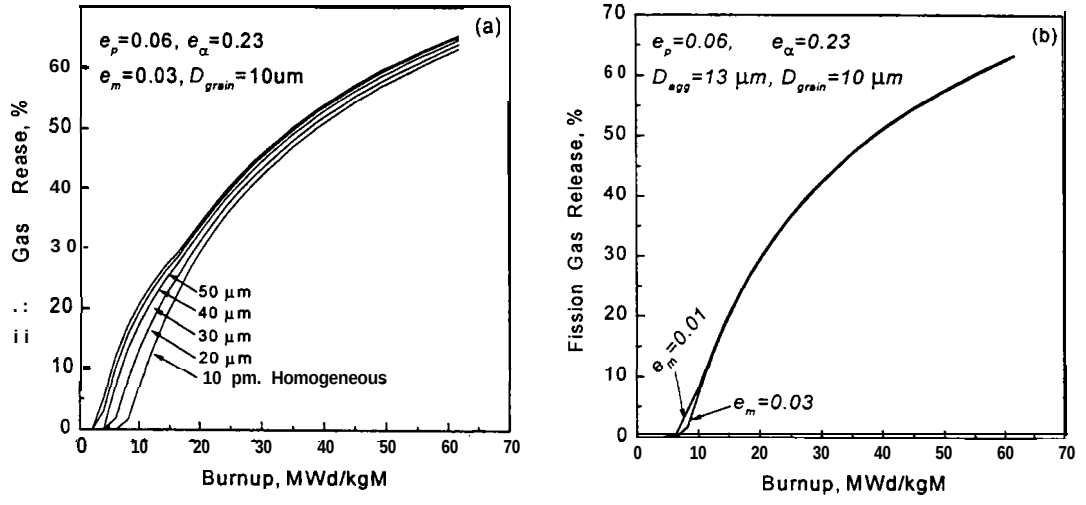

FIG. 3. Effect of (a) the size of Pu-rich agglomerate and (b) the average Pu content in the matrix **on** *fission gas release in MOX fuel.*

Fig. 3(b) shows that the MOX fuel with lower Pu content in the matrix gives a little higher gas release at low burnup. The reason is that, for the same amount of total Pu, more Pu is available in the agglomerates of the MOX fuel with lower Pu content in the matrix, and this yields more fission and therefore more release. However, since D_{eq} is larger than D_{cell} for both cases, MOX fuel can be considered homogeneous, explaining the lack of difference between them except for low burnup.

4. APPLICATION OF THE MODEL TO MOX AND UO_2 FUEL

The present model has been applied to compare the gas release behavior of MOX and U02 fuel that would be irradiated in a typical PWR with a constant average linear power of 220 W/cm during their lives. In addition, two fuel rods are assumed to have the same chopped cosine type of axial power shape. Fig. 4(a) shows gas release in a U02 fuel with an enrichment of 5% and a MOX fuel with a fissile Pu content of 5% both in the agglomerate and in the matrix. Considering a typical grain size of 10 pm in the U02 fuel, both the agglomerate size, D_{agg}, and the grain size in the matrix, D_{grain}, are also taken to be 10 pm, respectively, in the MOX fuel. In other words, the MOX fuel has a very homogeneous microstructure that is the same as that of U02 fuel. However, the two fuels would have slightly different thermal conductivity and radial power profile across the fuel pellet due to the different thermal neutron absorption cross section of U-235 and fissile Pu isotopes. Fig. 4(a) shows that these two factors, which would produce a slightly different temperature distribution within the

pellet, are combined in such a way that the gas release from the U02 fuel is a little higher than that from the MOX fuel. Gas release is accelerated for bumups greater than about 30 MWdkgM because the grain boundary is saturated with gas bubbles, and additional gas atoms arriving at the grain boundary are released to the fuel outside for this burnup range.

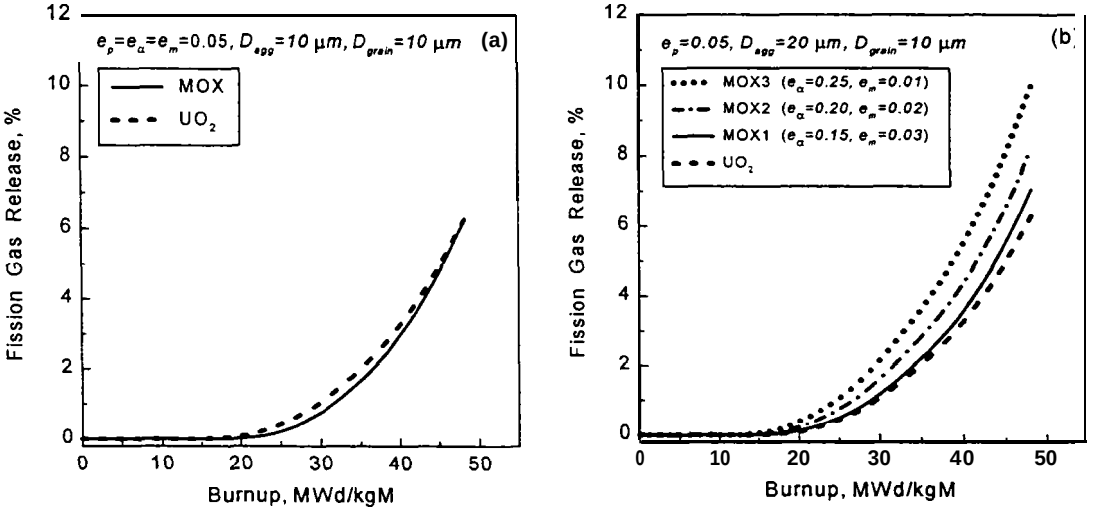

FIG. 4. Fission gas release in (a) homogeneous and (b) heterogeneous MOX fuel and its comparison with U02 fuel

Fig. 4(b) compares the gas releases from a UO_2 fuel and a MOX fuel that has the same Pu content but with three different microstructures, that is, three different distribution of Pu between the agglomerates and the matrix. Here, D_{agg} and D_{grain} are taken to be 20 pm and 10 μm, respectively, in the MOX fuel. The Pu content in the matrix e_m is calculated using a number density of $4 \cdot 10^{13}$ /m^3 agglomerates in the MOX fuel [6]. In these cases, according to calculations made by Eqs. (1) and (3) for the manufacturing parameters given here, the fission densities in the equivalent particles are about 20 % to 40% higher than those in the U02 fuel. This would lead to the earlier saturation of the grain boundary with gas bubbles, the earlier formation of the release path and finally, more fission gas release in the equivalent particles.

The situation, however, is the opposite in the MOX fuel's matrix region, whose fission density only amounts to 20 to 60% of the U02 fuel's, depending on the cases analyzed, and whose volume is about 30% of the U02 fuel's. Consequently, this situation in the MOX fuel's matrix region would yield slower gas release than in the UO_2 fuel. Since these two opposite situations compensate each other in terms of gas release, the dominant effect of the two, which is influenced by the size, Pu content, and number density of the agglomerates, would ultimately determine the overall gas release in MOX fuel. Fig. 4(b) indicates that the former effect plays a dominant role over the latter in the present three combinations of manufacturing parameters.

5. COMPARISON OF THE MODEL WITH EXPERIMENTAL DATA

To verify the present gas release model for MOX fuel, it has been incorporated into a computer code, COSMOS [l], which was developed for the analysis of MOX and U02 fuel. One of COSMOS's features is that it can analyze fuel segments refabricated from base-irradiated fuel rods. This is possible by saving all the relevant information at the end of base irradiation for the position that would be used for refabrication. They are then used as the initial conditions for the analysis of refabricated segments that are usually irradiated in an experimental reactor. The relevant information

for MOX fuel that needs to be stored for the analysis of the refabricated MOX fuel is as follows: first, the respective amounts of fission gas stored in the interior and the boundary of the grains located both in Pu-rich agglomerate and in the matrix. Second, each grain size in the two regions is required. Other information, such as fuel geometry, power history and coolant condition, is given in the input data used for the analysis of refabricated fuel segments.

The COSMOS has used the FIGARO results [2] to verify the present model. In the FIGARO program, two MOX fuel rods that had been irradiated in the Swiss BEZNAU-1 PWR of NOK during five cycles at moderate power were chosen for refabrication and subsequent irradiation in the Halden reactor. The main difference between the two fuel rods was the grain size in the U02 matrix; 15 and 8 pm, respectively. Four segments, where two segments had been cut from each fuel rod, were refabricated and then reirradiated in the Halden reactor in two phases. Each segment was equipped with a central thermocouple at the top of the fuel column and a pressure transducer at the bottom.

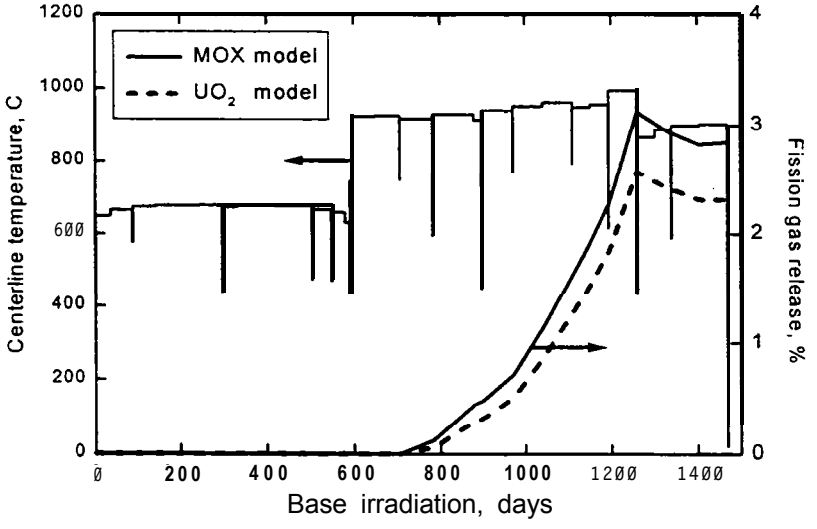

FIG. 5. Comparison of fission gas release for base-irradiated MOX fuel.

Fig. 5 compares the calculated gas release for the MOX fuel base-irradiated with the power history of Fig. 1 of Ref. 2. While the dashed line represents the calculation for U02 fuel enriched to 6%, the solid line shows the result for the MOX fuel used in the FIGARO program. To produce the MOX pellets for the FIGARO program, a master blend enriched to about 24% Pu/(U+Pu) was diluted with depleted UO_2 up to an average Pu content of 6%. This suggests that the maximum Pu contents were 24% in some Pu-rich agglomerates. Gas release for MOX fuel in Fig. 5, therefore, was calculated using the assumption that all the agglomerates in this fuel have the same size with the same Pu content of 24%. However, according to a characterization report on the MOX fuel rods used in the FIGARO program [5], the size of agglomerates has a log-normal type distribution and most of them are similar to or smaller than the grain size in the matrix. In addition, the Pu contents in the agglomerates have a distribution ranging from 1% to 20%. This suggests that measured release would be located between the two calculated releases for MOX and UO_2 fuel using the present model. This argument was confirmed when the measured release was found to be located between the two calculated ones. This also indicates that the size and Pu content in the agglomerates in the MOX fuel for this experiment hardly enhanced gas release due to the non-uniform distribution of Pu in the fuel matrix and a number of Pu-rich agglomerates.

The fission gas release for one of the fuel segments irradiated in the Halden reactor during the second phase of the experiment with the power history of Fig. 6(a) is given in Fig. 6(b). The predicted release was about 2% higher than the measured one. Although the COSMOS predicted a burst release of about 4% during the period of stepwise power increase, which was not found in the experiment, the

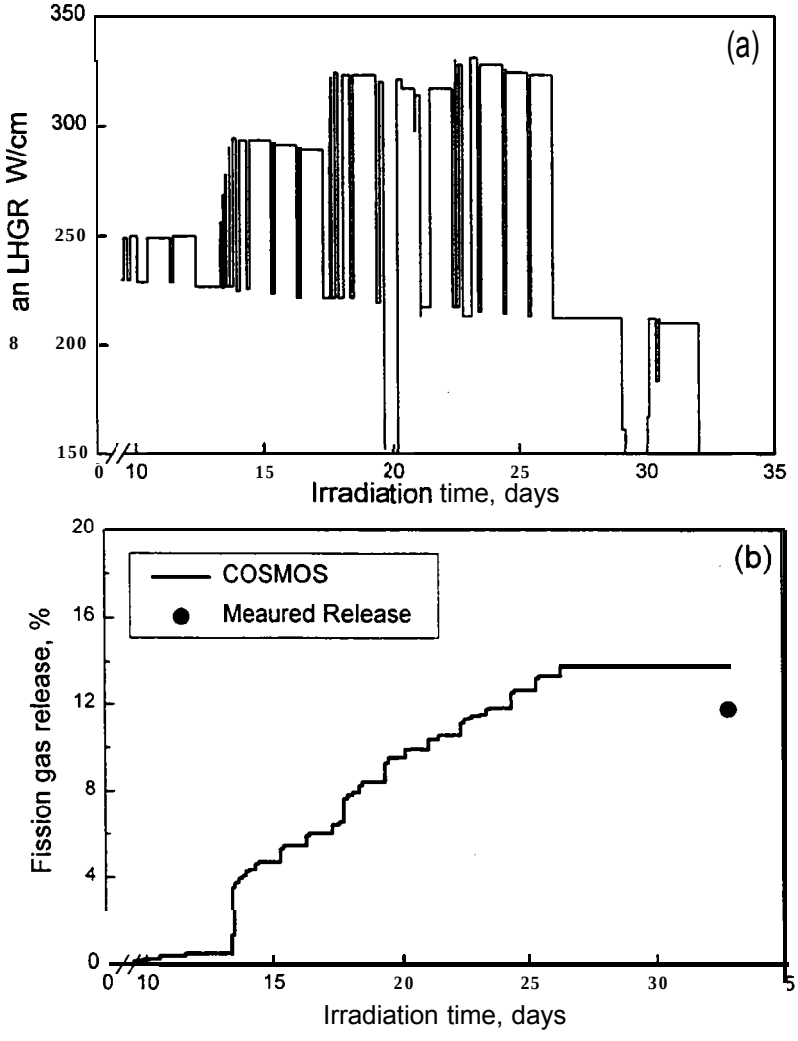

FIG. 6. (a) Power history and (b) fission gas release for phase 2 of the Halden irradiation

trend of gas release by diffusion during the high power period was reasonably simulated. This difference of 2% between the calculated and measured release could arise from the predicted burst release. Another possibility for this difference is that the same assumption as for the analysis of base irradiation, that is, all the agglomerates have the same size with the same Pu content of 24%, was used. This assumption would have increased the predicted gas release, as in the case for the base irradiation. Considering these circumstances, it can be concluded that the present model satisfactorily predicted the gas release behavior of the MOX fuel used in the FIGARO program.

6. CONCLUSION

To evaluate the in-pile behavior of MOX fuel, a fission gas release model for MOX filel was developed using the concept of equivalent spherical cell based on a gas release model for UO_2 fuel. This model considers the uneven distribution of Pu within the fuel matrix and a number of Pu-rich particles that could lead to a non-uniform fission rate and fission gas distribution across the fuel pellet.

Parametric study using the present model shows that gas release in MOX fuel is influenced by the size, Pu content and number density of the agglomerates. However, it reveals that it is not only one parameter but the combination of the above three parameters that determines the release behavior of MOX fuel compared with UO_2 fuel with the same fissile content.

Comparison with the FIGARO results shows that the present model satisfactorily predicts the measured ones. In addition, it is found that the microstructure of the MOX fuel used in the FIGARO program is such that it has produced little difference in terms of gas release compared with UO_2 fuel.

ACKNOWLEDGEMENT

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MOX FUEL TESTS IN THE SHIFT FACILITY IN THE HFR

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Abstract

The study of the properties of fuels at high burnup is an important subject in nuclear research. Knowledge of the changes in the fuel properties at high burnup is essential for safe and economical operation of the fuel. To that purpose a special sample holder for in-pile irradiation testing has been designed for the HFR. This so-called SHIFT (Sample Holder for Instrumented Fuel Tests) sample holder, of which the first one is presently being constructed, is described in this paper, together with some relevant aspects of the nuclear infrastructure at Petten. The first SHIFT facility will be used for the irradiation of rodlets containing the following materials: U_{1-x} Pu,O2, UO_2, $TH_{1-x}Pu_xO_2$, $Th_{1-x}U_xO_2$ and ThO_2.

1. THE SHIFT FUEL TEST FACILITY

The SHIFT has been designed as a versatile fuel testing facility for the HFR. The SHIFT allows performing fuel tests and parametric studies on a set of small fuel rodlets simultaneously. The rodlets can either be contained in an aluminium drum or in a liquid sodium environment depending on the required fuel and cladding temperature. The aluminium drum or the sodium bath are surrounded by a stainless steel containment (first containment). The SHIFT sampleholder is inserted in one of the three legs in a TRIO rig position of the HFR. The stainless steel TRIO rig forms the second containment. Helium gas or neon gas filled gas gaps are present between the aluminium drum and the first containment and also between the first containment and the second containment. Changing the gas composition in these gaps allows for temperature control of the experiment. The legs of the TRIO rig are surrounded by cooling water with a temperature of 40°C. An example of a horizontal cross section of the SHIFT fuel test facility is shown in figure 1. The fuel rodlets can be equipped with pressure transducers and thermocouples. The thermocouples can be situated both inside the fuel pellets and inside the fuel cladding. The combination of thermocouples inside the pellets and inside the cladding allows to study the impact or irradiation on the thermal conductivity of the fuel. Detailed pre- and post irradiation neutronic and thermal computations combined with a data set of the temperature field as measured during irradiation and a data set of the neutron metrology give an accurate prediction and evaluation of the fuel irradiation conditions. The thermal computations are performed with the Finite Element Method (FEM) code ANSYS 5.4. Using this code a detailed thermal description of the complete SHIFT facility is made.

Neutronics computations are performed with the HFR-TEDDI code. This is a two dimensional (2D) code especially made to perform neutronics computations for the core geometry of the HFR. The code takes all materials into account used in the irradiation experiment. Besides the 2D HPR-TEDDI computations detailed 3D Monte Carlo neutronics computations can be performed in order to gain a more detailed insight in the neutron-flux distribution in the complete SHIFT assembly.

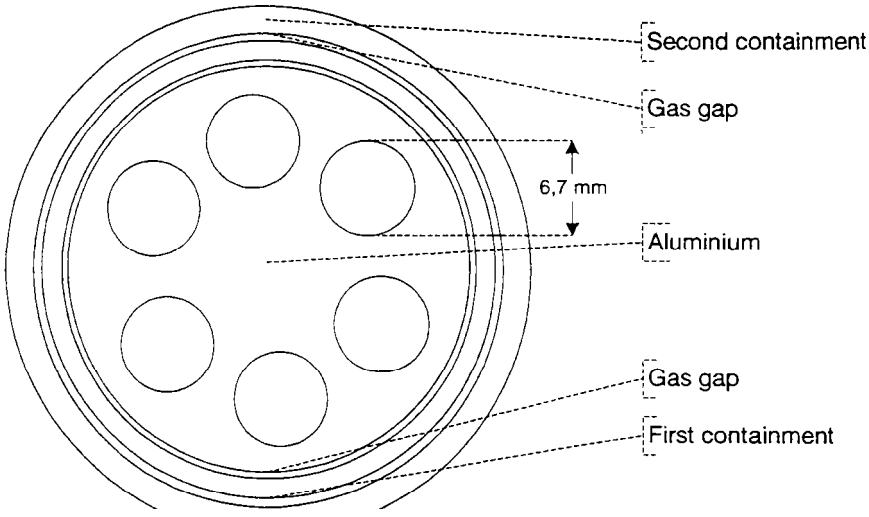

Fig. 1. Cross section of a SHIFT fuel test facility containing six holes in the aluminium ***drum in*** *which the five fuel rodlets are to be inserted.One of these holes will be used for another type of irradiation experiment.*

The SHIFT facility is very suitable for various types of MOX research such as studies on the behaviour of MOX up to high burnup (e.g. fission-gas release, changes of the thermal conductivity), or studies on the influence of modifications of the MOX production technique, (e.g. changes of the homogeneity of the Pu-distribution, Pu-content, Am-concentration or Pu-vector). Since the SHIFT facility can contain sets of fuel rodlets under approximately identical flux and temperature conditions, the facility is very suitable for comparative tests between different types of fuels.

The wide range of nuclear characteristics of the in-core positions and the large number of annual full power days of the HFR, together with the use of small diameter fuel test rodlets makes it possible to achieve a high burnp of the fuel in a short time in the SHIFT, avoiding a non-typical fuel temperature. By selecting a suitable in-core position and by tailoring the SHIFT facility, the rodlets can be irradiated under similar temperature and neutron flux spectrum as those in a LWR. Since SHIFT uses only l/3 of an in-core position and up to six rodlets can be accomodated in one SHIFT facility, the tests are very cost effective.

In the SHIFT-l experiment, which is planned to start at the end of 1999, $U_{1-x}Pu_xO_2$, UO_2, $Th_{1-x}Pu_xO_2$, $Th_{1-x}U_xO_2$ and ThO_2 samples will be irradiated to a burnup of about 60 MWd/kgHM in 2 years. ThO_2 and $Th_{1-x}U_xO_2$ are known to have good thermal and physical properties and good radiation stability [l]. These properties, and the fact that very little plutonium and americium is formed during the irradiation of thorium, makes ThO_2 a good candidate matrix for the transmutation of plutonium and americium. The combined irradiation of the uranium based fuels $U_{1-x}Pu_xO_2$ and UO_2 and the thorium-based fuels $Th_{1-x}Pu_xO_2$, $Th_{1-x}U_xO_2$ and ThO_2 under identical conditions allows for a direct comparison of the properties of these fuels in the complete burnup range of interest to the application of thorium based fuel.

2. THE HIGH FLUX REACTOR PET-TEN

The HFR is one of the most powerful multi-purpose research and test-reactors in the *world*. *The HFR* has a recognised record of consistent and reliable operation with approximately 275 full power days per year. The HFR has 19 in-core and 12 poolside irradiation positions, plus 12 beam tubes. The SHIFT facility occupies l/3 of one of the in-core positions.

Besides the SHIFT facility, at the HFR a wide variety of other fuel test facilities are available [2]. These facilities are mostly reloadable irradiation capsules of modular design, allowing easy adaptation to various instrumentation options (e.g. measurement of fuel rod length, fuel stack

displacement, fuel rod pressure, central temperature, diameter profiles and testing under various load conditions). They provide typical LWR system conditions at the fuel rod surface and continuous monitoring for fuel rod failure by activity monitoring of the fuel rod surrounding water.

3. THE NRG HOT LABORATORY

The NRG hot cells have the full range of fuel and cladding testing facilities [3] available, such as: visual inspection, gamma spectrometry, micro gamma scanning, eddy current measurement, profilometry, puncturing and fission-gas analysis, X-ray analysis, metallography, cermography, density measurement, EPMA, fuel re-fabrication and re-instrumentation facilities.

4. THE NRG ACTINIDE LABORATORY

In order to be able to handle unirradiated alpha-emitting actinides and beta-emitting fission products a new actinide laboratory has been taken into operation. Extensive fuel and target production and characterisation facilities are available in this laboratory. Since both hot laboratories are located at the Petten site, pre- and post-irradiation transportation problems are avoided.

5. CONCLUSIONS

The SHIFT facility, the HFR, the hot laboratory and the actinide laboratory, together with 2/3D computer codes for thermal-hydraulic and nuclear modelling are offering a powerful combination to study the properties of MOX fuel. The SHIFT facility can perform both scoping tests on innovative MOX types to study their overall behaviour and perform dedicated measurements on fuel properties such as fission-gas release and thermal conductivity. In the SHIFT-l experiment the behaviour of $U_{1-x}Pu_xO_2$, UO_2, $Th_{1-x}Pu_xO_2$, $Th_{1-x}U_xO_2$ and ThO_2 samples will be studied.

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THE VIPEX PROGRAMME FOR NEUTRON CODE VALIDATION FOR MOX FUEL

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Abstract

A short description is given of the VENUS critical facility and the experimental techniques used in this facility. These techniques comprise the measurement of the critical water level, the reactivity effect of the water level, the determination of axial and horizontal pin-by-pin fission rate distributions, the measurement of several spectrum indices and both angular and radial fission rate distributions inside a fuel rod and the determination of the delayed neutron fraction β_{eff}. With help of the measurement of the critical water level and its reactivity effect other reactivity effects can be determined. Examples are control rod worth, americium effect in MOX fuel and the influence of the water density.

The VIPEX programme aims at vaiidating neutron codes for different aspects of the control of reactors with MOX fuel. Parameters that have been investigated in the framework of this programme are the ^{241}Am effect on the reactivity, control rod worths in MOX, the delayed neutron fraction β_{eff}, the detector response in MOX, the reactivity effect of the water moderator density and the flux tilt at the border of a MOX and UO_2 assembly.
Comparisons are given between measurement results and calculations of several parameters determined in the framework of the VIPEX programme.
Some future programmes will be discussed.

1. INTRODUCTION

During already more than 25 year, the VENUS critical facility is used for benchmarking calculation codes for in-core and out-of-core neutron physics. The introduction of MOX in nuclear power plants has created a need for neutron physics benchmarks that deal with fuel configurations (partially filled) with MOX. Due to the high number and the diversity of the available MOX rods, the VENUS critical facility is particularly suited for establishing these kind of MOX benchmarks.

The last 10 years several international programmes have been executed in the framework of MOX benchmarking [1,2,3]. These programmes were VIP-BWR, VIP-PWR, VIP0 and VIPEX-PWR The most recent programme VIPEX-PWR aimed at completing the VIP database with experimental data more related to mixed fuel core operation and safety. Parameters that have been investigated in the framework of this programme are the ^{241}Am poisoning (^{241}Pu decay), control rod cluster (AgInCd) worths in MOX, the delayed neutron fraction β_{eff}, the detector response in MOX, the reactivity effect of the water moderator density (simulating hot to cold condition) and the flux tilt inside a MOX rod at the border of a MOX and UO_2 assembly.

2. DESCRIPTION OF THE VENUS FACILITY

The VENUS critical facility is a water-moderated zero-power reactor. It consists of an open (non- pressurized) stainless-steel cylindrical vessel including a set of grids which maintain fuel rods in a vertical position. After a fuel configuration has been loaded, criticality is reached by raising the water level in the vessel. (see figure 1).

Parameters that are measured with the VENUS reactor are the critical water level h_c, the reactivity coefficient $\delta\rho/\delta h$, the axial fission rate distribution, the horizontal fission rate distribution, spectrum indices F5/F9, F8/F9, C8/F9, fission rate distribution inside fuel rod, detector response and the delayed neutron fraction β_{eff}.

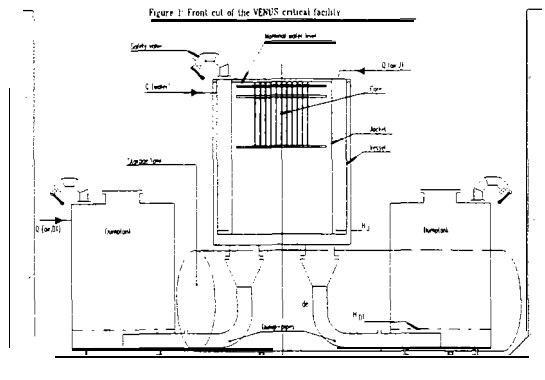

Figure 1. Vertical cross-section of the VENUS critical facility

3. PARAMETERS THAT CAN BE MEASURED WITH THE VENUS REACTOR

The following parameters are measured with the VENUS reactor.

the critical water level h_c
the reactivity coefficient $\delta\rho/\delta h$
the axial fission rate distribution
the horizontal fission rate distribution
spectrum indices F5/F9, F8/F9, C8/F9
fission rate distribution inside fuel rod
detector response
the delayed neutron fraction β_{eff}

3.1. **The critical water level**

The critical water level is measured in order to determine the critical mass of the loaded configuration. Due to adjustments of the outer feeding zone, the critical level can be arranged to a limited extent. Recent configurations had a critical height of about 40 to 45 cm. The critical level is measured with a random uncertainty of 0.02 cm and a systematic uncertainty of about 0.07 cm.
The comparison of the same or similar configurations will only be affected by the random uncertainty. This is in particular important when reactivity effects of small changes of the configuration are measured.

With help of the critical level measurements also changes in reactivity can be measured. These changes can be due to e.g. Am effect, changes in moderator density, replacements of absorber rods (Gd, B_4C, AgInCd), etc.

3.2. **Reactivity effect of the water level**

The reactivity coefficient $\delta\rho/\delta h$ is measured to make a link between the uncertainty of the critical level measurement and the uncertainty of the reactivity. Normally a value for $\delta\rho/\delta h$ is measured of about 0.30 to 0.35 %/cm. Given the random uncertainty of the critical water level of 0.02

cm, the uncertainty of the reactivity is about 0.007% or 7 pcm. In case the systematic uncertainty plays a role, too, the uncertainty of the reactivity is about 24.5 pcm.

The reactivity coefficient is determined by the measurement of two successive periods, created by making the reactor supercritical with an increment of the water level. It is calculated on basis of the Nordheim equation with the measured periods T together with some calculated parameters like the delayed neutron fraction β_{eff}, the prompt neutron lifetime l, the relative fraction of each delayed neutron group a_i and the corresponding decay constant λ_I

The uncertainty on the value of the reactivity coefficient due to the experimental uncertainties (period, water level increment) is estimated to be between 3 and 5%.

3.3. **Axial and horizontal fission rate distribution**

The axial and horizontal fission rate distribution can be used directly for validation purposes by comparing the measured and calculated fission rate distribution. Moreover, the axial fission rate distribution is used to determine the axial buckling. The axial buckling is measured with an uncertainty between 2 and 4%. The axial fission rate distribution is measured with an uncertainty from point to point of approximately 2% for UO, rods and 3% for MOX rods. For the horizontal fission rate distribution the uncertainty from pin to pin is estimated to be 1% for UO, rods and 1.5% for MOX rods. These numbers account for comparison of similar rods. If one compares different types of rods (e.g. UO, and MOX), an additional systematic uncertainty of 1.4% has to be taken into account due to different fission yield and selfshielding corrections for U and Pu.
Both the axial and horizontal fission rate distribution are measured by gamma-scanning.

The axial fission rate distribution is determined by measuring 30 to 40 positions on one fuel rod. From this the axial buckling can be derived by fitting the results with a cosine function.
The horizontal fission rate distribution is determined by measuring an amount of 50 to 100 rods. On each rod 5 positions are measured around midplane, together with a measurement of the monitor. The measurement results are corrected for decay, axial fission rate distribution, gamma self-shielding and fission yield.
An alternative for using gamma-scanning is the use of fission chambers. Fission chambers can be loaded either at the position of a fuel rod or diagonally in between two fuel rods. In both cases there is a perturbation of the fuel configuration due to the lack of one fuel rod or the introduction of the fission chamber, therefore the gamma-scanning method is preferred.

3.4. **Spectrum indices**

In some validation cases general fission rate distributions do not give sufficient information. In these cases more information is requested about the contribution to the fission (or other nuclear reaction) rate by the different isotopes. Therefore some spectrum indices can be measured. A spectrum index is the ratio of two different reaction rates. Spectrum indices that are measured at the VENUS are F5/F9, F8/F9 and C8/F9. F and C stand for fission and capture reactions, resp., while 5, 8 and 9 stand for ^{235}U, ^{238}U and ^{239}Pu. Random uncertainties on the measured values are between 2 and 2.5%.
A spectrum index is measured by placing two foils in a dismountable fuel rod at the same distance from midplane. The foils are chosen in function of the spectrum index that is measured, e.g. a ^{235}U and a ^{239}Pu foil for the F5/F9 spectrum index. After irradiation the foils are unloaded and measured by gamma-spectrometry (Ge-detector). Dependent on the reactions that are considered, the activities of fission products or activation products are measured and corrected for e.g. foil mass, decay, gamma self-shielding, fission yield, branching ratio, etc. Since both foils have been irradiated under the same conditions, no corrections for the position of the foils are necessary.
For some spectrum indices fission chambers can be an alternative for foil measurements by performing measurements with e.g. both ^{235}U and ^{239}Pu fission chambers.

3.5. Fission rate distributions inside a fuel rod

Radial fission rate distributions inside a fuel rod have been measured in the past. This method has been innovated and can also be applied for the measurement of the angular fission rate distribution (flux tilt). The method is based on irradiation of a foil and cutting the foil into pieces. The activity of the pieces is a measure of the fission rate distribution. The uncertainty of the activity measurement per piece is estimated to be 2%.

3.6. β_{eff}

Two methods are applied to measure the effective delayed neutron fraction:

- the critical size increase
- the negative reactivity insertion

These methods have been described elsewhere [6].

4. THE VIPEX-PWR PROGRAMME

The aim of the VIPEX-PWR programme is to determine reactorphysical parameters of MOX-assemblies that are mainly of interest for reactor operation. With help of these measured parameters reactorphysical codes can be validated with respect to these specific features. It is sponsored by SCK•CEN, Belgonucléaire, BNFL, KAERI, CRIEPI and MHI (including the Japanese utilities).

The VIPEX-PWR programme has been divided in two phases. During phase 1 the delayed neutron fraction β_{eff}, the ^{241}Am effect and the control rod worth have been determined. Phase 2 dealt with the flux tilt, the moderator density effect on reactivity and the detector response and fission rate distribution.

Figure 2 shows the overall layout of the PWR MOX configuration that has been studied in the VIPEX-PWR programme.

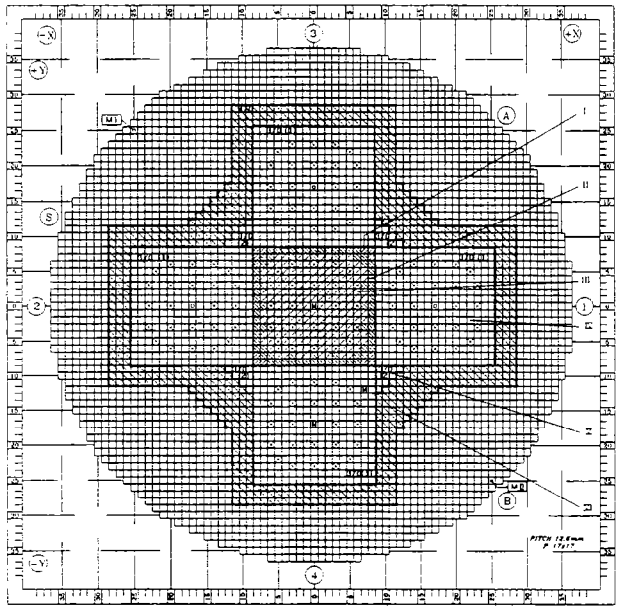

Figure 2. The basic VIPEX layout

Other aspects of the shown configuration have been studied in the VIP programme. For a good link with this programme the shown MOX configuration has been loaded with all the fuel rods at exactly the same position as in the VIP programme.

The fraction of delayed neutrons β_{eff} is very important for reactor control. For ^{239}Pu it is about twice to three times as small as for ^{235}U. So in principle plutonium becomes much faster prompt critical than uranium. In mixed plutonium-uranium configurations this effect is off course less pronounced, but it is still very important to have a good quantification of the delayed neutron fraction. Furthermore many reactivity effects are measured in the framework of the VIPEX-PWR programme, and a good knowledge of the value for β_{eff} is indispensable for the evaluation of these measurements.
The β_{eff} measurement is based on a method called negative reactivity insertion, which is verified by the increase of critical size method.

The ageing of plutonium by the decay of ^{241}Pu to ^{241}Am has a significant influence on the reactivity of a MOX assembly.
The ^{241}Am effect has been measured by determination of the critical height of the VIPEX configuration and comparison with the results from the VIP programme.

For a determination of the difference between “cold” and “hot” condition of a reactor, in other w words during start-up or operation, the reactivity effect of the moderator density has been investigated.
The moderator density effect on the reactivity has also been measured by determining the critical height of several configurations. The original VIPEX configuration has a moderator density of 73% compared to the moderator density at room temperature. The density reduction is obtained by introduction of aluminum rods between the fuel rods. To measure the effect of moderator density these aluminum rods have been pulled from the configuration in three subsequent steps. After each step the critical height has been measured to determine the reactivity effect.

The reason for measuring the flux tilt is that the peripheral rods of the MOX assembly are subject to a large shift of the neutron spectrum, since the neutrons from the UO_2 assembly have a lower average energy than those from the MOX assembly. The fission cross-section for low energy neutrons is higher, resulting in a higher reaction rate for these neutrons. This is partially compensated by the higher neutron flux from the MOX assembly; still large power differences inside the fuel rod are expected.

It has already been mentioned that the neutron spectrum in a MOX assembly is faster than in a UO_2 assembly. Since the neutron absorbing capacities of control rods are mainly based on the absorption of thermal neutrons (B, Cd), their effectiveness (control rod worth) is less in a MOX fuel assembly.
The control rod worth has been measured by inserting stepwise control rods at several positions and determining the resulting difference in critical height.

In a nuclear power plant the power of some fuel assemblies is monitored by fission chambers. Since the neutron spectrum in a MOX fuel assembly is harder, the response of the fission chamber will be different in the MOX assembly. To determine the difference in response of a fission chamber in a UO_2 and a MOX assembly the so-called detector response has been measured.
The detector response has been measured by placing identical fission chambers in the center of the MOX assembly and the UO_2 assembly and measuring the countrate at several power levels. The fission chambers have been interchanged of position and again the countrates have been measured at several power levels. In this way the ratio can be determined of the response of a fission chamber in a MOX assembly and in a UO_2 assembly.
Au and Mn foils have been irradiated in the same positions to investigate whether there could be a large gamma contribution to the fission chamber countrate.
The horizontal and vertical fission rate distribution has been measured in order to have an experimental indication of the power in the MOX assembly and the UO_2 assembly.

5. RESULTS OF THE VIPEX PROGRAMME

In this section several parameters measured in the framework of the VIPEX programme are compared to calculated values [4]. The (C-E)/E values will be given.

5.1. ^{241}Am effect

The (C-E)/E value of the ^{241}Am effect is 5.8%. The reactivity effect by the ingrowth of ^{241}Am is calculated by multiplying the measured difference in critical height of the previous and present configuration with the measured reactivity effect. The measured reactivity effect also evolutes in the course of time, therefore it has been evaluated and a mean value has been derived. The uncertainty of the evaluated value is estimated to be 4%. Since the uncertainty of the difference in critical height is only 1%, the total uncertainty of the reactivity effect is estimated to be 4%.

5.2. Control rod worth

Several control rod configurations have been investigated. A comparison between the measured and calculated values is given in table 1. The measured value has been determined by fitting the values of the reactivity coefficient with respect to the reciprocal third power of the critical level plus extrapolation length.

$$\frac{\partial \rho}{\partial h} = a\frac{1}{h^3} + b$$

where a and b are fitting parameters.

Table 1. Control rod effect. Difference between calculation and measurement

start configuration	end configuration	(C-E)/E (%)
basic	4 control rods	2.3
4 control rods	4 CR plus extra feeding zone	5.8
4 CR plus extra feeding zone	8 CR	1.4
8 CR	12 CR	1.3
12 CR	16 CR	2.9
16 CR	16 CR plus extra feeding zone	10.2
16 CR plus extra feeding zone	20 CR	1.8
total		**1.9**

From the table it appears that the calculation of the influence of an extra feeding zone is less accurate than the calculation of the influence of control rods, the actual goal of this experiment. No explanation has been found for this phenomenon. Since the measured reactivity effect of the control rods has an uncertainty of about 3%, the calculations of the control rod worth fall within the uncertainty range.

5.3. Detector response

The measured detector response gives a good agreement with the calculation (WIMS 7b, 1998 AEA Technology). The C-E/E value of the count rate ratio between the MOX and UO_2-assembly is 1.2%. The calculation of the detector response measured in the framework of the previous programme VIP-PWR resulted in a C-E/E value of 0.4%.
The uncertainty of the measured value is 0.3%. It can be concluded that the calculation gives a result with an uncertainty of about 1%.

5.4. Fission rate distribution

The C-E/E values are shown in figure 3. For the MOX assembly the standard deviation of the C-E/E values is 0.8%, for the UO_2 assembly this is 1.5%. These values are comparable to the measurement uncertainties; 1.5% and 1% for MOX and UO_2, respectively.

5.5. **Flux tilt**

A comparison between the calculated and measured maximum value of the flux tilt shows that the difference is less than 1%. However, the calculated and measured position of the maximum differ 45". In this case the flux tilt has been determined by measuring the activity of an Al foil surrounding the irradiated fuel pellets.

Another type of measurement has been performed, where the irradiated fuel pellets have been measured with a very narrow (2mmx2mm) collimator in order to measure directly the fission rate distribution in the pellet. These measurements have been performed with a low accuracy, but they confirmed the distribution as measured via the activity of the Al foil.

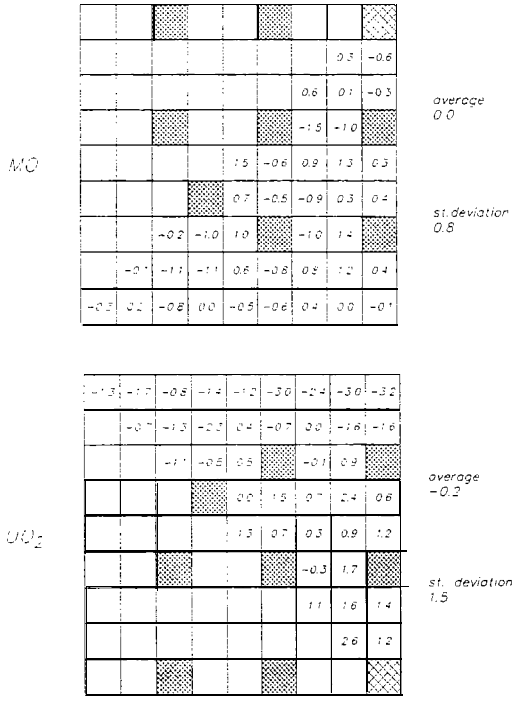

Figure 3. (C-E)/E values

6. FUTURE PROGRAMMES

6.1. **The VIPOX programme**

The VIPOX programme is an extension of the VIP0 programme, where the aim was the creation of a series of benchmarks for void calculations in highly enriched MOX lattices.

The strength of the VIPOX programme is the fact that the critical height of the investigated configurations will be increased to about 80 cm. In this way the axial leakage is reduced considerably (- factor 3). An eventual increase of the radial leakage can be countered by introducing boron in the water moderator.

Critical level and water level reactivity effect will be measured for validating criticality calculations. The horizontal and axial fission rate distribution will be measured for the validation of power distribution calculations. Some spectrum indices will be measured optionally (F5/F9, F8/F9, C8/F9) for even finer evaluation of the power distribution.

The void box to be used will be bigger than the previously used one for a higher void effect.

6.2. **The OVERMOX programme (overmoderated MOX configurations)**

The OVERMOX programme aimed at investigating overmoderated 100% MOX configurations. There are several ways to increase the moderator-to-fuel ratio: enlarging the lattice pitch, replacing selected MOX fuel pins by water holes and reducing the fuel pin diameter. It is obvious that the last way is the most expensive and the less interesting from a market point of view since technological compatibilities between actual cores and overmoderated cores are not maintained.
Preleminary calculations with respect to criticality, kinetics (prompt neutron life time, effective delayed neutron fraction) and F/M ratio have been performed. They show that several overmoderated configurations can be loaded in VENUS taking into account the available infrastructure. Full MOX square configurations with lm active length and pitch lattices of $1.26*\sqrt{2}, 2*1.26, 1.303*\sqrt{2}, 2*$ 1.303 could be experimentally investigated. The withdrawal of MOX fuel pins to achieve the overmoderation could occur in a PWR configuration.

6.3. **A programme proposal for the investigation of weapon-grade plutonium.**

For the investigation of weapon-grade plutonium a limited amount of MOX rods of type 0.7/4.3, that contain 4.3% Pu and natural U, are available at the VENUS, as shown in table 2. However, only about 25 rods with 50 cm fuel stack length of this fuel type are available, which may limit the experimental possibilities.

There are also about 400 MOX rods available with weapon-grade plutonium. A major drawback of these rods is that they contain only 1% Pu and that the plutonium is mixed with 3% enriched uranium. These rods are not considered in the programme proposal for the moment.

Some preliminary calculations have been performed to investigate the usefulness of these rods. These calculations are reported in [5].
To investigate the behaviour of weapon-grade plutonium in a nuclear power plant, it is possible to execute an experimental programme that determines basic parameters for neutron code validation in a BWR core with 4.3 enriched weapon grade plutonium. This programme can be executed with presently available fuel rods.

A similar programme can be considered for PWR cores where a quarter of a PWR assembly can be studied. Also BWR Island MOX cores can be investigated.

Table 2. Characteristics of fuel rods available at the VENUS critical facility

Fuel type	$^{235}U/Pu$ (w/o)	Isotopic composition Pu (w/o) 238/239/240/241/242	Fabrication process	Number of rods
MOX	0.6/4.8	1/63/24/8/4	MIMAS	75
MOX	0.3/5.4	1/63/24/8/4	MIMAS	66
MOX	0.3/9.7	1/61/24/9/5	MIMAS	215
MOX	0.4/14.3	1/62/24/9/4	MIMAS	229
MOX	3/1	0/92/7/1/0	vibrated	408
MOX	2/2.7	0/79/17/3/1		470
MOX	1.7/3	0/79/17/3/1		65
MOX	0.7/4.3	0/96/4/0/0	pelleted	25
MOX	0.7/5.1	0/79/17/3/1	pelleted	62

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REPROCESSING OF MIXED U-Pu FUEL BY RECRYSTALLIZATION IN MOLTEN MOLIBDATES

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Abstract

The method of mixed uranium - plutonium dioxides fuel reprocessing consists in its dissolution (after the cans separation) in the $MoO_3 - Na_2MoO_4$ melt and in the precipitation of the homogeneous solid solutions $(U,Pu)O_2$. On this stage the plutonium content in the melt is corrected. During the dissolution process the volatile fission products (I, Kr, Xe, T) are escaped from the melt as soon as the non-volatile ones remain in the melt in the molibdate form. The melt is separated from $(U,Pu)O_2$ crystals including fission products imitators or without ones. The rate of purification from fission products as large as 2-3 orders has been achieved.

1. INTRODUCTION

Pyrochemical and pyrometallurgical methods of spent nuclear fuel regeneration attract attention of investigators since the appearing of the spent fuel regeneration problem. The waited distingueshnesses of these methods are follows: the compactness of the technology, apparatus and wastes; the radiative stability of the used inorganic reagents, and, consequently, absence of the limitations on the activities level; high chemical activity of the reagents and high processing rates because of high temperatures; the possibility of the compact reprocessing plants creating using pyrochemical technologies near power stations. With these conditions the ecological safety of the nuclear fuel cycle would be rised.

The pyrochemical methods of the oxide fuel regeneration may be classified in the next directions: the gaseous methods, the solid state methods, the liquid salt - metal extraction methods, the crystallization methods, the refining, the precipitation from a melt, the electrodeposition from a melt [1] The methods of precipitation from a melt with low melting temperature as easy and compact ones attract the largest atttention of investigators. Some chloride, fluoride, nitrate, sulfate and alkaly metals melts are proposed to be used. After carrying out some analysis of the defects of these melts we have offered use the molybdates systems. Molten molybdates widely are used for the recrystallization of different refractory oxides but not for UO_2, PuO_2.

2. PHYSICO-CHEMICAL PRINCIPLES

For investigation of the regeneration process with the recrystallization of UO_2 (PuO_2) in molten molybdates we have selected the follow systems: $Na_2MoO_4 - MoO_3$, $BaO - MoO_3$, $ZrO_2 - MoO_3$, $La_2O_3 - MoO_3$, $CeO_2 - MoO_3$, $RuO_2 - MoO_3$, $UO_2 - MoO_3$, $UO_3 - MoO_3$, $PuO_2 - MoO_3$, $UO_2 - Na_2MoO_4$, $U_3O_8 - Na_2MoO_4$, $PuO_2 - Na_2MoO_4$, $U(MoO_4)_2 - Na_2MoO_4$, $Pu(MoO_4)_2 - Na_2MoO_4$. Our investigation has been carried out by using the termography, the rentgenography, the radiography and other methods. Then we have separated fields of the UO_2, PuO_2, $U(MoO_4)_2$, $Pu(MoO_4)_2$ crystallization in the $UO_2 - MoO_3 - Na_2MoO_4$ and $PuO_2 - MoO_3 - Na_2MoO_4$ triple systems.[2].

All above mentioned oxides have been found to be soluble in molten $Na_2MoO_4 - MoO_3$ with the formation of the corresponding molybdates. Then UO_2 and PuO_2 being dissolved in molten $Na_2MoO_4 - MoO_3$ at 1000°C are crystallyzed after melt cooling.

It was found that the molybdates of uranium, plutonium and fission products are not isostructural with uranium and plutonium dioxides. This fact exepts the co-crystallization of the called molybdates with uranium and plutonium dioxides.

During the recrystallization of UO_2 in $UO_2 - MoO_3 - Na_2MoO_4$ melt by cooling from 1000 °C to 800 °C in an inert atmosphere the tracer-labelled fission products imitators (Cs, La, Zr, Nb) were separated with decontamination efficiency up to 99.9 %.

During the recrystallization of commensurable amounts of UOZ and PuO_2 by cooling from 1000 ° C to 800 ° C in an inert atmosphere homogeneous solid solution $(U,Pu)O_2$ was precipitated with concentration of PuO_2 corresponding the initial proportions of PuO_2 / UO_2.

During the recrystallization of PuO_2 in $PuO_2 - MoO_3 - Na_2MoO_4$ melt by cooling from 1000°C to 800°C in an inert atmosphere the fission products imitators (Cs, La, Zr,) were separated with decontamination efficiency more than 99 %.

The commensurable amounts of UO_2 and PuO_2 may be separated by recrystallization of PuO_2 in molten $MoO_3 - Na_2MoO_4$ with the simultaneous oxidation of uranium to $Na_2U_2O_7$ and by the dissolution of $Na_2U_2O_7$ in the melt.

After the melting of solid solutions $(U,Pu)O_2$ (20 w. % PuO_2) in air atmosphere 99 % of the uranium amount passed to the melt and 97 % of the plutonium amount separated as crystals. These crystals were washed by a new melt Na_2MoO_4.

Due to the recrystallization of $U(MoO_4)_2$ in the melt $UO_2 - MoO_3$ with real fission products the separation to 98 - 99 % for ^{137}Cs, $^{144}Ce + ^{144}Pr$, $^{95}Zr + ^{95}Nb$ has been achieved.

During the recrystallization of $U(MoO_4)_2$ in the $UO_2 - MoO_3 - Na_2MoO_4$ melt in an inert atmosphere with 5 w. % of plutonium the plutonium separation up to 99.9 % has been achieved.

The UO_2 recrystallization process was verified on the pilot plant with 1 kg UO_2 charge. The follows simulators of fission products were used: the non-radioactive cesium, barium, strontium, zirconium, lantanum, ruthenium isotopes as oxides. The separation of: Cs, La - 99.9 %, Ba, Sr - 99.7 %, Zr -99 %, Ru - 98 % has been achieved.

There were determined the limiting minimal concentrations of fission products after many cycles of UO_2 recrystallization : $^{106}Ru + ^{106}Rh$ - 3.3 . 10^{-9} w. %, $^{144}Ce + ^{144}Pr$ - $8.2 \cdot 10^{-9}$ w. %. Apparently, these amounts of impurities remain in the UO_2 lattice points.

3. THE REGENERATION PROCESS

The uranium and plutonium dioxides recrystallization may be used for the regeneration of the spent oxide nuclear fuel. With this purpose the separated from the can fuel is melted with $MoO_3 - Na_2MoO_4$ mixture of the determined proportion at 1000 "C in an inert atmosphere and after the total dissolving of the fuel and after the separation of volatile fission products (iodine, tritium, xenon, krypton) the melt is cooled. As a result the solid solution $(U,Pu)O_2$ corresponding to the initial proportion of UO_2 and PuO_2 is precipitated. On this stage the plutonium concentration correction is possible. After the recrystallization all non-volatile fission products remain in the melt. The melt and the crystals are divided. The continuous elimination of the crystals, the washing by the recent Na_2MoO_4 melt and by water, drying and directing to distant producing of fuel elements are supposed. The melt is used many times.

From the sodium molybdate aqueous washing solution by adding of acids molybdic acid is precipitated and after heating it turns into the molybdenum trioxide which can be used again. The irreversible losses of molybdenum as a part of wastes are l-5 % of the used amount of molybdenum salts. As a result of many cycles the melt accumulates large amounts of the non-volatile fission products. By gradual adding of alkaline metals oxides or carbonates the uranium and plutonium dioxides are consequently precipitated from the melt and on the second stage the oxides of fission products are precipitated. The last ones are conditioniered and bum.

By the changing of the MoO_3 and Na_2MoO_4 proportions in the melt and by the changing of the temperature the conditions for the concentration and crystallization of Np, Am and Cm as oxides are created.

The molybdate processes may be used for the separation and escaping from the melr of the uranium and plutonium in the commensurable proportions too.

The whole totality of these processes ensures the process continuity and the full closeness of the regeneration cycle. Another merit of the above subscribed process is the fact that an oxide fuel dissolving reagent is a salt bath component, namely molybdenum trioxide, not agressive gases (chlorine, fluorine, hydrogen chloride. hydroden fluoride).

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FABRICATION AND CHARACTERIZATION OF MOX FUELS WITH HIGH PLUTONIUM CONTENT USING ALTERNATIVE PROCESSES

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Abstract

The Institute for Transuranium Elements (ITU) in Karlsruhe is part of the Joint Research Centre of the European Commission. Its particular aims are (i) to perform nuclear R&D in support of EU policies, (ii) to further enhance the Institute's role as a recognized centre of European basic actinide research, (iii) to contribute to an effective nuclear safeguards system in Europe and elsewhere, and (iv) to strengthen the position of the European industry by evaluating and testing the potential for technological and medical applications of transuranium elements. In particular, ITU is actively engaged in the performance of the R&D programmes "Safety of Nuclear Fuels" (development of advanced fuels able to sustain safely high burnup operation) and "Mitigation of Long Lived Actinides and Fission Products". Within the framework of these projects, ITU is participating in several international research groups on plutonium incineration (CAPRA [1]) or Partitioning and Transmutation (EFTTRA [2]). Development of advanced LWR MOX fuels is also being pursued in close collaboration with the nuclear fuel cycle industry. This paper describes the fabrication methods developed for MOX (and other Pu-bearing compounds) at the Nuclear Technology Department of ITU. Results obtained from fabrication of MOX fuel at very high Pu content are presented and discussed. Finally, examples of MOX fuel fabrication (LWR representative, with high Pu content) leading to large grain formation are presented.

1. Fabrication processes

For MOX fuels, three processes, leading to different fuel structures are being developed at ITU: powder metallurgy, the liquid (SOL GEL) process, and a third option, which combines the first two: a hybrid route, called SOLMAS.

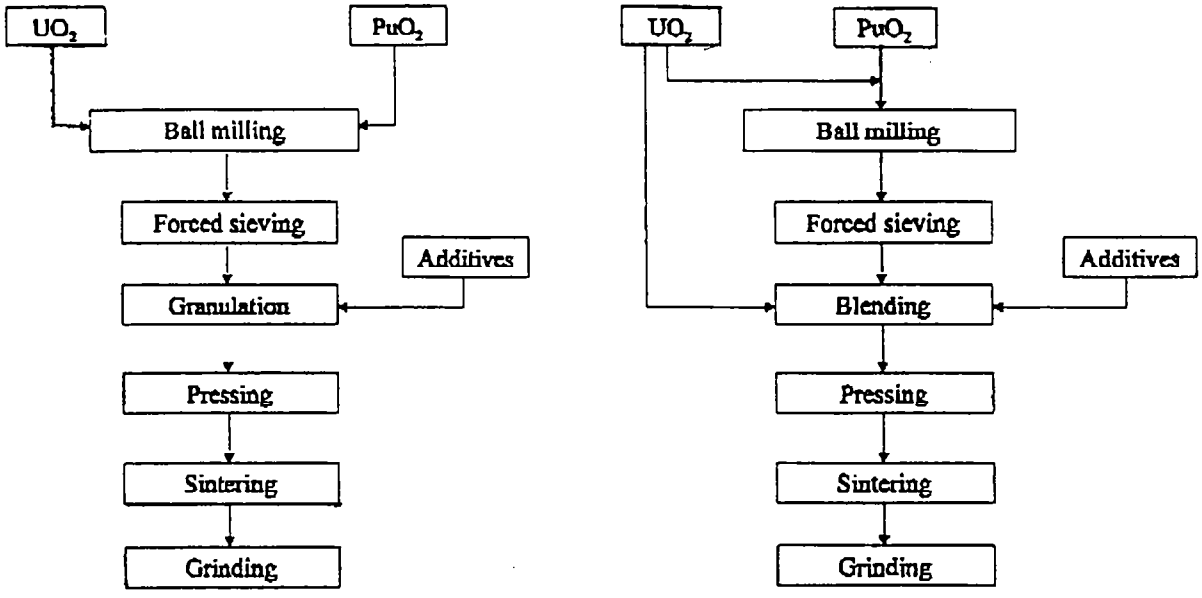
1.1 Powder metallurgy

The powder metallurgy process consists of a variety of methods of oxide powder mixing. Each method is selected to produce the desired fuel, as far as its main properties are concerned: degree of homogeneity, fuel specifications (LWR or FBR), dissolution properties, plutonium content. Similar processes, used commercially in Europe (BELGONUCLEAIRE, BNFL, COGEMA [3]) are well proven on an industrial scale. However, the developments of modified fuels and the testing of new parameters, like UO_2 powder type, role of additives, sintering conditions, need to be done on a laboratory scale, prior to their qualification on an industry level. ITU's programmes on powder metallurgy cover two aspects:

- within the CAPRA programme, ITU has fabricated experimental fuel pins with very high plutonium content (up to 45% Pu/U+Pu); in some cases, neptunium was also added to the MOX fuel [4];

- ITU's research involves the development of LWR MOX fuels that are able to sustain higher burnups. For this purpose, methods to produce large grains are being tested.

Practical experience of ITU in MOX fabrication by powder metallurgy mainly includes two processes, shown in the flowsheet in Fig. 1. Direct Powder Blending is commonly used for FBR applications, while the MIMAS method is used for LWR fuels. The products obtained by both processes meet the usual commercial requirements in terms of geometry, density, thermal stability, O/M or chemical impurities. Moreover, particular microstructures can be obtained, as shown later.



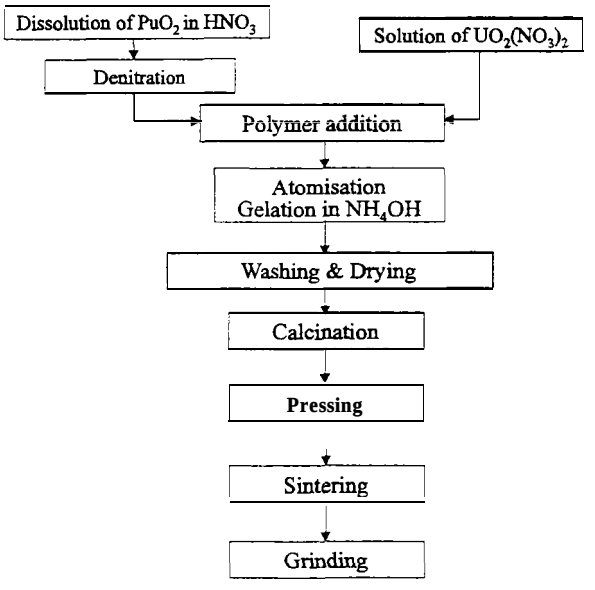
1a) Direct Powder Blending 1b) Micronized Master Blend (MIMAS)

Fig. 1: Powder metallurgy fabrication processes at ITU.

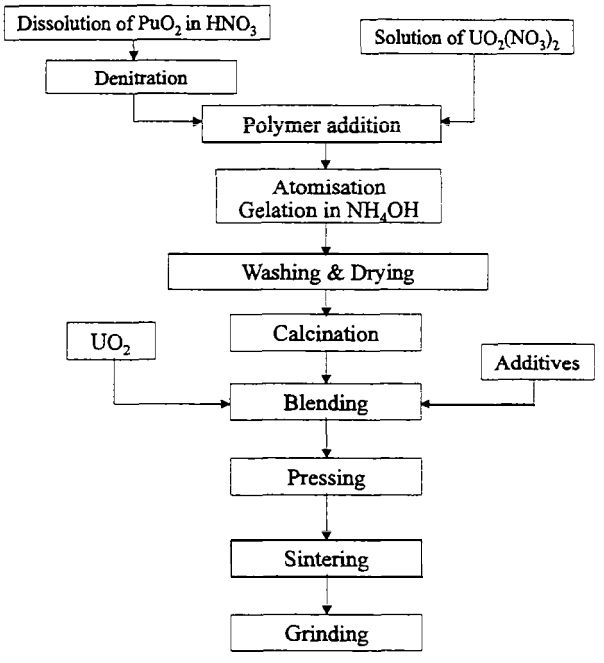
Fig. 2: SOL GEL and SOLMAS processes The use of powder metallurgy processes is limited by its application to highly radioactive components, e.g. when using plutonium with high neutron emitter (Pu238) content (high burnup Pu) or high gamma emitter (Am241) content (aged Pu), or in the extreme case when minor actinide fuels and targets have to be manufactured within the Partitioning and Transmutation framework. The limitation of this process arises due to the formation of dust during fabrication, and from the resulting difficulty in operation, maintenance or decommissioning of the equipment. For these reasons, the SOL GEL and SOLMAS processes, based upon the use of liquid phases of active material, have been developed in our Institute.

1.2 SOL GEL process

The SOL GEL [5] process involves the dissolution of the starting materials in nitric acid (alternatively, if located at the reprocessing plant, the active solutions are readily available). As shown on the flowsheet in Fig. 2, the uranyl and Pu nitrate solutions are mixed in the required proportion. To produce the feed solution, the viscosity is adjusted by addition of trace



2a) SOL GEL



2b) SOLMAS

Fig. 2: SOL GEL and SOLMAS processes

amounts of long chain organic thickeners. This feed solution is dispersed into droplets, which are then collected in a hydroxide (typically ammonia) bath, where gelation occurs within the original droplet, so that near spherical beads are formed. After washing and drying, the beads are in a hydroxide form, from which they are converted to oxide by thermal treatment in an oxidising atmosphere (typically 400 °C in air). The beads are then thermally treated in a reducing atmosphere (typically 800 °C in Ar/5% H_2) to obtain $(U,Pu)O_2$. The beads are free flowing, dust free and their size varies between 10 and 200 μm. Their tap density is usually of the order of 20% of the theoretical density. This powder is then pressed into pellets, and sintered. The product meets all specification requirements for LWR or FBR fuels; it is characterized by the preparation of a fully homogeneous solid solution $(U,Pu)O_{2-x}$. For experimental purposes, it can be used as a reference compounds to study the effects of heterogeneity in the microstructure on the fuel behaviour under steady state or off-normal conditions. The SOL GEL process is particularly suited for the fabrication of oxide compounds containing minor actinides (neptunium, americium), as has been demonstrated in the TRABANT 1 [3], TRABANT 2 and SUPERFACT [6] experiments.

1.3 *Hybrid methods*

If required for experimental or practical reasons, the two methods described above can be combined. For example, we call SOLMAS the process leading to the mixture of a master blend prepared by the SOL GEL method (because it contains highly active isotopes), with non-fissile or non-active powder: UO_2 in the case of MOX fuel, or inert matrix powder (spinel, MgO . ..) in the case of target fabrication for transmutation. The advantage of the process is that the active material prepared by SOL GEL is concentrated in the master blend, thus increasing the fabrication capacity and reducing the liquid waste. The flowsheet of SOLMAS (for MOX) is given in Fig. 2b.

2. Effect of Pu content on homogeneous MOX fuel fabrication

In the TRABANT 2 experiment (irradiation of fuel pins in HFR Petten), MOX fuel at high Pu content has been fabricated by the SOL GEL process. Despite several tests and modification in the fabrication steps, the required density (95% TD) could not be met in the fuel with 45% Pu/U+Pu. Since the resintering tests demonstrated the thermal stability of the pellets, the fuel pin with the high Pu content was nevertheless loaded in the experiment (started in 1999). In order to understand the reason for the lower density, a series of SOL GEL pellets were fabricated, with Pu contents ranging between 25 and 40% Pu/U+Pu, and results of previous experiments (TRABANT 1) were also analysed. The analysis of the results of the powder characteristics (tap density, average particle size), shown in Fig. 3, lead to following conclusions:

- the tap density of the beads increases with Pu content;
- the particle size of the beads decreases with the Pu content;
- the density of the sintered pellet is constant (92-95% TD) up to 35% Pu/U+Pu where an abrupt decrease to 86% TD is observed;
- the age of the feed solution (few weeks for "fresh" solutions, more than one year for "aged" solution) influences the density of the end pellets (but not the particle size) for Pu/U+Pu greater than 40%.

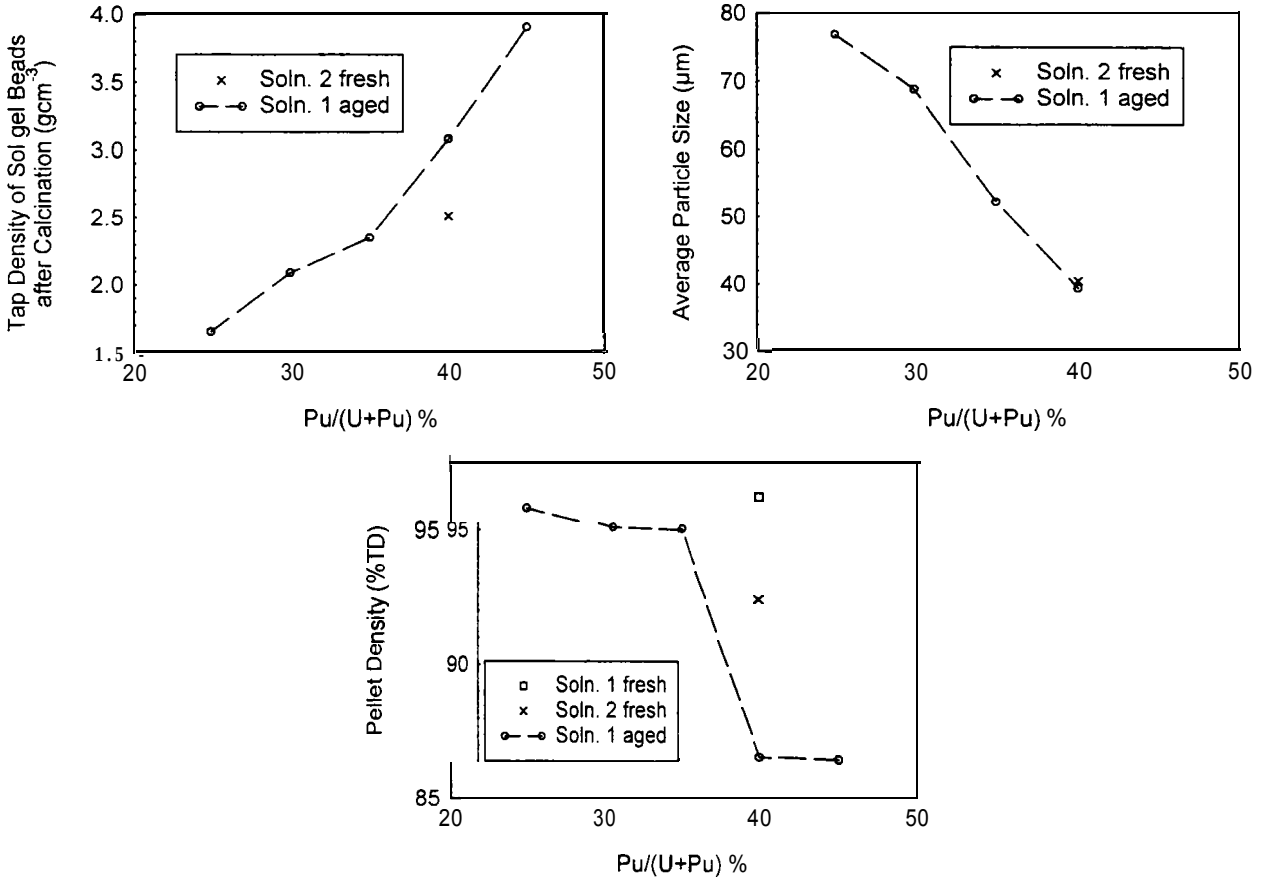
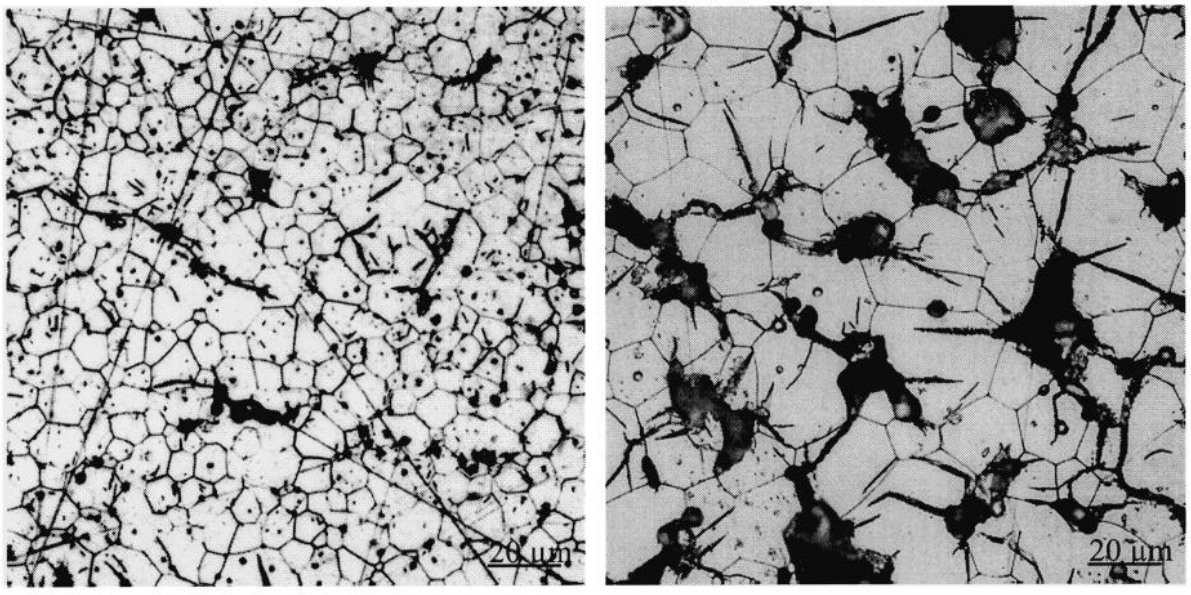


Fig. 3: Effect of Pu concentration on fabrication parameters of SOL GEL MOX fuel. We attribute these effects to polymerisation in Pu nitrate solutions [7]; this increases with Pu content, and with time. This modification of the solution properties leads to product beads which are more dense and harder. These effects have a detrimental influence on the sintering properties of the final pellets.

Examples of microstructures of MOX fuels at 40 to 45% Pu are shown in Fig. 4. Note the high porosity in the fuel at 45% Pu (low density fuel), and also the average grain size of about 10 - 15 μm in high Pu content fuel produced by SOL GEL. The sintering conditions were 1700 °C for 6 h in a dry Ar/5% H_2 atmosphere.



$(U_{0.55}Pu_{0.40}Np_{0.05})O_2$ (solution 1, fresh)

$(U_{0.55}Pu_{0.45})O_2$ (solution 1, aged)

Fig. 4: Microstructure of SOL GEL MOX fuel at high Pu content

3. MOX fuel fabrication with large grains

In UO_2 fuels, large grains (more than 30 microns size) have been tested for a number of years [8]. Their beneficial effect on the irradiation behaviour at high burnup (fission gas release) is still controversial, mainly due to the way these large grains have been obtained. Additives (TiO_2, Nb_2O_5 ...) have generally a high solubility rate in the UO_2 matrix and, during irradiation, tend to enhance the diffusion processes within the grains, leading to effects opposite to that expected. Therefore, other investigations [9,10] have studied additives that are insoluble, and form a phase at grain boundaries, which is viscous at the irradiation temperature.

MOX fuel is known to behave as good as UO_2 in the reactor, with a possible exception concerning fission gas release behaviour (under steady state and transient conditions) at high burnup. Therefore, to obtain MOX fuel with larger grains is one development underway on a laboratory scale. The problem lies in the structure of the fuel. The large grains should be obtained in the phase that is dominant (in proportion to the pellet volume), i.e. UO_2 in very heterogeneous MOX, and $(U,Pu)O_2$ in totally or partially homogeneous fuel.

At ITU, we have successfully tried two methods:

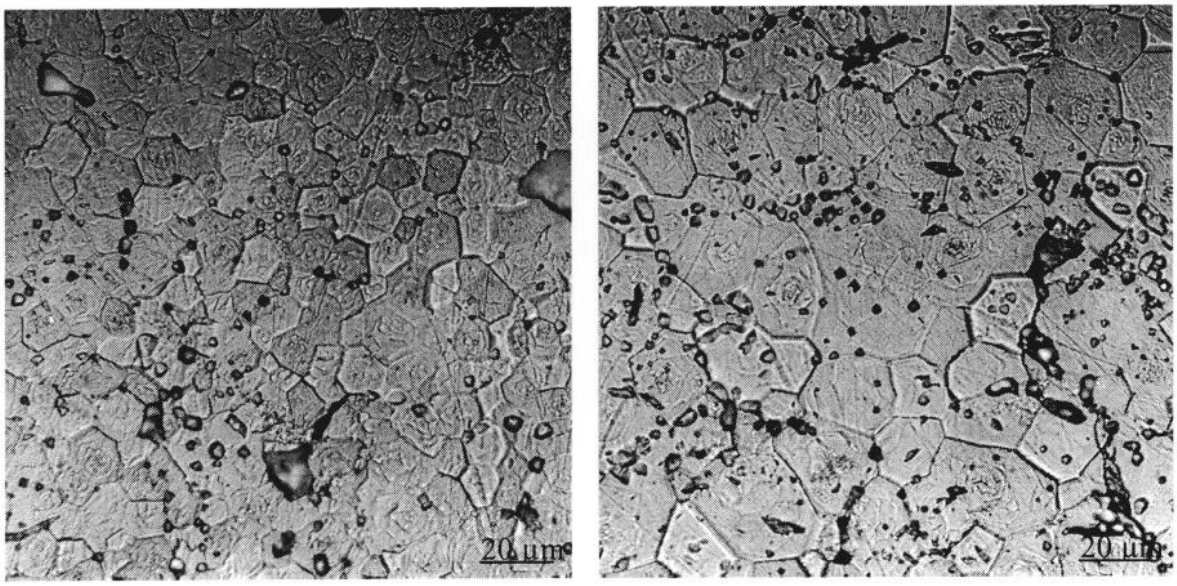
i) homogeneous SOL GEL fuel pellets sintered under dry atmosphere (Ar/H_2) lead to grain size of 10 - 15 microns (all grain size data are defined by the Mean Linear Intercept

method, without correction); this result has been shown to be independent on the Pu content (up to 45% Pu, see section 3).
After sintering under humid conditions (4 hours, $pH_2/pH_2O=33$), the grain size obtained depends on the sintering time. Grains of 20 microns on average have been obtained (Fig. 5) without additives. As the SOL GEL material is already a $(U,Pu)O_2$ solid solution, sintering is particularly effective in promoting grain growth. The high sinter activity may also be associated to relatively small crystallites produced in the SOL GEL process. This explains why large grains can be obtained without any dopant. Note that increasing the sintering time leads to further enhancement of the grain growth.

ii) in heterogeneous fuel (SOLMAS, MIMAS), large grains (either in UO_2 or $(U,Pu)O_2$ phase) are difficult to produce without additives. We have tested mullite ($3Al_2O_3,2SiO_2$) as an additive because of its ability to form a liquid phase at grain boundaries in UO_2 during the sintering phase. The addition of 0.1 w/o (mullite to total oxide) in MIMAS fuel (15% Pu/U+Pu) leads to two effects on the microstructure (Fig. 6):

- an enhancement of the Pu-U interdiffusion: the $(U,Pu)O_2$ phase becomes dominant (in volume) in the pellet. The UO_2 phase is limited to islands, in which the grains are very small (a few microns).
- an increase (up to 20 microns on average) of the $(U,Pu)O_2$ grains.

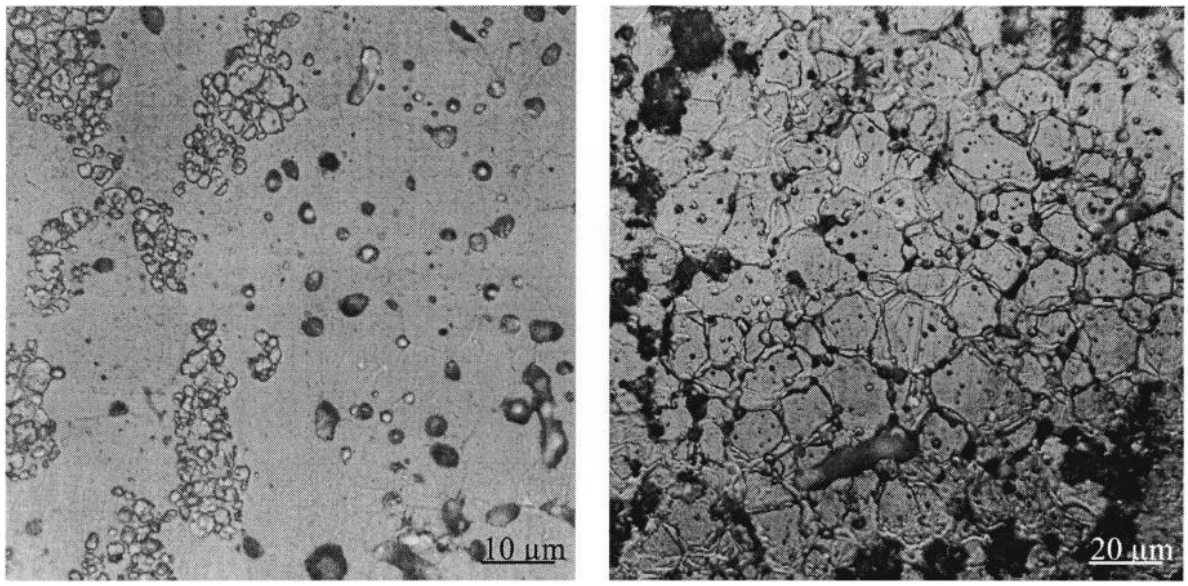
Studies are being pursued on both fuel types, to determine the effects of sintering conditions, level of additives and the degree of the homogenisation in the MIMAS-type fuel pellets.



SOL GEL 15 w/o Pu/(U+Pu)
dry sintering

SOL GEL 15 w/o Pu/(U+Pu)
wet sintering

Fig. 5: Microstructure of SOL GEL MOX fuel at 15 w/o Pu content, under dry and humid sintering conditions.



UO_2 etching $(U,Pu)O_2$ etching

Fig. 6: Microstructure of MIMAS MOX fuel at 15 w/o Pu with 0.1 w/o mullite.

4. Conclusion

Various processes for MOX fuel fabrication up to very high Pu content (45%) are being developed at ITU, within the framework of two institutional research programmes, namely plutonium incineration (CAPRA) in FBR's, and the Safety of Nuclear Fuels in LWR's. In this paper, the wet process SOL GEL and powder blending routes were described. Additional tests and characterization of the production of large grains are underway, and have to be completed by the demonstration of their good behaviour in pile. The main result of our present research is the fabrication of LWR MOX fuel with large grains. This could be obtained by the SOL GEL process, without additives, and also on MIMAS fuel, by including 0.1 w/o mullite. In the latter case, a strong interdiffusion of U-Pu, led to grain growth in the now dominant $(U,Pu)O_2$ phase.

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CLOSING STATEMENT

R.W. Stratton
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Baden, Switzerland

In his summing up of the Symposium, Mr Stratton, Chairman of the Panel Session, included two personal viewpoints aimed to motivate the participants to examine their assumptions on the long term future of MOX fuels and Plutonium recycle on their return to their laboratories and design offices. They were in the form of the two diagrams which are explained here for completeness and introduce a number of aspects which were not covered in the Symposium itself.

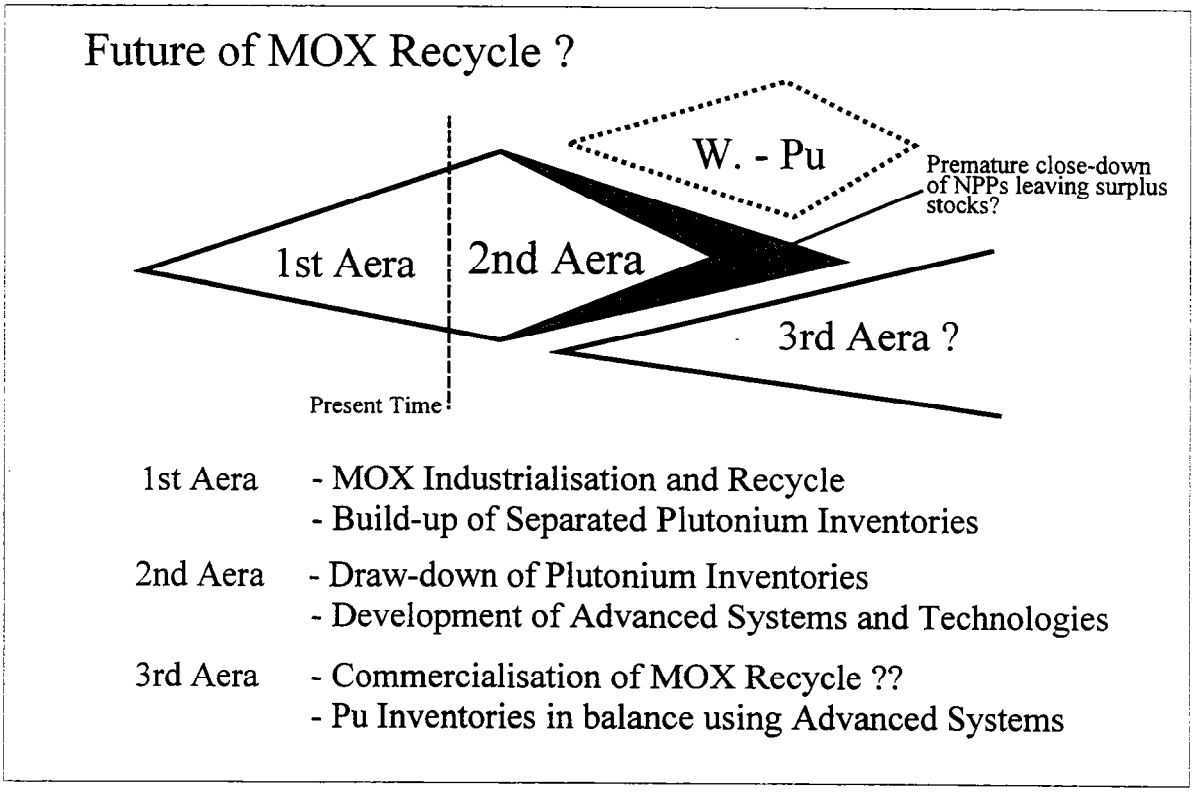

Figure 1 "The Future of MOX Recycle?

This Symposium marks a milestone in the development of MOX fuels. It may well be the last such Symposium of its type.

It comes at the end of the first era of MOX development in which the technology (alongside the reprocessing of spent fuel) has grown out of the research environment to an industrial level, with the full introduction of MOX recycle on a large scale in several countries.

This phase has also been marked by the build-up of stocks of separated plutonium.

The required Safeguards arrangements have also kept pace with the development of the technology and the volume of the MOX being handled, fabricated, stored, transported and irradiated.

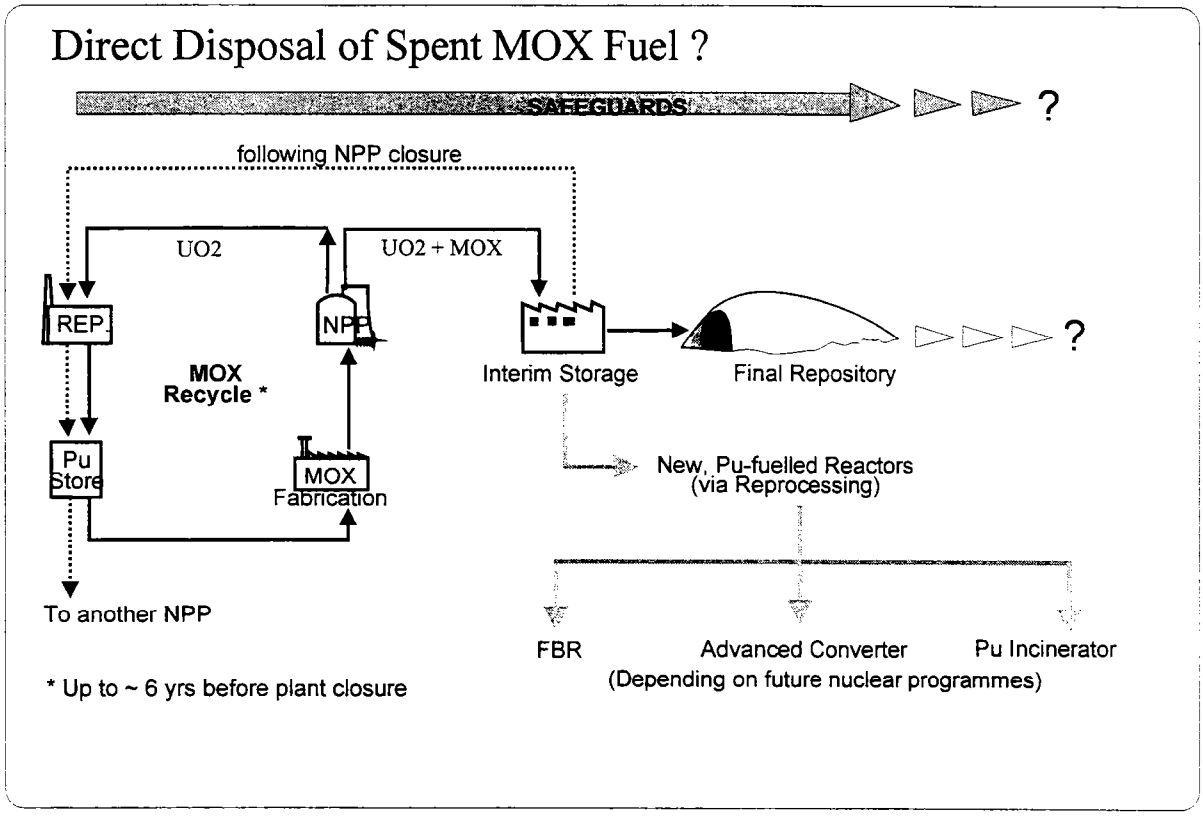

Figure 2. Direct Disposal of Spent MOX Fuel ?

The second era is now beginning. After a further expansion of MOX recycle to include other countries and additional nuclear plants licensed for MOX fuel, it can be expected that the separated Plutonium stockpiles would begin to diminish and move to significantly lower levels.

An important reservation is that for stockpile reduction, sufficient NPPs must be available and licensed for plutonium fuels recycle, and that any premature shutdown of nuclear plants may cause problems not only for the utilities concerned but also for the countries whose plants are no longer available to dispose of their plutonium reserves. For this the technology should be urgently developed for inert matrix fuels and advanced plutonium "incinerators", etc., possibly installed and operated under an international regime.

In a parallel, but only indirectly related programme, stockpiles of surplus weapons plutonium in Russia and the USA will also be recycled (in part) in nuclear power plants to make the plutonium "inaccessible" and unattractive for any future weapons use (conversion of weapons grade plutonium into "the spent fuel standard").

A third era of MOX Recycle might be envisioned in which MOX fuels become one of the standard nuclear fuels for plants specifically designed for their use. However it is very possible that MOX recycle would end at the end of the second era.

There will be no third recycle era without MOX recycle becoming commercially viable to the utilities.

The electric power utilities, newly faced with competitive open markets, can no longer subsidise the MOX and reprocessing industries using the argument of optimised resource

management and the belief in the final introduction of the long-promised fast breeder reactor systems. For there to be a third and sustainable era of MOX fuel, prices must be significantly reduced, helped in part by the introduction of new, environmentally acceptable plants and processes with minimum waste production.

In the final analysis, under the current arrangements, all spent MOX fuel assemblies must be disposed of by direct interim storage followed some 50 years later by deposition in a final repository. This is simply because at the end of life and some 6-8 years prior to closure, no more recycle in the parent plant is possible. Even the transfer to another reactor only delays the inevitable final disposal of fuel elements. This also of course applies to the final charges of spent uranium fuels but becomes acute, when as today the nuclear industry is believed by outsiders to be a declining industry.

For MOX fuel the question arises whether the direct disposal route has been sufficiently thought through regarding non-proliferation aspects. Due to the accessibility of the remaining fissile material (plutonium) increasing in future centuries, safeguards measures (accounting, containment, physical protection) will have to be applied indefinitely, in particular on repositories containing spent MOX fuel and especially if these repositories are of the newly proposed "retrievable" type. Can such a long term surveillance and control be guaranteed?

If direct disposal places an open-ended commitment on Society to safeguard the spent fuel repositories in perpetuity (unlike those for radioactive wastes and residues), then an alternative must be sought in separating plutonium from spent fuel and returning it as quickly as possible into a reactor where it is truly inaccessible. There are two scenarios to consider:

Case 1. Declining nuclear power industry, and/or premature shutdown of individual plants in one country, or the situation of a reactor reaching the normal end of its operating life:

- The need is for the development and deployment of advanced fuels and reactor systems for the complete destruction of plutonium and the actinides in so called incinerating plants. Some decades can be allowed (interim spent fuel storage) until these systems and materials have been developed.
- For those countries abandoning nuclear power an international solution (fuel cycle centres) will need to be found.
- In this scenario there continues to be a strong need for reprocessing of the spent fuel. For non-proliferation reasons alone, reprocessing cannot be abandoned.

Case 2. An expanding nuclear industry

- The need is for efficient Pu-fueled reactors which can match an expanding fuel demand by breeding, extraction and recycle of the unused fissile material, at the same time maintaining the strong non-proliferation characteristic of Pu "storage" in a power producing plant. Fast Breeder Reactors and Advanced Converter Reactors are required.
- In view of the need for a viable reprocessing industry to support this scenario, advanced processes must be developed (as in the previous diagram) to allow low-cost and environmentally benign reprocessing processes to be introduced and advanced fuel fabrication facilities to be developed all with low waste arisings.

- Case 2 also implies a strong research and development component but this can only be attained if the sources of financing are assured. The industry relies for its R&D financing on its utility customers. If the utility customers cannot survive in the liberalised market the postulated expansions will not be sustained and the source of funding needed for the advanced reactors and recycle technologies will dry up.

It must be left to Governments and Society as a whole to consider their duties and obligations, to what extent they will be required to support (subsidise) the development of these advanced systems and to put the necessary international agreements and controls in place to secure a robust proliferation-resistant fuel cycle regime in support of nuclear power in the next Millennium or to ensure its controlled and safe abandonment.

GENERAL CONCLUSIONS

(1) MOX MATURITY

This symposium was attended by more than 150 participants from 28 countries and 4 international organisations. In total, 48 oral papers and 21 posters reported on the current status of the technology and some of the major issues encountered for further development.

More than thirty years of reactor experience using MOX fuel as well as the fabrication of 2000 MOX assemblies with the use of 85 t of Pu separated from spent fuel from power reactors indicates that the recycling of Plutonium as MOX fuel in LWRs has become a mature industry. The technology is well understood and the facilities, institutions and procedures are in place (capacity extensions being planned) to meet the anticipated arisings of separated plutonium from the production of nuclear power.

The number of countries engaged on plutonium recycling could be increasing in the near future, aiming the reduction of stockpiles of separated plutonium from earlier and existing reprocessing contracts. Economic and strategic considerations being the main factors on which to base such decision to use MOX.

(2) MOX TRANSPORT

Transport of MOX fuel assemblies (as well as spent fuel transports and to a lesser extent PuO_2 powders) is a vital element in these recycle programmes but could have the potential to be a weak link in the chain. To avoid problems, it is essential that sufficient numbers of transport flasks of the required types, licensed for the increasing Pu contents, be made available in a timely manner to keep pace with the planned increases in fabrication rates. Despite the excellent safety records for radioactive and MOX transports over many decades, continuous attention should be drawn to establish the transport modalities, buffer stores, secure vehicles, and transport routes at the same time accounting for public sensitivities on radioactive transports in general and MOX transport in particular.

(3) MOX PERFORMANCE, DESIGN AND SAFETY

A large number of technical presentations and posters updated and reconfirmed the good and almost defect-free performance of MOX fuel at increasingly high burn-up levels. MOX fuel is designed to meet the same operational and safety criteria as uranium fuels under equivalent conditions. This is also confirmed by the parallel development of design codes to accommodate the special characteristics of MOX. The neutronic behaviour of mixed cores of uranium and MOX fuels can also be modelled in a satisfactory way.

To reach burnup levels equivalent to uranium fuels up to 70 MW d/kg, work is still ongoing to confirm models of fission gas release and limiting this release by changing the fuel microstructures. Increased fuel plasticity (creep) helps to explain the superior performance of MOX fuel in ramping tests.

Some results from the RIA tests in the French CABRI Programme indicate that further careful study of MOX fuel behaviour under extreme transient and accident conditions at high burnup is needed. As stated above, MOX fuel is designed to meet the same key safety parameters as for uranium fuel for all normal and off-normal events and it needs to be demonstrated at each core loading that there is no challenge to the design basis.

Integral and specific parameter testing of MOX fuel in normal and off-normal operation is under way in a number of countries with particular emphasis on high burnup behaviour. Here the important contributions of the OECD/NEA Halden BWR programme should be mentioned.

(4) DISPOSITION OF SURPLUS WEAPONS GRADE PLUTONIUM

The reactor-based weapons-grade plutonium disposition approaches proposed by the United States of America and the Russian Federation build upon proven commercial MOX fuel technologies. It is subject to their own special rules and time scales:

- The political dimension and direct involvement of the governments (disarmament and arms-control)
- The specific geographical locations involved which govern the scale and scope of the associated transport programmes
- The need for transparency and safeguards
- The absence of a 'commercial' component while recognising the significant financing problems
- Complex interlinkage of international agreements.

The goal of the US and Russian plutonium disposition programmes is to render the surplus plutonium as inaccessible and unattractive for retrieval and weapons use as the residual plutonium in spent fuel from power reactors.

(5) TECHNICAL IMPROVEMENTS

(involving cost reduction, environmental and safety aspects)

It was noted that the following developments were taking place:

- Extending MOX burnup to ≥70 MWd/kg HM with increasing Pu contents
- Further developments of transport flasks for MOX fuel assemblies
- Waste reduction (reprocessing and fabrication)
- Operator dose reduction
- Direct disposal of spent MOX fuel
- Advanced cost effective reprocessing and MOX Fabrication methods
- Development of efficient Pu fuelled reactors and their fuel cycles
- Development of advanced MOX fuels (low FGR, inert matrix, etc.)
- Demonstrating the applicability of current fuel safety criteria for advanced use of MOX fuel.

(6) INSTITUTIONAL IMPROVEMENTS

(involving international co-operation management, policies, safeguards)

It was noted that the following developments were taking place:

- Establishing an international framework for the management of excess civil Pu inventories also in the case of premature reactor closures.
  - exchange procedures and controls
  - storage facilities
  - centres for Pu-incineration
- Establishing a stable and publicly acceptable transport environment
- Ensuring Safeguards are adequate for the new technologies and management regimes
- Complete WG Pu disposition
- Introduction of Pu-fuelled reactors appropriate to nuclear power requirements
- The IAEA and OECD/NEA should continue in a complementary way to perform studies and analysis on design, fabrication and performance data as well as on updating fuel cycle information on MOX and Pu-recycle and disposal, e.g. :
  - Data Bank (OECD/NEA)
  - Working groups/technical meetings (e.g. IAEA IWG on Water Reactor Fuel Performance and Technology, IWG on Fuel Cycle Options; OECD/NEA Working Party on Pu Recycling and Innovative Fuel Cycles, Expert Group on the Integrated Assessment of the Nuclear Fuel Cycle)
  - Fuel cycle codes
  - Public information.

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CHAIRPERSONS OF SESSIONS

Opening Session	V.M. MOUROGOV	International Atomic Energy Agency
Session I	A. BONNE	International Atomic Energy Agency
	P. WILMER	OECD Nuclear Energy Agency
Session II	L. HEINS	Germany
	J. MALONE	United States of America
Session III	L. BRUNEL	France
	S. NOMURA	Japan
Session IV	F.G. RESHETNIKOV	Russian Federation
	D. FARRANT	United Kingdom
Session V	P. DERAMAIX	Belgium
	F.C. DIMAYUGA	Canada
Panel Discussion	R.W. STRATTON	Switzerland

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