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Recycling of plutonium and uranium in water reactor fuel

Proceedings of a Technical Committee meeting held in Newby Bridge, Windermere, United Kingdom, 3–7 July 1995

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FOREWORD

Mixed oxide (MOX) fuel used in power water reactors is made of a mixed uranium and plutonium oxide and clad with zirconium alloy.

The utilization of MOX fuel has been regarded for many years as limited to fast breeder reactors (FBR). Since the mid-1980s, and the delay of FBR programmes in many countries, it has been thought that the plutonium produced in reprocessing plants should be used in water reactor fuels instead of being stored.

MOX fuel has been industrially produced and is now a mature technology. However, beyond the obvious difference in fabrication technology, the experience accumulated on MOX fuel is less than that accumulated on uranium fuel; as a consequence MOX fuel has been licensed for a lower burnup than uranium fuel. To reach comparable performance MOX fuel has to be studied further and more accurate models of thermal behaviour, fission gas release, or pellet-clad interaction, have to be established. The database on MOX fuel irradiation should be expanded especially in the area of high burnup.

For countries with a large amount of weapons grade plutonium the option of burning this plutonium in MOX fuel is now under consideration.

The Technical Committee Meeting on Recycling of Plutonium and Uranium in Water Reactor Fuel was recommended by the International Working Group on Fuel Performance and Technology (IWGFPT). Its aim was to obtain an overall picture of MOX fabrication capacity and technology, actual performance of this kind of fuel, and ways explored to dispose of the weapons grade plutonium. The subject of this meeting had been reviewed by the International Atomic Energy Agency every 5 to 6 years and for the first time the problem of weapons grade plutonium disposal was included.

The papers presented provide a summary of experience on MOX fuel and ongoing research in this field in the participating countries. The meeting was hosted by British Nuclear Fuels plc, at Newby Bridge, United Kingdom, from 3 to 7 July 1995. Fifty-six participants from twelve countries or international organizations took part.

The IAEA wishes to thank BNFL for the organization of the meeting and all the participants for their contributions to this publication. The officer of the IAEA responsible for the organization of the meeting and the compilation of this TECDOC was P.M. Chantoin of the Nuclear Fuel Cycle and Materials Section.

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SUMMARY

1. INTRODUCTION

All the contributions in this meeting were on plutonium recycling, mixed oxide (MOX) fuel fabrication and performance, and weapons grade plutonium disposal.

Fifty-six participants from twelve countries or international organizations attended the formal presentations and panel discussions.

The papers were presented in five sessions: MOX and Recycled Uranium Fuel Utilization, MOX Fabrication and Reprocessing, MOX Fuel Design, Plutonium Disposal and MOX Fuel Performance.

The technical discussions were conducted in three panel discussions: MOX Fabrication, MOX Fuel Performance and Utilization of Weapons Grade Plutonium in MOX Fuel.

The main findings of this meeting are that the production of MOX fuel is increasing worldwide and should be sufficient, in the near future, to absorb most of the plutonium production and to respond to the increasing demand. The database of MOX fuel has to be extended for both irradiated and non-irradiated fuel to allow further burn-up increase. The MOX solution to the problem of disposal of weapons grade plutonium was considered to be very valuable.

The technical discussions are summarized below.

2. MOX FABRICATION - Panel 1

2.1. Present capacity and status

The key to the issue of reducing the present stockpile of plutonium and keeping it at a minimum in the future is to match the MOX production capacity and subsequent utilization to the Pu output at the reprocessing plants. In France this is already becoming policy with the ultimate aim of having a Pu inventory of not more than a 2 year requirement as a working buffer stock.

The panel attempted to define the expected MOX manufacturing capacity known today. For reference the 16 t/a Pu generation rate in La Hague equates to a 230 t/a MOX fabrication requirement which is related to the design, burnup and isotopic composition of the feed plutonium. The detailed annual throughput for a given capacity will depend on batch size and number of Pu contents. Table I shows the revised expected capacities without the Hanau plants and ignoring Russian expectations due to lack of firm information. Table II shows in more detail the Franco-Belgian production plans based on the icronized master blend (MIMAS) process.

Product flexibility (range of fuel types produced) will increase over the next decade. For example while MELOX will remain a basic 17 × 17 PWR plant for EdF type fuels, Cadarache will cover PWR and BWR for mainly German customers. A MELOX extension (50 t/a) would produce PWR and BWR MOX fuels of all designs as would a new plant if built at La Hague (2 × 50 t/a lines). In the United Kingdom the Sellafield MOX Plant (SMP) is being designed to make PWR, BWR and fast reactor fuels and in France a small (10 t/a) fast reactor fuel production capacity will also be retained.

The Dessel line P1 of Belgonucleaire could be considered as an alternative to one or the other of the French extensions noted above. In Japan the plant size and process to be used is still under review but will clearly handle both PWR and BWR fuels when built. Adequate capacity is also provided in Japan to cope with fast breeder reactor (FBR) and advanced thermal reactor (ATR) requirements. The independent planned programme of MOX recycle in India was noted as well as the difficulties facing the Republic of Korea with their large reactor programme.

It appeared that any capacity for dealing with Pu from military stockpiles would have to be provided by a new facility and should perhaps incorporate special design features due to the nature of the material.

The chairman noted the importance of having adequate MOX transport capabilities and drew attention to possible changes in flask licensing and testing brought about by forthcoming changes in the revised IAEA Safety Series No. 6, Regulations for the Safe Transport of Radioactive Materials. This applied particularly to the air transport of MOX powder and fuel assemblies.

2.2. MOX fabrication process

There has been an unexpected reduction in MOX processes being used. Since the last meeting in 1989 the COCA and OCOM processes have disappeared, the latter due to the demise of the Hanau MOX plant. The processes now available are MIMAS and the Short Binderless Route (SBR). At least two process routes are desirable and should be maintained to preserve diversity of supply. Regardless of the process there are still many improvements which could be made to reduce fabrication costs, improve product quality and increase efficiency.

One of these improvements might, for example, be improved interchange between the vendor specifications perhaps leading to common standards in some areas (e.g. solubility, impurity levels, pellet damage, broadening isotopic tolerances). This could benefit both the supplier and the utility customer. For the customer it is also important to maintain the diversity of supply and the element of competition.

The panel was unwilling to enter into a fundamental discussion of the costs and price of MOX fabrication. The best available data is still the MOX fuel cycle cost study of the OECD/NEA of 1989 and the fuel cycle study of 1994. The actual prices are always dependent on the contract conditions, giving, for example price reductions for large batch orders.

2.3. Future developments

For the longer term (specifically in Europe) it was thought unlikely that any fundamental new fabrication process could or would be introduced commercially before the installation of major new reprocessing facilities with which it would be associated. The introduction of a new process could also coincide with the need to deal with the reduction of military plutonium in specially designed reactors. In the commercial area, product or process improvements must provide a price incentive to the customer.

When considering the long term evaluation of the Pu fuel cycle over the coming decades, account needs to be taken of the opportunity offered by the CANDU system in synergy with the LWR spent fuel strategy.

For the user, the long term requirement is not to limit any aspect of the use of UO_2 fuel and to retain full operating flexibility in-reactor. The burnup of MOX fuel must increase at least in line with UO_2 fuels, and if possible exceed them to realize the added economic potential of high burnup MOX.

The panel endorsed the view that MOX fuel plants needed to meet the highest standard of safety and noted that a minor incident could close down a plant permanently as a result of heightened public or political sensitivity.

Similarly the heightened demands on MOX fuel behaviour called for an increased robustness in design and product quality.

3. MOX FUEL PERFORMANCE - Panel 2

3.1. Summary of discussions

It is essential that the fuel produced by fuel fabricators satisfies the customer's requirements - it must allow the utilities to operate their reactors safely, reliably and economically. By assessing the performance of the MOX fuel under both normal and off-normal conditions fuel fabricators can provide the evidence required by the customers and safety authorities to assure them that their fuel is safe and reliable.

At present European experience of MOX irradiations extends to 52 GWd/t (rod average burn-up) in commercial PWRs and up to 60 GWd/t in experimental assemblies. The operational modes include load-following conditions. In addition there have been many demonstration assemblies irradiated in reactors throughout the world.

The main conclusion from these irradiations is the good general behaviour of the MOX fuel at burn-ups up to 5-55 GWd/t. Specific features include:

- Good dimensional behaviour of assembly components and fuel rods
- Good water side corrosion behaviour of the cladding
- Internal rod pressures which are not significantly different from those for UO_2 under comparable conditions
- Good PCI resistance, even superior to UO_2
- No apparent relationship between fuel microstructure and enhancement of fission gas release
- Failed fuel behaviour similar to UO_2.

In addition the acquisition of physical properties data has progressed well since the last IAEA technical committee meeting on MOX fuels in 1989.

A number of items, however, which require further investigation, can be identified. Panel discussion of these items led to the following conclusions and recommendations.

- Further acquisition of physical properties data for all manufacturing routes is required - this will reveal any possible relationship with the plutonium content and with the microstructure.

TABLE I. COMMERCIAL MOX FABRICATION CAPACITY AROUND THE YEAR 2000

	Plant	Operator	Product	Start-up	Nominal annual capacity (tHM/y)	Process
Belgium	Dessel PO	Belgonucleaire	LWR	1973	35	MIMAS
	Dessel P1*			?	50 or 0	MIMAS
France	Cadarache	Cogema	LWR/FR	1990	30/10	MIMAS/COCA
	MELOX		LWR	1995	160 plus 0 or 50	
	La Hague				0 or 50	
	(New line)*					
Japan	Tokai	PNC	ATR/FR	1972	10/10	Standard blending
	Rokkasho *	not known	LWR(?)	2005(?)	100(?)	Open
UK	MDF	BNFL	LWR	1993	8	SBR
	SMP	BNFL	LWR	1998	120	SBR

* Note that authorization to construct these plants has not yet been obtained.

TABLE II. MOX FUEL FABRICATION TO 2004
Detail: Plants in France and Belgium (in tHM)

	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004
Cadarache*	2	10	15	20	30	30	30	30	30	30	30	30	30	30
PO (Dessel)	35	35	35	35	35	35	35	35	35	35	35	35	35	35
MELOX*					25	86	110**	135**	160	160	160	160	160	160
MELOX extension									30	50	50	50	50	50
and/or La Hague U2***												30	50	50
and/or P1 **** (Dessel)												30	50	50
Total	37	45	50	55	90	151	175	200	255	275	275	275-335	275-375	275-375

* Capacity as fabricated or expected to be fabricated)
** Subject to the receipt of the necessary authorization (modification of the decree)
*** U2 = modular-flexible (1st stage: 50 t max; 2nd. stage: add 10 t FBR fuel; 3rd. stage: add 50 t max.)
**** P1 = subject to licensing authorization

It was agreed that the scope of the database should aim to approach the same level as that of UO_2. It was also agreed that the database should cover both fresh and irradiated fuel, in particular fuel tested in in-pile instrumented experiments.

It was also noted that techniques used should be tested for consistency in an attempt to reduce the scatter in the data; this would be of value in both the UO_2 and MOX fuel areas. Control of stoichiometry during laboratory testing was raised as a specific point requiring attention.

- While it was agreed that internal corrosion was unlikely to ever be a problem in MOX fuel rods it was accepted that the situation should be continuously monitored.

- Further acquisition of fission gas release statistics is required at high burnup under steady state and ramp conditions.

  This will reveal any possible relationships with plutonium content, microstructure, stoichiometry etc.

  The topic of helium release at high burnup was raised. It was agreed that more information was required with regards to the production and release.

- It was agreed that the rim effect if it occurs is associated with local burn-up level and not with the original fuel microstructure.

- It was suggested that a rationalization of specifications may result in a reduction of fabrication costs of MOX fuel without prejudice to safety.

- It was believed that MOX fuel at high burnup will not behave differently from UO_2 in accident conditions in terms of a RIA (reactivity insertion accident). Experimental investigations which are foreseen in the near future could provide support for a safety demonstration.

Finally, it was concluded that consolidation of all MOX fuel data in a single document would be a valuable exercise.

4. UTILIZATION OF WEAPONS-GRADE PLUTONIUM IN MOX FUEL - Panel 3

This panel looked at some issues associated with reactor options (mainly MOX fuel) for disposing of military plutonium. The intent is to render the plutonium more proliferation resistant.

4.1. Reactor options for positioning plutonium

The discussion began with a description of 3 reactor options that would achieve this aim.

a) Denaturation

Weapons-grade plutonium would be rendered more proliferation resistant by modifying one or more essential nuclear or physical attributes of the plutonium:

Critical mass: Plutonium would be made more proliferation resistant by increasing the higher plutonium isotopes. Civilian plutonium is less effective as a weapon than weapons-grade plutonium (around 94% Pu-239); however, there is no international consensus as to the effectiveness of civilian plutonium as weapons material. One immediate means of reducing the proliferation risk from military plutonium would be to dilute it with civilian plutonium. (This would be difficult in the context of some national nuclear policies.)

Neutron activity: Increasing the neutron activity of the plutonium would render it less attractive as a weapons material by reducing the effectiveness through premature detonation. This could be achieved by increasing the O, Pu-238, Pu-240 or Am-241 content of the plutonium, by oxidizing the military plutonium and mixing it with civilian plutonium.

Heat rating: The heat generated by alpha-decay of Pu-238 or Am-241 would make it more difficult to assemble in an explosive device.

One paper in this meeting examined the high level of Pu-238 generated in a mixture of weapons uranium and plutonium in an inert matrix as a means of providing a high degree of proliferation resistance through the neutron activity and heat rating.

b) MOX Fuel

Mixing the plutonium with UO_2 was suggested as an option for achieving an immediate increase in the proliferation resistance of the plutonium. The increase is, however, small, since a simple nitric acid dissolution could isolate the Pu.

One paper looked at using CANDU for burning military derived plutonium as MOX fuel in existing reactors. Other papers discussed conventional MOX fuel from reprocessed LWR fuel; quarter cores of weapons plutonium could likely be accommodated in existing PWRs, while full core MOX fuel could likely be utilized with BWRs.

It was pointed out that the so-called spent fuel option does not eliminate the risk of diversion, and that the plutonium could still be extracted in future generations. The diversion times for plutonium in various forms defined by the IAEA take into account the relative accessibility of the plutonium.

c) Plutonium annihilation

One paper showed that the high neutron economy and on-power refueling would enable over 80% of the plutonium to be annihilated if mixed with an inert matrix and burned in CANDU. Near total elimination could be achieved upon subsequent passes through the reactor.

There are currently many experiments, both in- and out-reactor, exploring potential inert matrix materials. This is acknowledged to be a longer-term option.

4.2. Processing technology

Public figures indicate that there will be a total of about 150 t of military plutonium released from all warheads in the United States of America and the Russian Federation. This would add only about 20% to the civilian plutonium to be processed during the same period.

Use of this as MOX fuel would require conversion of the metal 'pits' to oxide, MOX fuel fabrication, and transport to reactors. Conversion of the metal to oxide was not seen to be a problem technically, although processing facilities would have to be built. New MOX fuel fabrication capability would have to be built to meet these additional fabrication requirements. Current and projected MOX fuel fabrication capacity in Europe is planned to meet only the needs from reprocessing civilian spent LWR fuel. The French repeated their position, reported in Obninsk, that their nuclear programme maintained an equilibrium between the output from their reprocessing plants, and MOX fuel fabrication requirements, and that fabrication of MOX fuel from military plutonium in French facilities would not be compatible with this policy.

The multitude of security and safeguards systems associated with the large inventories of civilian plutonium and commercial use of MOX fuel could be applied to military plutonium processing.

In terms of transport, the impact of new transportation regulations drafted by the IAEA on transporting MOX fuel was raised. It was hoped that the ability to transport plutonium or MOX fuel by air would continue.

4.3. Fuel technology

During this conference, PWR and BWR fuel designs for civilian MOX fuel were described. It was felt that the designs would not be much different for military plutonium. A fuel design for accommodating military plutonium in either a conventional 37-element CANDU bundle, or in the new CANFLEX bundle was also described in this conference. Further validation of codes, for either military or conventional plutonium is required. The smaller delayed neutron fraction, and the smaller Doppler effect with weapons plutonium will require reassessment of reactivity initiated accident response of MOX fueled LWRs.

SESSION 1
MOX RECYCLED URANIUM FUEL UTILIZATION

MOX IN FRANCE: STATUS AND PROSPECTS

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Abstract

At the outset of its nuclear programme, France opted for a closed fuel cycle. After the strong slowing-down of the FBR implementation, emphasis have been carried on the plutonium recycling in PWRs. In 1985, the current policy was defined and the industrial investment was decided. Now the whole industrial system is being operated. 1) Spent fuel reprocessing: Every year, EDF unloads 1200/1300 tons of spent fuel. 850 tons are reprocessed by UP2, 800 tons of slightly enriched uranium and 8 tons of plutonium are recovered. 2) Manufacturing facilities: The annual EDF's need is about 110/136 tons (e.g. 240/300 assemblies). In Europe, more than 300 tons are required by 2000. 3) The "moxified" reactors: The 900 MW units are reloaded with UO2 and MOX assemblies. The share of MOX is 30 % (16 assemblies for a 3 cycle-loading and 12 for a 4 cycle-loading). The annual production corresponds to 20 and 25 reloadings. Today 16 reactors are open and authorization is required for 12 additional units. 4) Prospects: The consequences of this clear policy is analysed and the situation in 2005/2010 is predictable. The future of the Pu recycling is varying with the international constraints and the evolution of the nuclear requirement. Four ways are studied :

- continuation of the same policy (850 t spent fuel, 110/120 t MOX)
- reprocessing of all the spent fuel (1300 t spent fuel, 160 t MOX, 28 PWRs)
- reprocessing of the irradiated MOX
- introduction of Pu burners.

1. INTRODUCTION

From the very origin of the French nuclear programme, reprocessing of spent fuel and recycling of valuable materials such as uranium and plutonium has been the thorough and coherent French strategy. Today, recycling in water reactors is the option chosen to close the fuel cycle. It implements a set of well-mastered industrial operations, the impact on environment and human health of which is in accordance with the ALARA principle. At three places, the same flow of recycled materials is to be achieved: at the output of the reprocessing plants, at the input the fuel manufacturing facilities and at the input of the recycling-dedicated reactors.

Plutonium can be recycled in reactors, since its energetic content is still important: one gram of plutonium is equivalent to almost two tons of oil. The possibility for the use of plutonium into Mox fuel appears economically sound, and that is why EDF decided in 1985 to recycle plutonium in some of its PWR 900 units.

Thanks to reprocessing and plutonium recycling, spent fuel is no longer considered as an issue, but as a value. While reducing the demand for natural uranium, recycling also offers the great advantage to reduce significantly the volume and potential radiotoxicity of long-lived nuclear wastes.

The French nuclear power programme is an outstanding success both in terms of national independence, preventing reliance on finite fossil fuel, and as regards the economy and the balance of trade. Besides, Mox fuel, having a thirty years experience, is about to rapidly attain an equivalent operating experience as its « elder » UO2.

2. SPENT FUEL REPROCESSING

During the beginning of this decade, reprocessing has become a fully demonstrated industry. The techniques have been proven. Activities, begun over 30 years ago in order to improve quality, productivity and safety, have been patiently pursued and are being strengthened even more.

EDF's spent fuel undergoes reprocessing in the UP2 plant at La Hague, operated by Cogema:

- The UP2 plant, which started operation in 1966, had reprocessed various types of fuel up to 1987, after which it was dedicated to LWR fuel only. In order to meet the requirements of France's nuclear power programme, the annual capacity of UP2 has been recently increased from to 400 tons to 800 tons by adding new processing facilities.
  Moreover, there is another reprocessing plant at La Hague, operated by Cogema.

- The UP3 plant successfully started operations in 1990 and its first ten years of operation, has been assigned to the reprocessing of 7000 tons of spent fuel from other European and Japanese utilities. Its production is ramping up according to schedule from 350 tons reprocessed in 1991 to full capacity operations in 1995.

  As of the end of 1994, total reprocessed fuel at La Hague facilities amounts to near 7000 tU. Plant reliability and availability are well demonstrated by reprocessed quantity.

Tons per year	UP2-800	UP3
1994	573	700
1995	850	800

This unique industrial complex will be able to reprocess 1650 tons of spent fuel, serving 60 to 80 LWRs and offering high quality recyclable energetic products and conditioned residues. In the French case, the reprocessed quantity will yearly yield 8 to 8.5 tons of separated plutonium. It should be noted that reprocessing of Mox fuel has been successfully tested in 1992, during a 4,5 tons campaign.

The favourable performance of the La Hague plants is the result of an approach to safety in which safety features are incorporated early in the design phase, particularly those relating to containment not only for normal operation, but for maintenance and repair operations as well.

The La Hague plant design has made real strides in plant performances in the area of waste volume minimisation, following the ALARA principle.

Furthermore, the operating experience has shown outstanding operational achievements in terms of process performances as well as environmental impact, occupational exposure and waste management. Mainly based on an improved effluent management, on the use of additional evaporation capacities and on new volume reduction techniques. Further steps in waste minimisation are currently implemented or planned in the near future. Since 1995,

practically all the activity is routed towards the vitrification units, thus eliminating the need for precipitation and bituminisation of low and medium level effluents.

This substantial experience of reprocessing at La Hague demonstrates the industrial mastery of commercial reprocessing. France masters the reprocessing process: over 99,88% of the plutonium and the uranium contained in the fuel is separated and recovered, in order to be recycled in nuclear fuel manufacturing, while the rest is confined in the glass and other final waste forms. Another consequence is the very high values of decontamination factors, up to one hundred times the design specifications.

The reprocessing of more than 850 tonnes per year of the spent fuel discharged from French reactors is now a hypothesis which is under consideration.

As regards long term trends, technical options will depend on political evolution but an extension of reprocessed quantities can be contemplated, up to the total discharged spent fuel, including irradiated Mox fuel.

3. MANUFACTURING FACILITIES

Mixed oxide fuel is now commercially well-established and is playing an important role in France. Since a number of years, plutonium oxide is routinely transported from La Hague to the Mox fabrication facilities, and more than 200 tons of fresh Mox fuel assemblies have been transported to the EDF reactors.

a) MELOX

Melox, which is dealt with in another presentation, will be the EDF's main producer of Mox assemblies. The Melox plant (with a nominal capacity of 120 tHM/years which can be easily extended to 160 tHM/y) is located in Marcoule, enters in service in 1995 and includes all manufacturing operations from UO2 and PuO2 powders reception to assembly delivery. It appears as the first high-throughput Mox fuel manufacturing facility to be put into operation in the world. It is designed to fulfil the requirements coming from foreseeable trends in both fuel management and modern safety and environment concern.

Although dedicated to a number of Framatome assembly designs, MELOX offers utilities a flexible use of Mox fuel. As a result, it must receive and handle a wide range of basic nuclear materials. This implies the capability of using high-burnup and aged plutonium, which has consequences, through plutonium 238 and americium content, both on the specific thermal energy to be evacuated and on radiation shielding to be installed. On the other hand, the trend to an increased Mox fuel discharge burn up leads to a high plutonium content in the fabricated fuel. In the light of Melox's large scale factor, Mox fuel manufacturing costs are expected to be lower. The reduction of the annual dose of most of the factory operators to a value of 5 mSv/yr was an important objective. Besides, a design effort has been achieved in waste minimisation.

The main milestones and the manufacturing programme are presented elsewhere: startup activities are on schedule. Full qualification will be achieved by the end of summer.

MELOX will be able to supply Mox assemblies for 20 to 25 reloads of EDF 900 MWe units per year, which corresponds to the 8 to 8.5 tons of plutonium that are separated at La Hague.

b) Other plants

MELOX has taken full benefits on the outstanding Mox fuel experience of the existing facilities at Dessel and Cadarache. The activity of the two plants, Belgonucleaire Dessel and Cogema Cadarache will be adapted with the total need for Mox fuel (EDF and other customers.).

The fabrication of Mox fuel was initially realised in the BN Dessel plant (with an annual capacity of 35 tons). Cogema Cadarache plant was previously dedicated to FBR fuel, but since 1989, has been modified to produce Mox fuel in parallel with the assemblies of FBR. It will reach a 30 tons production this year.

Even if the MELOX plant is at full operation, the Mox fuel fabrication capacity will not be enough to absorb the plutonium separated in France by the reprocessing of French and foreign fuel. Cogema considers the possibility of increasing its capacities in order to serve customers who want their MOX fuel to be fabricated in France.

4. IN- REACTOR RECYCLING

The aim of EDF's strategy is to avoid the creation of a plutonium stockpile larger than what is needed by global operational conditions (i.e. about 2 years of normal utilisation). This means that the same flow of plutonium should exist simultaneously at the outlet of reprocessing plants, at the throughput of the fabrication facilities and at the input of moxified reactors.

Another EDF's target is to make Mox fuel as similar to UO2 fuel as possible, avoiding penalties on associated UO2 fuel as regards achievable burn ups and reaching the same operational flexibility as classical UO2 fuel.

4.1 Present status

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 of one-third of the core (annual cycle). This corresponds to 16 Mox assemblies per reload, out of 52. The average concentration of plutonium is limited to a level of 5,3%.

a) Present number and hybrid management

At present, 7 units out of the sixteen already licensed for Mox fuel operation are loaded with this fuel. In four of them, the proportion of Mox has reached the equilibrium. These reactors operate in baseload with a third core reloaded annually; two of them are authorised for load follow operation, for demonstration purposes. The target today is to obtain an homogeneous management of 4 cycles. EDF has decided to use

temporarily an « hybrid management » : 3 cycles for Mox fuel and 4 cycles for UO2 (3,7% enrichment). It will be applied to the other Mox units in 1995. The demonstration is performed in Dampierre 2 and Gravelines 4. Such a core management allows to draw benefit from the advantages of uranium four batch refuelling while securing an acceptable degree of burn up of the Mox fuel.

b) Plutonium recycling in 900 MWe EDF reactors

7 reactors loaded	St Laurent B1 & B2 (1987) Gravelines B3 & B4 (1989) Dampierre 1 (1990) & 2 (1993) Blayais 2
9 reactors licensed	Dampierre 3 & 4 Tricastin 1, 2 ,3 & 4 Gravelines B1 & B2 Blayais 1
12 reactors compatible	Gravelines C5 & C6 Blayais 3 & 4 Chinon B1 through B4 Cruas 1 through 4

4.2 Near term trends

a) Number of reactors

EDF will progressively burn Mox fuel. A total of 16 reactors are already licensed in France to use Mox fuel (7 units in 1994, increasing to 9 in 1996 and to 16 in 1998). 12 additional reactors are technically designed to receive Mox fuel, but they still have to undergo the licensing process (the first of these might be the four Chinon B units.)

20 to 28 reactors could receive Mox fuel, thus absorbing the entire plutonium output of Cogema's UP2-800 reprocessing plants and requiring most of the MELOX capacity.

b) Load follow

According to EDF's goal of getting same services from Mox fuel as from uranium fuel, implementation of load follow is desirable. The involved operating modes (frequency adjustment and power level modulation) have been authorised in St Laurent reactors for testing, and EDF is expecting soon a generic licensing from French Authorities. Operation in load-following mode is presently authorised on an experimental basis at the Saint Laurent B1 and B2 reactors. Operators at St Laurent have not experienced any difficulty during load-following mode operation.

c) Increase of burn up

The maximum authorised value of average Mox fuel assembly burn up is 36 GWd/t. This rate has been effectively achieved in three units, and the corresponding assemblies were unloaded without having caused any problem during irradiation. In order to improve industrial Mox performance, araised burn up should be implemented as soon as the French Safety Authorities agree. By the year 2000, over 16 reactors will be loaded with Mox fuel at high burn up (about 45 GWd/t) and a 4-cycle core management policy. By the end of 1992, 4 Mox fuel assemblies have been reloaded for a fourth cycle in order to extend data-base of Mox fuel behaviour and to prove that such burn ups are actually achievable without any problem. It could represent future Mox management. This trend in the case of Mox provides an economy in the fuel cycle cost, since the fabrication cost is independent of the plutonium content.

d) Increase of plutonium content

Research focuses on increasing the Mox burn up required by the 4 cycle management, on developing new Mox fuel design with plutonium content up to 6,5% that will answer EDF's need for higher burn ups with sufficient margins. Beyond the year 2000, the target is to reach discharge burn ups of 52 GWd/t with a plutonium content greater than 9%.

5. PROSPECTS

Today, the policy is clear and well-defined. It will reach equilibrium in 2000. Several solutions can be considered afterwards :

a) Continuation of the same policy

This possibility can be contemplated, but in this case, by 2010, several thousands tons of spent UO2 fuel, as well as more than 1000 tons of Mox irradiated fuel, will have to be stored.

b) Increasing the reprocessed quantities

This will lead to reduce spent fuel storage

c) Reprocessing of the irradiated Mox fuel

In principle, the reprocessing of Mox fuel is not different from that of the UO2 spent fuel and it has already been studied since the end of 1985 by Cogema.

Mox assemblies have been designed to be reprocessed in La Hague and the guarantee of their reprocessing was one of the points to be demonstrated to the French Safety Authorities. This leads to the monitoring of fresh pellets solubility during fabrication. In November 1992, a 4,7 ton campaign of reprocessing Mox fuel was conducted at UP2 plant and gave quite satisfactory results. Cogema has planned to adapt the UP2-800 plant to the Mox fuel reprocessing.

d) Multi-recycling

Beyond first recycling, a long term strategy involves multirecycling, whereby spent Mox fuel is reprocessed and a second generation plutonium is recovered and reintroduced 10 to 15 years later as Mox fuel. Such successive recycling offer a means to reducing the total plutonium inventory in the coming decades.

e) Introduction of 100 % Mox reactors

The all-Mox reactor concept is currently under study. This concept provides several advantages :

- a homogeneous core,
- a simpler assembly design and management (no zoning),
- a reduced number of reactors receiving Pu-containing fresh fuel

An increased moderation ratio could allow burning plutonium in better conditions. Advanced PWR reactors accepting 100 % Mox loading will burn plutonium at a rate of 60-80kg Pu T/Wh.

f) Introduction of Fast Neutron Reactors (FNR)

When fuelled with 30% Mox, as currently implemented, a water reactor gives a near zero balance in plutonium. FNR can operate in breeder or burner mode. The burner mode results in a negative plutonium balance, and has the added advantage of burning « dirty » plutonium, coming from multi-recycling. The French programme CAPRA is aimed at designing such a core.

CONCLUSION

We are now in a position to assert that the technical and industrial maturity of the RCR (Reprocessing, Conditioning and Recycling) strategy in France has been reached. Through a consistent and large-scale programme, recycling of plutonium in water reactors is fully mastered. From the operational point of view, plutonium is about to be used in the same conditions as uranium. As far as economics are concerned, plutonium is likely to take the best place: when increasing the discharge burn-up, which is the normal trend, Mox fuel costs will remain stable while uranium fuel costs will inevitably grow.

For the future, all the options remain open: most of them have a proven feasibility. The most advanced solutions (such as Pu burners) are still in progress. They will certainly appear as a very promising objective which leads to an extremely lower level of residual plutonium in the geological disposal.

BASIC EVALUATION ON NUCLEAR CHARACTERISTICS OF BWR HIGH BURNUP MOX FUEL AND CORE

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Abstract

MOX fuel will be used in existing commercial BWR cores as a part of reload fuels with equivalent operability, safety and economy to UO_2 fuel in Japan. The design concept should be compatible with UO_2 fuel design. High burnup UO_2 fuels are being developed and commercialized step by step. The MOX fuel planned to be introduced in around year 2000 will use the same hardware as UO_2 8 x 8 array fuel developed for a second step of UO_2 high burnup fuel. The target discharge exposure of this MOX fuel is about 33 GWd/t. And the loading fraction of MOX fuel is approximately one-third in an equilibrium core. On the other hand, it becomes necessary to minimize a number of MOX fuels and plants utilizing MOX fuel, mainly due to the fuel economy, handling cost and inspection cost in site. For the above reasons, it is needed to develop a high burnup MOX fuel containing much Pu and a core with a large amount of MOX fuels. The purpose of this study is to evaluate basic nuclear fuel and core characteristics of BWR high burnup MOX fuel with batch average exposure of about 39.5 GWd/t using 9 x 9 array fuel. The loading fraction of MOX fuel in the core is within a range of about 50% to 100%. Also the influence of Pu isotopic composition fluctuations and Pu-241 decay upon nuclear characteristics are studied.

1. INTRODUCTION

We have been studying MOX fuel which can be applied to existing commercial BWR cores as a part of reload fuels. We have established three fundamental policies on the MOX fuel design from operability, safety and economy points of view.

(1) to maintain compatibility with UO_2 fuel
(2) to meet same safety design criteria as UO_2 fuel
(3) to adopt same hardware as UO_2 fuel

Accordingly, a design concept of MOX fuel should be compatible with UO_2 fuel design. We describe the current status on our recent BWR UO_2 fuel developments and relationship between UO_2 and MOX fuel design development in section 2.

On the other hand, the needs to decrease a number of MOX fuel bundles and plants introducing MOX fuel will increase more and more in future. Because a amount of MOX fuels strongly affects the costs of fabrication and transportation. Also, an introduction of MOX fuel will need to install new equipment for handling and inspection and safeguard in the plant site. Considering these situations, it is important to develop higher burnup MOX fuel containing more Pu and a core with a larger amount of MOX fuels.

The purpose of this study is to make clear the feasibility of high burnup MOX fuel and core through nuclear characteristics evaluations. The next target exposure of the MOX fuel is about 39.5 GWd/t, and the fuel hardware is 9 x 9 array fuel, which has been developed for a third step of high burnup UO_2 fuel. The loading fraction of MOX fuel in the core is about 50% to

100%. Both the discharge exposure and MOX fuel loading fraction in the core are larger than those of the 8 x 8 MOX fuel and core.

The nuclear design has been performed on 9 x 9 MOX fuel. One design example of the MOX fuel is made, and the core characteristics are evaluated on equilibrium cores in 1100 MWe BWR/5 plant.

Larger Pu inventory in a MOX fuel and larger loading fraction of MOX fuel in a core will increase the effects of Pu isotopic composition fluctuations and Pu-241 decay upon nuclear characteristics. So such influences were also studied.

2. STATUS OF UO_2 AND MOX FUEL DEVELOPMENTS

The status of high burnup UO_2 fuel developments[1] is shown in Table I. These fuels have been developed and commercialized in stepwise manner to improve fuel cycle cost and decrease amounts of spent fuel. They are named high burnup STEP-1, STEP-2, STEP-3 fuel respectively and are applicable in current BWR plants. STEP-1 and STEP-2 fuel with 8 x 8 array have been introduced, while STEP-3 fuel with 9 x 9 array is planned to be introduced after about year 2000.

STEP-2 fuels are mainly used in Japanese BWR plants at present. STEP-2 fuel is designed to achieve a batch average exposure of about 39.5 GWd/t within a maximum assembly exposure of 50 GWd/t. STEP-2 fuel is composed of sixty zirconium liner fuel rods and one large central water rod.

At a first batch-size introduction of MOX fuel to BWR, STEP-2 fuel hardware will be adopted from standpoints of reliability and compatibility with existing UO_2 fuels. We have developed the design of this type of MOX fuel. The target discharge exposure of this MOX fuel

Table I Development Step of High Burnup UO_2 and MOX Fuel

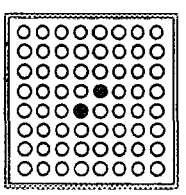
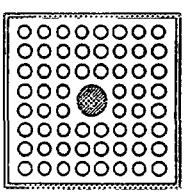
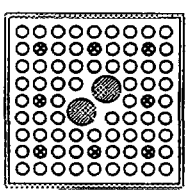
UO_2 fuel	Step	STEP－I	STEP－II	STEP－III
	Batch Average Exposure (GWd/t)	33	39.5	45
	Max Assembly Exposure (GWd/t)	40	50	55
	Bundle Average U^{235} enrichment (w/o)	～3.0	～3.4	～3.7
Fuel Lattice Design		 ・8x8 Array ・2 Water Rods	 ・8x8 Array ・1 Large Water Rod	 ・9x9 Array ・2 Large Water Rods ・8 Part Length Rods
MOX fuel	Step	－	I	II
	Batch Assembly Exposure (GWd/t)	－	33	～39.5
	Max Bundle Average Exposure (GWd/t)	－	40	～50

Table II Summary of Core Characteristics

Item	50% MOX CORE	100% MOX CORE	STEP-3 UO_2 CORE	Operational Limit
Min. Shut Down Margin (% Δ k)	2.0	2.8	3.1	> 1.0% Δ k
Max. Linear Heat Generation Rate (kW/m)	40	39	38	< 44kW/m
Min. Critical Power Ratio	1.45	1.45	1.47	> 1.23
MOX fuel batch average exposure (GWd/t)	39.8	40.2	–	(Target) 39.5GWd/t
MOX fuel max. assembly exposure (GWd/t)	41.7	46.0	–	< 50GWd/t

Table III Summary of Safety Parameters

Item	50% MOX CORE	100% MOX CORE	STEP-3 UO_2 CORE	note
Static Void Coefficient (Relative)	1.08	1.17	1.0 (base)	at EOC, 40% void fraction
Dynamic Void Coefficient (Relative)	1.19	1.44	1.0 (base)	at EOC, 40% void fraction
Doppler Coefficient (Relative)	1.005	1.01	1.0 (base)	at EOC, Cold
Delayed Neutron fraction (%)	0.48	0.43	0.53	at EOC

is about 33 GWd/t, and the loading fraction of the MOX fuels is approximately one-third in an equilibrium core. This core meets all design and safety criteria and w as very similar to that of UO_2 fuel core.

We are interested in the MOX fuel and core design after STEP-3 fuel will have succeeded to STEP-2 fuel. STEP-3 UO_2 fuel is aimed at increasing burnup to about 45 GWd/t as a batch averaged exposure within a maximum assembly exposure of 55 GWd/t. STEP-3 fuel has sixty-six full length rods and eight partial length rods and two large central water rods within a 9 x 9 array. This configuration has been optimized in order to maintain performance at a higher burnup.

We think also a next high burnup MOX fuel will prefer to adopt the STEP-3 fuel hardware in order to have a compatibility with UO_2 fuel and core. So we expect that a development step of high burnup MOX fuel design will be like as shown in Table I . The target exposure of this MOX fuel using STEP-3 hardware is tentatively set to about 39.5 GWd/t under a principal of stepwise manner, which is lower than that of STEP-3 UO_2 fuel.

3. HIGH BURNUP MOX FUEL AND CORE

One design example of MOX fuel was made. The Pu inventory is determined to be able to achieve a target discharge exposure of 39.5 GWd/t. Fuel nuclear calculations were performed

by using TGBLA[2] code, which is BWR fuel lattice physics code. Two MOX cores of different MOX fuel loading fractions were studied on a typical 1,100 MWe (3,293 MWt) BWR/5 plant. Core design and performance calculations were performed by LOGOS[3] code, which is three dimensional BWR core physics simulator. A weight fraction of Pu fissile in Pu used here is 67% as a reference.

3.1 Nuclear Fuel Design Concept

The nuclear design of MOX fuel was made. Pu inventory is considered to be as large as possible. Generally, BWR fuel includes several gadolinia fuel rods (UO_2-Gd_2O_3) in order to suppress an excess reactivity at the beginning of life (BOL). MOX fuel also must have them. But in this study it is not allowed to use PuO_2-Gd_2O_3 rods. Accordingly, increasing gadolinia fuel rods in the bundle leads to fewer Pu inventory. Number of gadolinia fuel rods must be optimized between Pu inventory and allowable reactivity at BOL. The optimized number of gadolinia fuel rods is sixteen in this design.

BWR fuel is composed of several different enrichment fuel rods. An enrichment distribution is established to make a local power distribution flat.

The number of different enrichment types must be reduced as few as possible from a fabrication standpoint under an acceptable local power peaking factor. There are four fuel types of different Pu enrichment in this design. Outer rods are relatively low Pu enrichment and inner rods are high.

A matrix blended with PuO_2 is depleted uranium. Gadolinia is contained in enriched uranium fuel rods.

K-infinity comparison among this MOX fuel, STEP-2 and STEP-3 UO_2 fuel is shown in Fig. 1. Reactivity of MOX fuel is established to be same K-infinity as STEP-2 UO_2 fuel at exposure 25 GWd/t, which corresponds to an average exposure at the end of cycle (EOC) of STEP-2 fuel equilibrium core achieving an average exposure of 39.5 GWd/t.

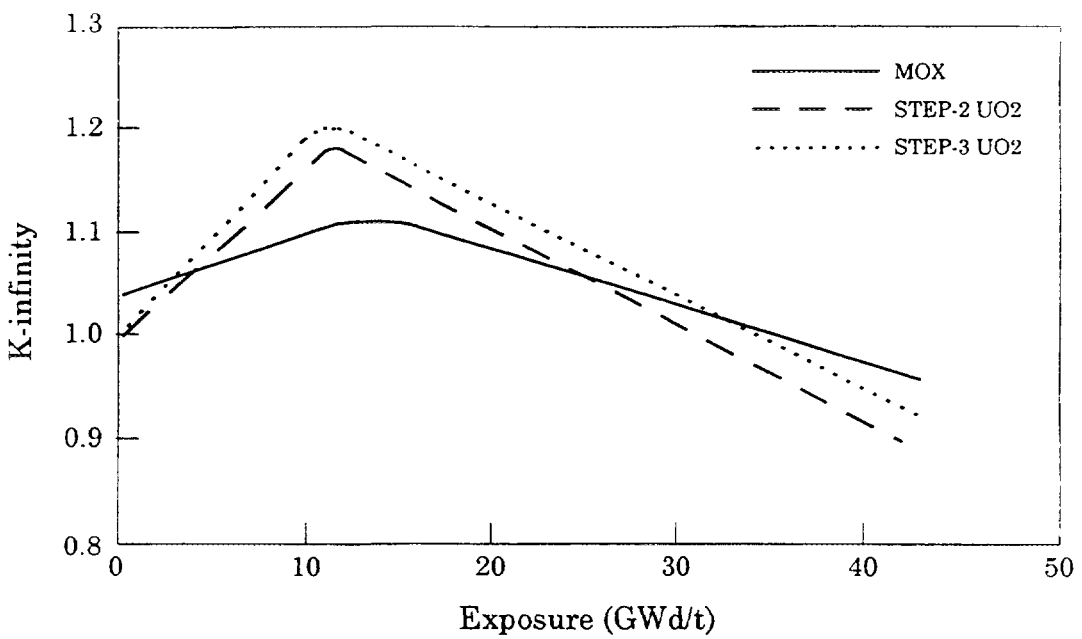
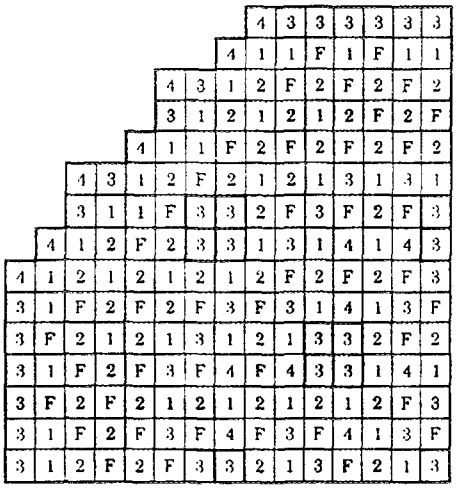
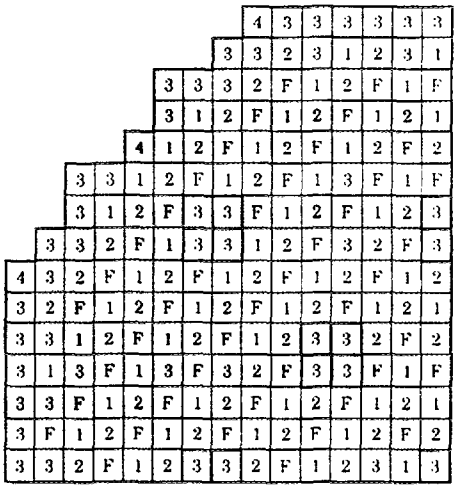


FIG. 1. K-Infinity comparison among MOX, STEP-2 & STEP-3 UO_2 Fuel

(a) 50% MOX CORE



(b) 100% MOX CORE



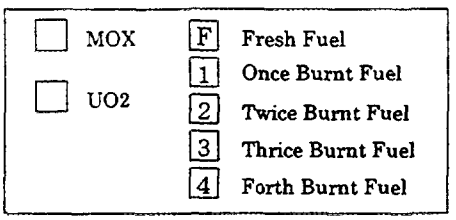


FIG. 2. Fuel Loading Patterns

3.2 Core Design Concept and Core Characteristics

Two MOX cores of different MOX fuel loading fraction were studied. One is the mixed core of the MOX and STEP-3 UO_2 fuel, and the loading fraction of MOX fuel is about 50%. Another is 100% MOX fuels loading core. Both fuel loading patterns are shown in Fig. 2. The cores are designed under the following plant and operating conditions.

- Plant and operating conditions
  - plant type — BWR/5
  - electric power — 1,100 MWe
  - core rated thermal power — 3,293 MW
  - core rated flow — 483,000 t/h
  - number of bundles — 764
  - number of control rods — 185 rods
  - cycle length — 13 months

(a) 50% MOX fuel loading core

The total number of MOX fuels in the equilibrium cycle core is 352 bundles including fresh and burnt fuels. The rest are STEP-3 UO_2 fuels of 412 bundles. The refueling batch size of MOX fuels is 88 bundles per a cycle, then the batch number is 4.0. The refueling batch size of UO_2 fuels is 88 bundles, the batch number is 4.7. Therefore the MOX fuels can achieve substantially the target average discharge exposure of 39.5 GWd/t, and STEP-3 UO_2 fuels can also achieve that of 45 GWd/t. Meanwhile the fuel loading concept are the following, basically same as adopted in current UO_2 core.

(1) to scatter MOX fuels and UO_2 fuels uniformly in the core in order to flatten radial power distribution

(2) to load higher burnt fuels at the most peripheral in the core in order to reduce a neutron leakage

(3) to load higher burnt UO_2 fuels in Control Cell, which is composed of four bundles at the location of control rod insertion during a power operation

(b) 100% MOX fuel loading core

The core is composed of MOX fuels only. The refueling batch size of MOX fuels a cycle is 188 bundles, then the batch number is about 4.1. The MOX fuels can achieve substantially the target average discharge exposure of 39.5 GWd/t under this refueling plan. The core configuration concept is almost same as those of 50% MOX fuel core except that MOX fuels are loaded in Control Cells.

Calculations on core characteristics for both cores were performed at rated power condition. Main results are shown in Fig. 3 and summarized in Table II. All of Shut-down margin at one control rod stuck condition, MLHGR (Maximum Linear Heat Generation Rate), MCPR (Minimum Critical Power Ratio), which are key parameters for BWR core characteristics, meet each operational limit with margins.

Generally, a MOX core tends to reduce the control rod worth, but the cores have enough shut-down margin. Because the K-infinity peak value of MOX fuel is relatively low compared with that of UO_2 fuel, which cancels control rod worth decrease.

Thermal performances such as MLHGR, MCPR of both MOX cores are fairly good. The thermal performance of 100% MOX fuels core is better than that of 50% MOX core and UO_2 core because of smaller power mismatch and neutron spectrum mismatch between bundles.

3.3 Safety Parameters

Moderator void coefficient, Doppler coefficient and dynamic parameters were evaluated to investigate influences upon a transient and accident behavior of a high burnup MOX fuel and core. The results are summarized in Table III.

Generally a static void coefficient of MOX fuel tends to be more negative compared with that of UO_2 fuel. Consequently increasing a loading fraction of MOX fuel in a core leads to larger core average void coefficient. Dynamic void coefficient taking delayed neutron fraction into consideration is important from transient behavior standpoints. An increase of dynamic void coefficient of MOX core will be larger than that of static void coefficient because of smaller delayed neutron fraction of MOX fuel.

Generally, a transient behavior such as Turbine Trip event will be severe because of larger void coefficient. But, since current BWR has fast speed SCRAM mechanism, void coefficient increase does not affect a operational limit.

Doppler coefficient of MOX fuel is slightly more negative than that of UO_2 fuel. This will moderate a behavior of reactivity initiated accident such as control rod drop accident. A power excursion is suppressed by large Doppler effect, and a fuel enthalpy increase will be in low.

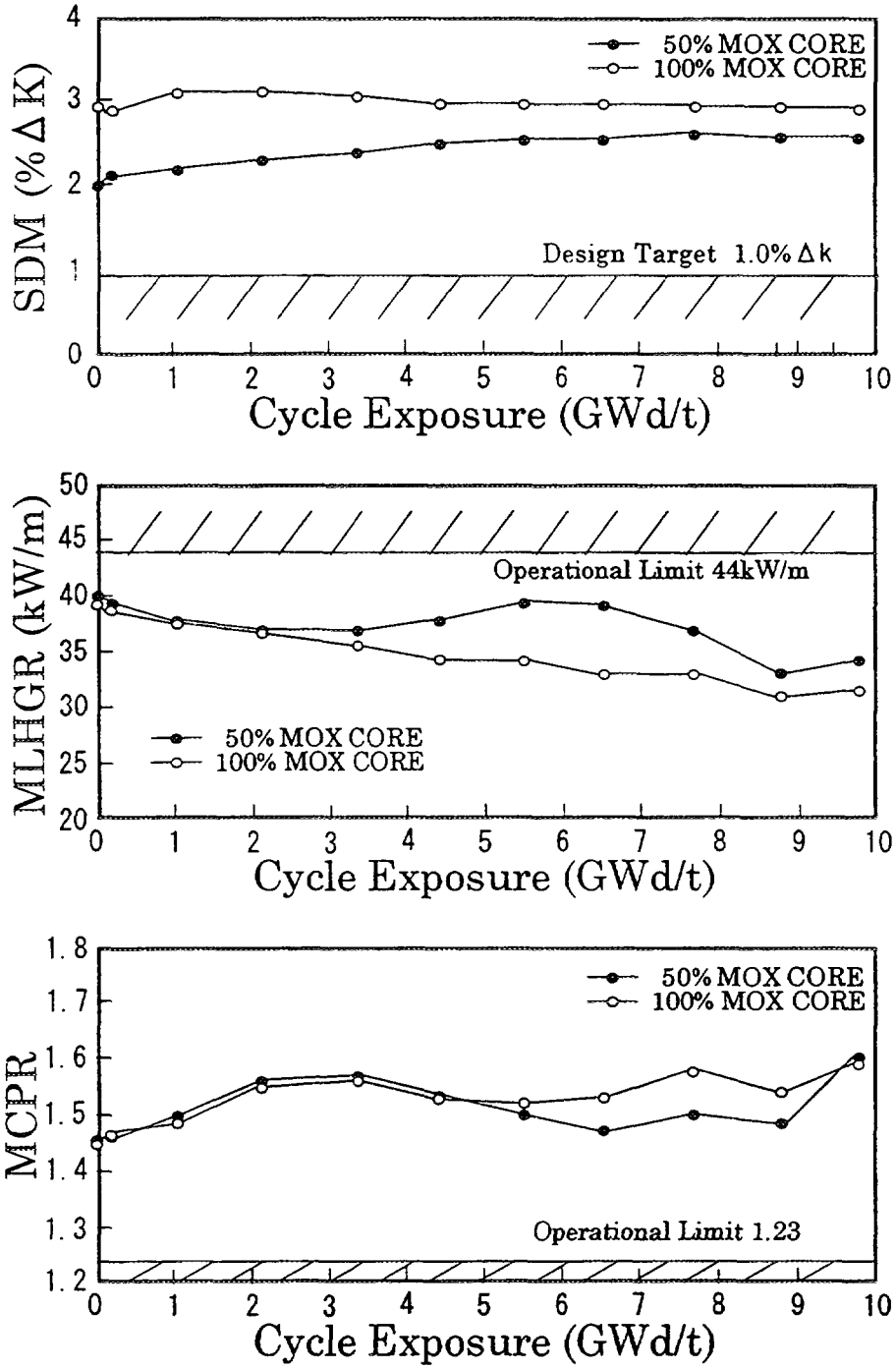


FIG. 3. Core Characteristics

Prompt neutron life time of MOX core is smaller than that of UO_2 core. But it is known that a prompt neutron life time dose not practically affect a transient and accident behavior.

3.4 MOX Fuel and Core Specific Considerations

(a) Influences of Pu isotopic composition

It is expected that an increase of Pu inventory in fuel and core has more influences upon a core characteristics due to a fluctuation of Pu isotopic composition ('Pu vector'). Two Typical MOX fuels with different Pu vector from the reference Pu vector used here were studied. One

is higher quality Pu with large fraction of Pu fissile than the reference. Another is lower quality than the reference. Pu contents of these MOX fuels are adjusted to have equivalent reactivity to the reference MOX fuel. Figure 4 shows a relationship between Pu vector and a required Pu inventory or void coefficient of 100% MOX core. This result shows that void coefficients among three kinds of MOX fuel are almost same. It is found that a influence of Pu vector fluctuation is eliminated by adjusting Pu content to achieve equivalent reactivity according to the Pu vector.

(b) Influences of Pu-241 decay

It is also expected that an increase of Pu inventory have more influence upon a core characteristics due to a decay of Pu-241 with half life of 14.4 year, which decays to Am-241. Influence of Pu-241 decay upon core reactivity was studied on 100% MOX core. Figure 5 shows

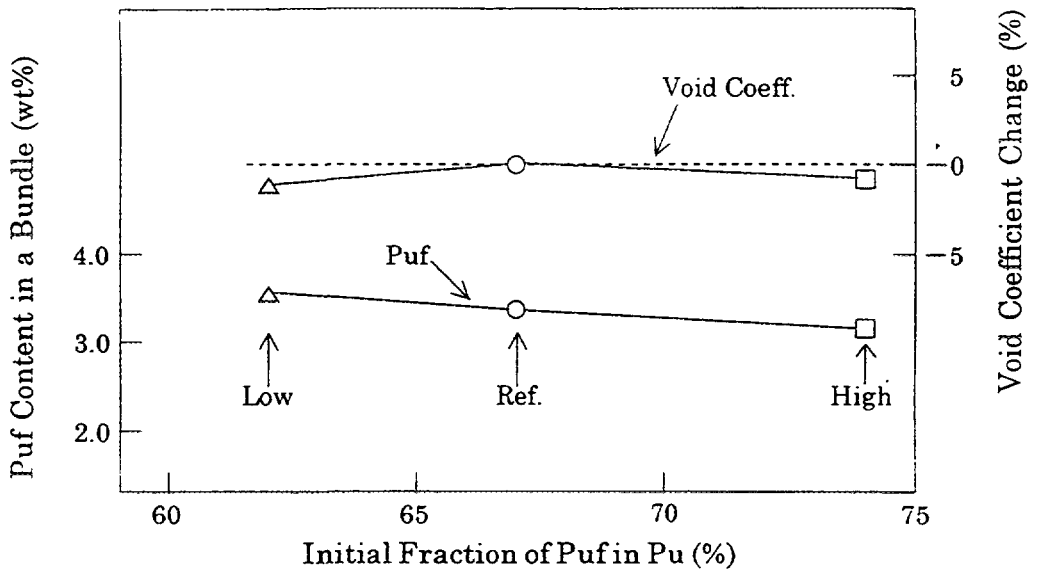


FIG. 4. Relationship between initial fraction of Puf and Pu content, void coefficient

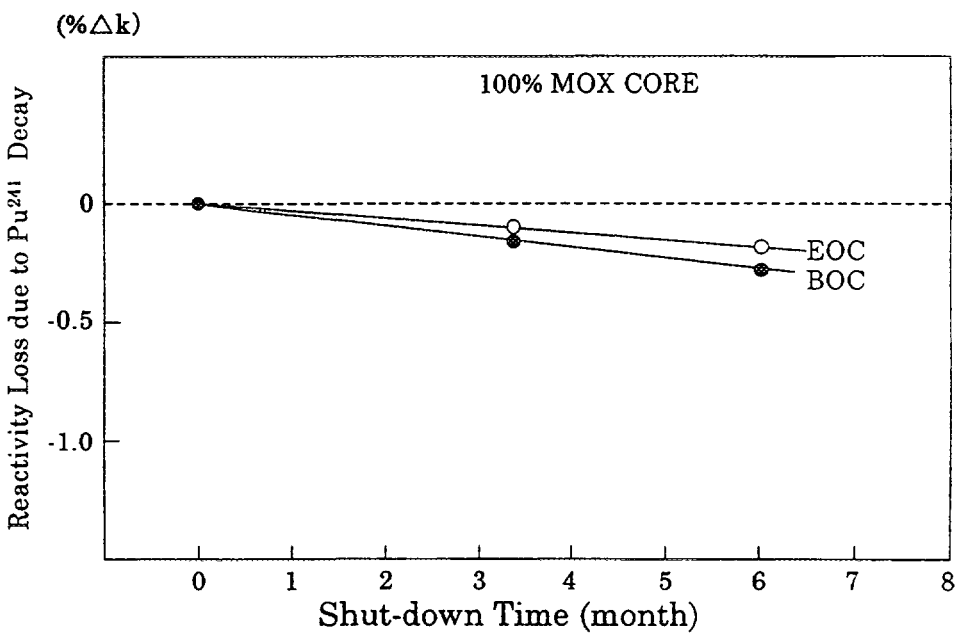


FIG. 5. Relationship between shut-down time during periodic inspection and reactivity loss at BOC and EOC due to Pu^{241} decay

a relationship between a shut-down time during a periodic inspection and reactivity loss at BOC and EOC. A reactivity loss due to a reduction of Pu-241 and a production of Am-241 during a shut-down is proportional to its length and is not negligible in magnitude.

While a reactivity loss at EOC is relatively small compared with that of BOC because of a consumption of Am-241 through burnup, so the effect to a cycle length is not so large in a case of short shut-down time, but it is not negligible in a case of longer shut-down time.

It must be taken account of a Pu-241 decay effect exactly in a nuclear characteristics evaluation for MOX fuel and core.

The above LOGOS code can treat the effect of Pu-241 decay on reactivity during operation and shut-down.

4. CONCLUSION

High burnup MOX fuel and core with a large amount of MOX fuels were studied. The following were found through nuclear characteristics evaluations.

(1) 100% MOX fuels loading core with a batch average exposure of 39.5 GWd/t is feasible from core characteristics standpoint in a typical 1,100 MWe BWR/5 plant.
(2) 100% MOX core has better core characteristics than the mixed core of MOX and UO_2 fuels because of smaller power mismatch and neutron spectrum mismatch between bundles.
(3) Increasing MOX fuel in a core leads to more negative void coefficient. This generally tends to cause severe on a transient behavior, but this doesn't affect a transient of current BWR with fast speed SCRAM mechanism.
(4) Pu inventory within a bundle is limited by the existence of gadolinia fuel rods in BWR MOX fuel. In order to in.crease Pu inventory further, it will be necessary to permit to use a mixed fuel rod of gadolinia and Pu.
(5) The effect of Pu vector and decay of Pu-241 should be treated for the MOX fuel and core especially with much Pu inventory.
(6) The influence of Pu vector upon a nuclear characteristics is eliminated by adjusting Pu content to have equivalent reactivity according to the Pu vector.
(7) The effect of Pu-241 decay during shut-down under periodic inspection must be considered in nuclear characteristics evaluation to predict a exact core reactivity and performance.

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PLUTONIUM RECYCLING: THE POINT OF VIEW OF ELECTRICITE DE FRANCE

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Abstract

EDF has more than 25 reactor years of operating experience with MOX fuel. This paper presents a synthesis of the available experience feedback relating to MOX fuel management and considers the various different aspects - operations, calculations, tests and measurements.

1. ANALYSIS OF EXPERIENCE FEEDBACK FROM MANAGEMENT OF MOX FUEL

1.1. Introduction

EDF now has more than 25 reactor years of operating experience with MOX fuel, involving 7 reactors.

In view of the prospects for development of fuel management towards hybrid MOX management, and the extension of plutonium recycling to a greater number of power plant units, it was considered important to assess the available experience feedback relating to the original three-batch fuel cycle.

The analysis was carried out on the basis of three areas :operations, studies and tests. Before examining these three aspects of experience feedback, the main characteristics of the scope of the feedback will be considered.

1.2. General experience

MOX fuel was first loaded into a reactor at St Laurent B1 in 1987. The experience feedback from the management of MOX fuel involves more than 25 completed, or almost completed, irradiation cycles in 7 reactors : Saint Laurent B1/B2, Gravelines 3/4, Dampierre 1, Dampierre 2 (first MOX loading : late 1993) and Blayais 2 (1994). These two last reactors were loaded with hydrid MOX fuel management (1/3 core for MOX fuel, 1/4 core for uranium fuel).

An increasing number of reactors is set to be opened up to MOX loading as the MELOX plant comes progressively on stream through 1996/97. The hybrid MOX fuel management will be generalized.

Table 1 shows the history of these irradiation cycles and the number of MOX fuel assemblies loaded at the beginning of 1995. More than 400 such assemblies have been irradiated, almost half of which were unloaded after 3 irradiation cycles.

Figure 1 shows the distribution of the discharge burnups of MOX assemblies. After 3 cycles, the average is 37.5 GWd/t, with a maximum of 40 GWd/t.

Four MOX assemblies, reloaded for a fourth cycle on the periphery of the core in Gravelines 4, reached 43 or 44 GWd/t. They were unloaded in March 1994.

DISCHARGE BURNUPS

Mean BU : 37613

As at 13/02/1995 (204 MOX assemblies)

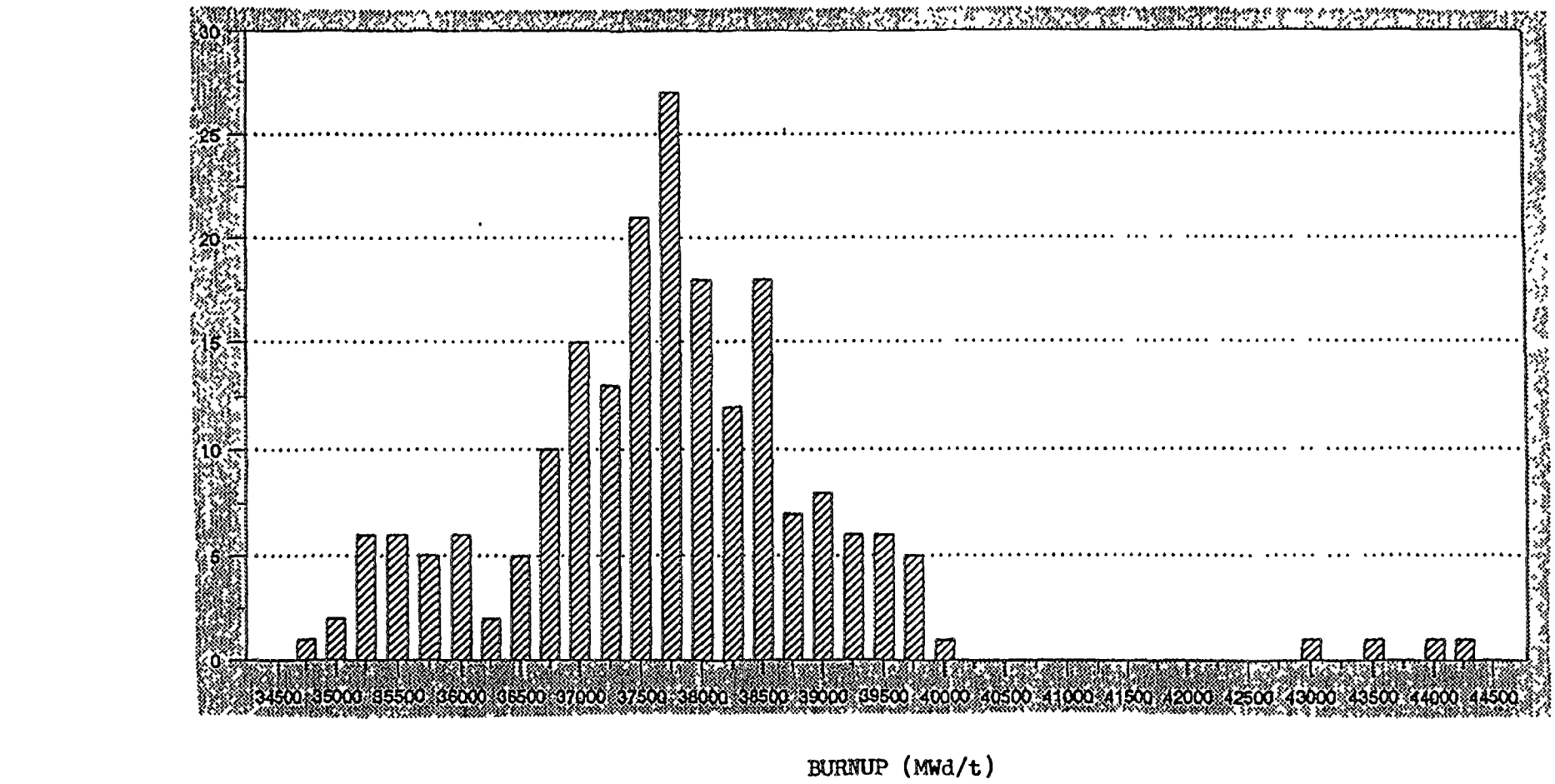


FIG. 1. Discharge Burnups (As at 13/02/1995 (204 MOX assemblies)

1.3. Operating experience feedback

1.3.1 Stretch-out operation

Stretch-out operation is necessary to optimize the timing of unit shutdowns. Studies have limited their duration to 40 equivalent full power days (EFPD) in MOX management, except during the first transitional cycle (60 EFPD, as for UO2 management). Figure 2 shows that this flexibility has been fully exploited in both MOX of UO2 management.

The limit of 40 EFPD has been extended to 60 EFPD in hybrid MOX management, as the IPG phenomenon which was behind it no longer appears to be a limiting factor with MOX.

1.3.2 Network-following/fine control operation

Only the two units St Laurent B1 and B2 have had approval for network following operation, which has been renewed for each irradiation cycle from 1990 to 1994. At the request of the Central Department for Radiological Protection, the generic approval was made conditional on the acquisition of sufficient experience feedback on these two units. The renewal of the approval, after conversion of these two units to hybrid MOX fuel management in 1994, is being discussed with the Safety Authority.

The analysis of operating experience feedback carried out in summer 1993 did not demonstrate any great difficulties in fine control. The analysis relates to 3.5 reactor years, 5 800 hours of remote load dispatch control and 87 load following operations. Favorable effects, in terms of a reduction in effluent volume, were observed in network following, due to the smaller proportion of xenon.

The boron concentrations in the various standard shutdown states are higher than with UO2 management, which may increase dilution times on restart.

Operation and fine control do not bring out any new problems in current recycling rate hypotheses.

1.3.3 Behavior of MOX fuel

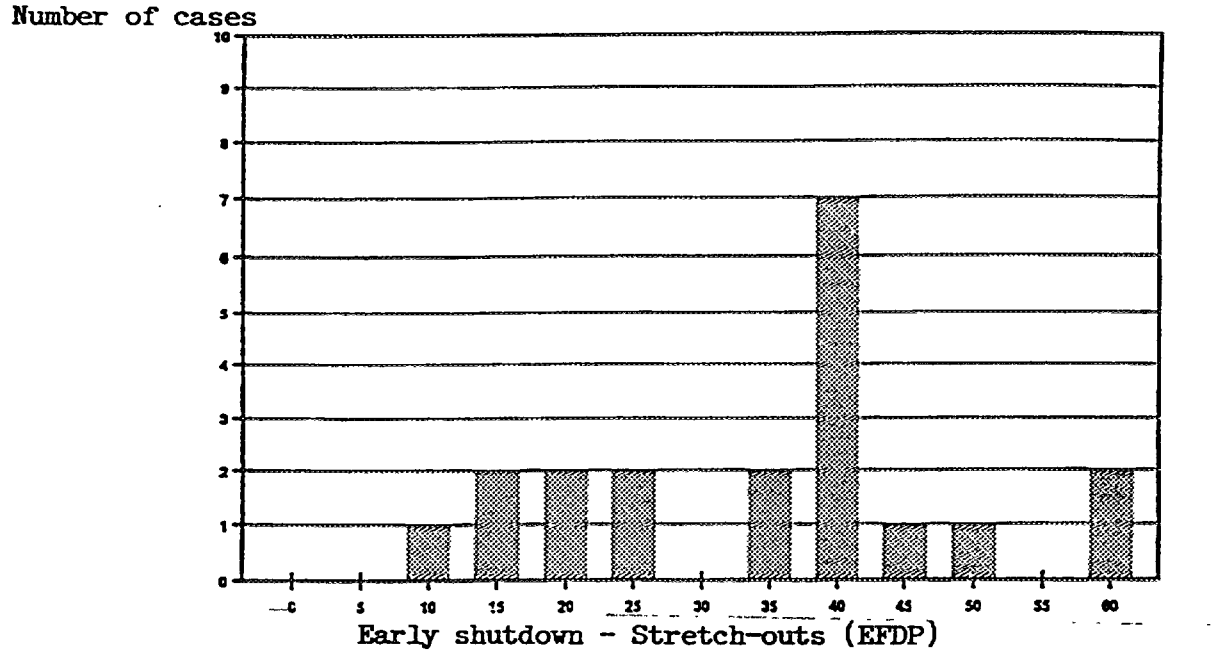
The behavior of MOX fuel assemblies has been completely satisfactory. The first failure, in fact, which was due to a loose part, was identified at the end of cycle 11 at Dampierre 1 in July 93. As this cladding failure met the reload criteria, the assembly was reloaded in the following cycle, and was subject to specific monitoring (radiochemical monitoring by the operator, and on-line monitoring of primary circuit activity by the CEA). It has been unload and will be repaired (replacement of leaked fuel roads) before reloading for a third cycle (cf. communication D. PARRAT "Behaviour of a defective MOX fuel rod in PWR).

1.4 Experience feedback from reload studies

Experience feedback from loading pattern studies can be illustraded using the results obtained for the key safety parameters, the impact of fluctuation of the isotopic composition of the plutonium and the impact of plutonium recycling on reactor vessel fluence.

1.4.1 Isotopic vector

The isotopic compositon of MOX fuel batches varies as a function of plutonium origin. Fluctuations in average total plutonium content of between 4.5 % and 5.3 %, équivalent to a uranium enrichment of 3.25 %, have been observed. A deterioration in plutonium quality, coupled with aging, has led to the average content leveling off at 5.3 %.



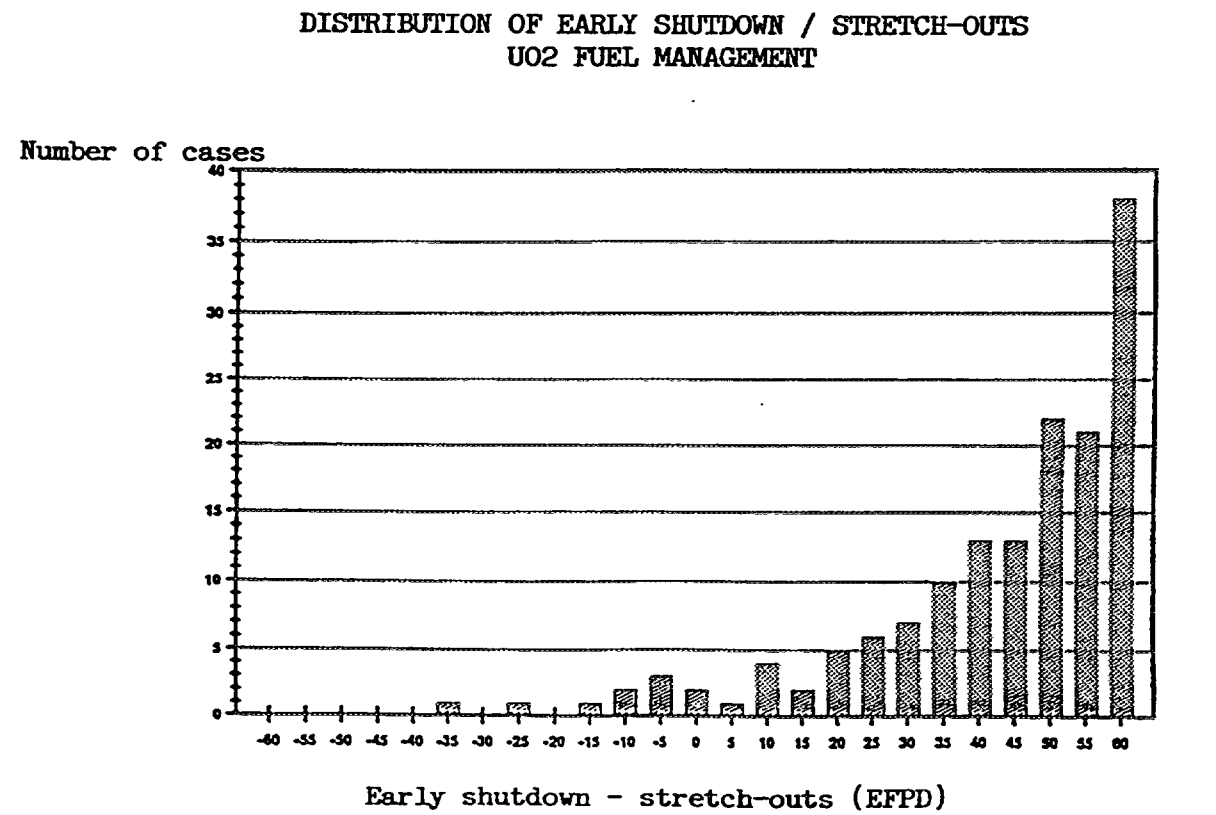


FIG. 2. *Distribution of early shutdown/stretch-outs MOX Fuel Management and UO_2 Fuel Management*

1.4.2 Reload safety analysis

Figure 3 shows a loading pattern which is typical of the equilibrium fuel cycle, i.e. with 3 batches of 16 MOX assemblies. The new MOX assemblies are loaded in the "entering" positions around the periphery, while irradiated MOX assemblies are reloaded towards the center of the core. MOX fuel supply problems can easily be countered by using UO2 3.25 % fuel as a replacement.

The different key parameters of the safety assessment files (radial hot spot factors reactivity, shutdown margin, modered coefficient) for each reload do not show any specific problems.

1.4.3 Influence on reactor vessel fluence

Loading of new MOX fuel at the periphery of the core instead of UO2 fuel would increase the flux. Current loading patterns place the new MOX fuel in a position which ensures that the hot point of the flux on the reactor vessel on the main core axes is not affected.

Loading of irradiated MOX fuel around the periphery of the core is not ruled out. The favorable effect of power decrease prevails over the increase in rapid flux. The 4 MOX assemblies reloaded for a 4 th cycle were in fact loaded around the periphery as the axis end, leading to a decrease in rapid flux at the hot spot.

1.5 Experience feedback from periodic test and measurements

Up to now, no special difficulty has been encountered in using MOX fuel. The safety assessment computations were validated by core physic tests at start up. Test campaigns have been carried out at various levels of power. The differences between measured and calculated parameters are similar to those obtained in the uranium cores (critical boron concentration, control rod worth, temperature factor, etc.). All the design and safety criteria are confirmed and, concerning the control rod banks worth, a very good agreement was noted between calculations and measurements.

All these results show the core conformity with design, confirm the accuracy of EDF calculation codes and methods, and validate the surveillance and protection systems.

1.5.1 Start up test at zero power

These tests include standard start up tests and specific ones for first MOX cycles. Three types of tests are performed :

- measurements of critical boron concentration (all rods out, control rod banks inserted, grey bank inserted),
- isothermal coefficients under the same three above described conditions,
- worth of control rod bank (using dilution measurement method) and other control rod bank (using a control rod bank exchange measurement method).

The main results show that all the differences meet design criteria :

- the difference between calculated and measured critical boron concentration is less than 50 ppm,
- concerning isothermal coefficients, less than 5,4 pcm/°c were found between calculation and measurement,
- control rods bank worth was better estimated than 10%.

1.5.2 Flux map and critical boron concentration at full power

According to the standard "out-in" core management strategy, the fresh fuel is loaded in the periphery of the core. Nevertheless, fresh MOX assemblies are not loaded close to the baffle in order to limit their contribution to the vessel fluence. During the following cycles, irradiated MOX fuel is moved towards the centre of the core.

According to flux maps established in the core at full power, the difference between measurements and calculations is in the same range as the one found with enriched uranium cores.

Moreover, flux maps showed that required criteria are satisfied for power peaking factor, hot channel factor and radial power peaking factor.

The agreement between calculations and measurements of the critical boron concentration were found excellent and not different from standard uranium cycles.

1.5.3 Primary coolant activity

The primary coolant activity is quite identical to that of the reactors burning uranium fuel (only one rod failure has occurred up to now).

1.6 Conclusion

This synthesis of the available experience feedback relating to MOX fuel management allows the various different aspects - operations, calculations, tests and measurements - to be considered.

The use of MOX fuel as standard should provide a justification for a relaxation in the areas of adminsitrative procedures and monitoring provisions.

The experience feedback analyzed showed that, in global termes, MOX cores display behavior which is equivalent to that of UO2 cores in terms of the various operating and safety parameters.

Experience feedback from the first cores operated with hybrid MOX management will further supplement this analysis.

2. THE PRESENT STRATEGY

2.1 Origins of the present strategy

At the beginning of the 80's, it became clear that, following the construction of Super-Phénix, the development of further breeders would be delayed.

Within this new environment, reprocessing was put into question. But, taking into account :

- the feasibility of recycling plutonium in PWR reactors which had been demonstrated through foreign and French programs and experimentations,
- the demonstration which had been made that, through reprocessing and recycling, a consistent back end solution for the fuel cycle was available,
- the economics of recycling considering investments already made.

EDF decided in 1985 to proceed with reprocessing and, as a consequence, to recycle plutonium in its PWRs on a large scale, in the form of MOX (mixed uranium and plutonium oxides) fuel.

EDF's present strategy is to adapt the reprocessing programme to the fuel industry's potential for MOX fabrication. The aim of the strategy is to avoid the creation of a large plutonium stock : no more plutonium should be separated per year than could be incorporated into MOX fuel. This strategy is currently called **"adjustment of reprocessing flows"**.
This strategy implies a future increase of the number of EDF's PWRs burning MOX fuel. Therefore, EDF's target is to make MOX fuel a **"common"** product, like uranium fuel.

2.2 EDF's program

By the end of the century, 16 to 28 900 MWe PWRs may be MOX fuelled, depending on the available manufacturing capacity and authorised burn-up and core management strategy.

If EDF uses more than 16 reactors, it will be necessary to carry out some public enquiry for the supplementary ones.

Experiments and studies are underway to improve current knowledge of MOX fuel behaviour in the reactors for satisfactory implementation of plans to increase burn-ups :

- post-irradiation examination on irradiated materials (cf. communication P. BLANPAIN "Plutonium recycling in French power plants : MOX fuel irradiation experience and behaviour").
- neutron physics critical experiments,
- theoretical studies of different management schemes (1/4 core, increased rate of MOX fuel per reactor, ...).

All these actions are related to the studies precising the type of reactors EDF will use when renewing its PWRs in the years 2010.

2.3 Multiple recycling for the future

As used MOX fuel assemblies contain more plutonium (20 kg Pu) than uranium used assemblies (4 kg Pu), one could imagine it would be more beneficial to give them priority in reprocessing.

Nevertheless, if used MOX fuel reprocessing is technically possible (such experiment have already been performed in La Hague plant), second generation plutonium produced from such reprocessing is rich in even isotopes which make it less energetic in a PWR than first generation plutonium. Moreover, as MOX matrix is made with depleted uranium, reprocessed uranium (REPU) separated from MOX assemblies cannot be recycled in PWRs.

For these reasons, EDF does not intend to reprocess MOX fuel assemblies in the near futur.

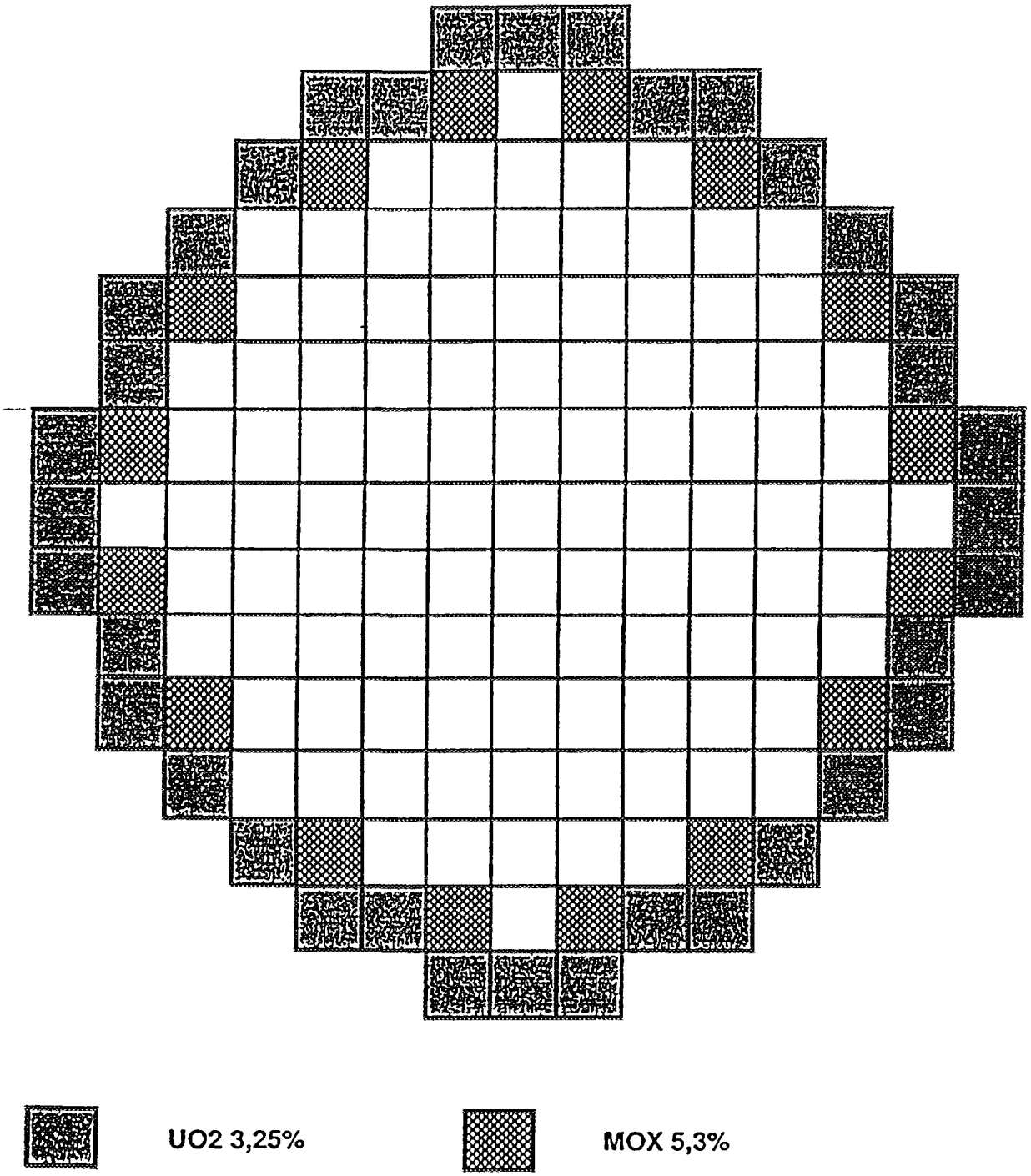


FIG. 3. *Three-batch Cycle UO_2 3.25% + MOX 5.3% Standard loading pattern*

TABLE 1

OPERATING EXPERIENCE FEEDBACK WITH MOX

MOX units	1987	1988	1989	1990	1991	1992	1993	Total Cycles	Total Mox assemb.	Program result 1994
SLB1	C5 16 MOX	C6 16 MOX		C7 16 MOX	C8 16 MOX	C9 16 MOX	C10 16 MOX	6	96	(C11) 16 MOX
SLB2		C6 16 MOX	C7 16 MOX		C8 16 MOX	C9 16 MOX	C10 /	5	64	(C11) 16 MOX
GR3			C8 16 MOX	C9 16 MOX	C10 16 MOX	C11 16 MOX	C12 8 MOX	5	72	(C13) 16 MOX
GR4			C8 16 MOX	C9 8 MOX	C10 16 MOX	C11 16 MOX		4	56	(C12) 16 MOX
DA1				C9 16 MOX	C10 /	C11 16 MOX	C12 /	4	32	(C13) /
DA2							C12 16 MOX	1	16	(C13) 16 MOX
BL2										(C12) 8 MOX
Total Cycles	1	2	3	4	5	5	5	25	/	7
Total MOX ass	16	32	48	56	64	80	40	/	336	96

TABLE 2

COMPARISONS BETWEEN MEASURED AND CALCULATED VALUES
OF CORE PARAMETERS
FUEL MANAGEMENT UO2 AND MOX

CORE PARAMETERS		CB ARO C - M (ppm)	αISO ARO C - M (pcm/°C)	CONTROL ROD WORTH (C - M)/M (%)								
				R	G1	G2	N1	N2	SA/SD	SB	SC	GG
MANAGEMENT MOX	MEAN	7.7	- 0.4	0.4	2.2	2.1	1.7	7.1	0.1	4.4	1.8	0.6
N = 22	STANDARD DEVIATION	17.7	1.0	2.2	4.3	2.1	4.1	6.3	3.9	5.7	3.2	4.4
MANAGEMENT UO2 (3 CYCLES 3.25 %)	MEAN	12.8	0.2	0.8	4.2	- 0.1	1.0	1.8	3.8	1.2	0.6	- 1.9
N > 22	STANDARD DEVIATION	19.6	1.0	3.1	4.2	2.6	4.1	7.2	6.7	5.7	3.7	3.1
MANAGEMENT UO2 (4 CYCLES 3.7 %)	MEAN	- 4.4	- 0.4	- 0.2	2.7	1.5	-1.5	4.5	1.8	3.7	2.2	- 0.3
N = 46	STANDARD DEVIATION	19.4	0.8	2.9	5.2	3.1	4.4	6.8	5.1	4.9	4.4	5.8

- STANDARD - DEVIATION
- N = NUMBER OF CYCLES
- GG : GRAY GROUPS OVERLAPPING

2.4 Conclusion

EDF, by its position, belongs to the small circle of utilities who keep a long term vision (as far as nuclear fuel is concerned, this latter represents several decades).

The reprocessing - recycling technology has attained maturity in Europe, from plutonium separation to irradiated MOX fuel reprocessing. Used fuel reprocessing and fissile material (uranium and plutonium whose energetic value is unquestionable) recycling is the fundamental option of the back end of the nuclear cycle in France.

However, this choice, though it is quite favoured by EDF, can be modified at any time. EDF is still optimistic for the future but keeps open the other options, such as direct storage without reprocessing. EDF could be ready to alter its policy if it was induced by R&D programmes or changes in raw material prices.

The results of this strategy have already proved positive :

- safety has been proved,
- technology has been mastered,
- economic equilibrium between MOX and uranium assembly has been obtained,
- international checks concerning fissile materials are efficient and thoroughly performed.

EDF is preparing for the future, taking plutonium recycling into account when designing the power plants of the next century.

Taking these results into consideration, French partners of nuclear industry rely upon the future of this young fuel, the MOX, and more generally are confident in recycling of nuclear materials. The accumulated experience for these recent years should be profitable for other utilities.

MIXED OXIDE FUEL DEVELOPMENT FOR THE INDIAN NUCLEAR POWER PROGRAMME

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Abstract

The Indian Nuclear Power Programme at present is based mainly on Pressurised Heavy Water Reactors (PHWRs) with the exception of two Boiling Water Reactors (BWRs) in operation since 1969. The former use Zircaloy-clad natural uranium oxide (Nat.UO_2) fuel and the latter low enriched uranium oxide (LEU) fuel elements. India has adopted the philosophy of closed fuel cycle and recycling of plutonium (Pu) essentially in the fast reactor programme. However, due to slower progress of fast reactor programme, utilization of plutonium as mixed oxide (MOX) fuel in a recycle mode in the thermal reactors is contemplated as an interim measure. Studies have been carried out for the introduction of MOX in the PHWR cores using identical Zircaloy hardware as for natural uranium core, consisting of 19-rod fuel bundles. Based on design calculations, substitution of the inner 7 rods by MOX and retaining the outer 12 as natural UO_2 rods appears encouraging and work is being taken up on fabrication of these fuel bundles. Further, based on the studies carried out for recycling of plutonium in BWRs, it has been decided to operate the Tarpur reactors with a mixed core consisting of LEU and MOX fuel assemblies. A few MOX fuel assemblies have been loaded into one of the reactors of this station. Earlier, a number of irradiation experiments had been carried out on short-length MOX fuel rod clusters in the Pressurised Water Loop (PWL) of the research reactor CIRUS at the Bhabha Atomic Research Centre, (BARC) Trombay, as part of the MOX fuel development programme. Some of the fuel characteristics for extended burn-up such as higher grain size, controlled porosity, etc. had been under study. These studies enabled working out an appropriate flow-sheet and establishing the requirements of process machinery and equipment for MOX fuel manufacturing plant. The standard process flow-sheet for ceramic fuel fabrication by pelletising route has been adopted for manufacturing, with selection of some of the process steps to suit glove-box operations. These include use of an attritor for mechanical milling of powders, addition of a synthetic binder, use of diamond wheel for pellet grinding and hydroclone for sludge separation, etc. The quality control steps include a gamma auto-radiography technique which enables the detection of micro-inhomogeneities on the pellet surfaces. The configuration of the fuel assembly is 6 x 6 all-plutonium rods, based on a design optimising 3 enrichments to match the existing LEU fuel bundle characteristics. Subject to satisfactory performance, plans are in hand for increasing the MOX fuel loading progressively in the two BWRs of the Tarapur Atomic Power Station (TAPS) thereby putting to use the plutonium contained in the spent fuel of TAPS.

1. INTRODUCTION

Plutonium recycling in thermal or fast reactors is an important nuclear power strategy and is essential for better utilisation of nuclear energy resources. It is well known that plutonium can be most efficiently used in fast reactors and hence our priority has always been to develop advanced plutonium bearing fuels for fast reactors as evidenced by the mixed carbide core of the 40 MW (th) Fast Breeder Test Reactor (FBTR) at the Indira Gandhi Centre for Atomic Research, Kalpakkam. This 70% PuC-UC fuel, developed and manufactured in the early eighties at our Radio-Metallurgy Division (RMD), has been used in the first core of the FBTR, which is a fore-runner of our fast breeder programme and test-bed for the development of advanced fuels/materials needed for the same. However, due to slower progress of the fast reactor programme, plutonium recycle in thermal reactors has been considered as an interim measure.

Indian nuclear power programme at present is based mainly on Pressurised Heavy Water Reactors (PHWRs) with the exception of two Boiling Water Reactors (BWRs) at the Tarapur Atomic Power Station

(TAPS), in operation since 1969. In this paper, some important aspects are brought out regarding the introduction of MOX fuel in these two types of reactors, the development of the fuel, manufacture and quality control of MOX fuel assemblies for the commercial BWRs.

2. MOX FUEL FOR PHWRS

The mainstay of Indian nuclear power programme is Pressurised Heavy Water Reactor (PHWR), which is characterised by very attractive features such as -- i) high utilisation of U due to excellent neutron economy; ii) use of natural uranium (NU),iii) on-load refuelling leading to high reactor availability and capacity factor, etc. Nevertheless the natural U fuel in PHWRs has a low burn-up capability of about 7000 MWd/T. This implies that PHWRs need larger fuel cycle industry (except enrichment) both in the front-end (fuel fabrication) and at the back-end (spent fuel storage, transport, reprocessing and waste management) per unit of electricity generated compared to Light Water Reactors (LWRs). Extension of fuel burn-up will be possible if MOX fuel is used and this results in reduction of capacity requirement of both front and back-end services. Introduction of MOX fuel will also facilitate once-through thorium cycle in PHWRs, enabling exploitation of the large thorium resources available in India.

A study "Plutonium Recycling in RAPS" [1] has been carried out at the BARC for back-fitting the PHWR core with MOX fuel using the standard zircaloy hardware used in the 19 element NU fuel bundle. The MOX fuel bundle consists of 12 nat. UO_2 rods in the outer periphery and 7 MOX fuel rods in the centre with a composition of 0.4% PuO_2 in nat. UO_2 (Fig.1). It has been calculated that in equilibrium condition, the PHWR core can have 44 central channels with Nat. UO_2 and the remaining 262 channels with MOX. The average burn-up of the core in this scheme is about 10,700 MWd/T. Some of the features of this fuel design are given in Table-I. Initial studies carried out in respect of thermal hydraulics and reactor physics indicate that this pu recycling scheme is feasible in our PHWRs. It is planned to commence the fabrication of such MOX fuel bundles and progressively introduce them in the PHWRs.

3. MOX FUEL FOR BWRS

Introduction of MOX fuel in the BWRs in several countries started after extensive experimental irradiations in the research reactors followed by power reactors.

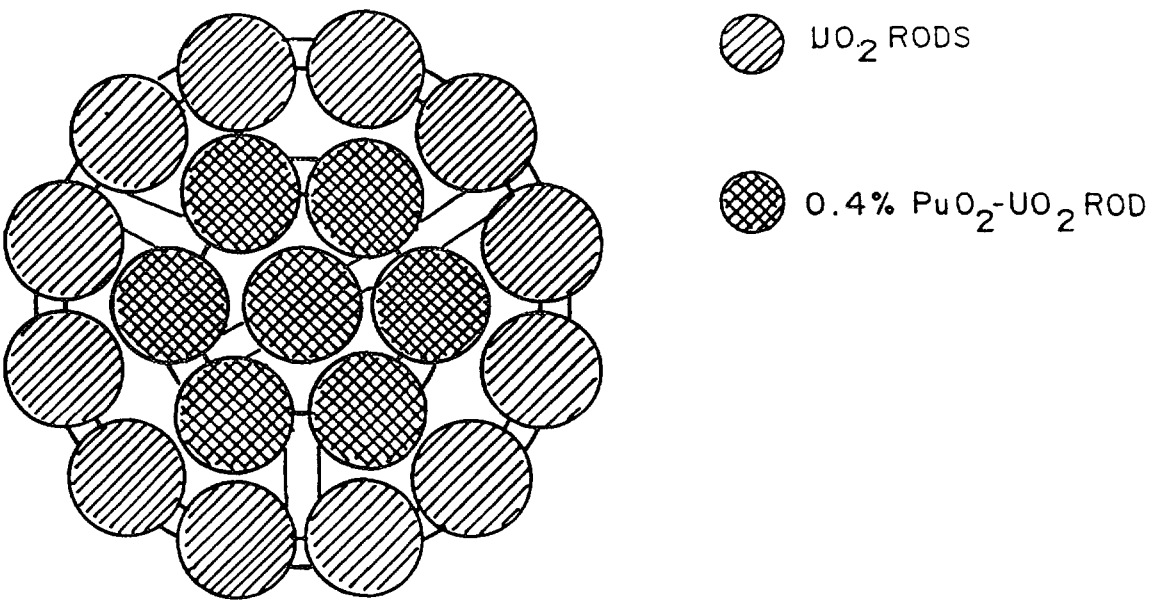

FIG. 1. MOX Bundle for PHWR

TABLE I

SOME FEATURES OF MOX-7 FUEL BUNDLE FOR PHWR

1. FUEL BUNDLE DESIGN	:	MOX-7 :	Inner 7 rods with 0.4% PuO_2 Outer 12 rods with Nat.UO_2	
2. BUNDLE POWER DISTRIBUTION [From Center to Periphery]				
a) Start of irradiation	:	1.032	1.175	0.910
b) 10 GWd/Te burn-up	:	0.910	0.969	1.023
3. FUEL/BURN-UP (MWd/Te)				
a) Inner Zone	:	Nat. U/11,000		
b) Outer Zone	:	MOX-7/10,200		
c) Average	:	10,700		
4. MAXIMUM BUNDLE POWER (kW)	:	422 in Nat. U bundle		
5. MAXIMUM CHANNEL POWER	:	3.11 MW		
6. REACTIVITY WORTH OF ADJUSTER RODS	:	13.2 milli-K		

In the early '70s, it was felt that the optimum design of MOX fuel for PWRs would be of `All-Pu' assemblies, and that for BWRs would be `Island'assemblies (containing LEU rods and MOX rods), to minimise control rod worth reduction. However, it has now been possible to use `All Pu' assemblies even in BWRs.

This approach has many logistic and back-end fuel cycle advantages. The number of MOX assemblies loaded in the core are at present restricted to about 30% to meet the operational safety requirements.

In India, the two BWRs of TAPS operating at 160 MW(e) each are designed to be fuelled with LEU. In order to utilise the nuclear fuel resources to the maximum extent possible, operation of TAPS with a mixed core consisting of MOX and LEU assemblies has been contemplated. Fig.2 gives the typical design of the "All Pu" MOX assembly worked out for TAPS and at present a few such assemblies have been loaded into one of the TAPS reactors.

Studies were initiated to establish the feasibility of a recycle scheme, addressing several significant issues, some of which are listed in Table-II. Some of the MOX fuel assembly characteristics arrived at based on the study[2] are given in Table-III.

4. DEVELOPMENT AND MANUFACTURE OF MOX FUEL

Since thebeginning of the '80s, a number of MOX rods have been irradiated in pressurised water loop (PWL) of the research reactor CIRUS as part of programme for the development and testing of MOX

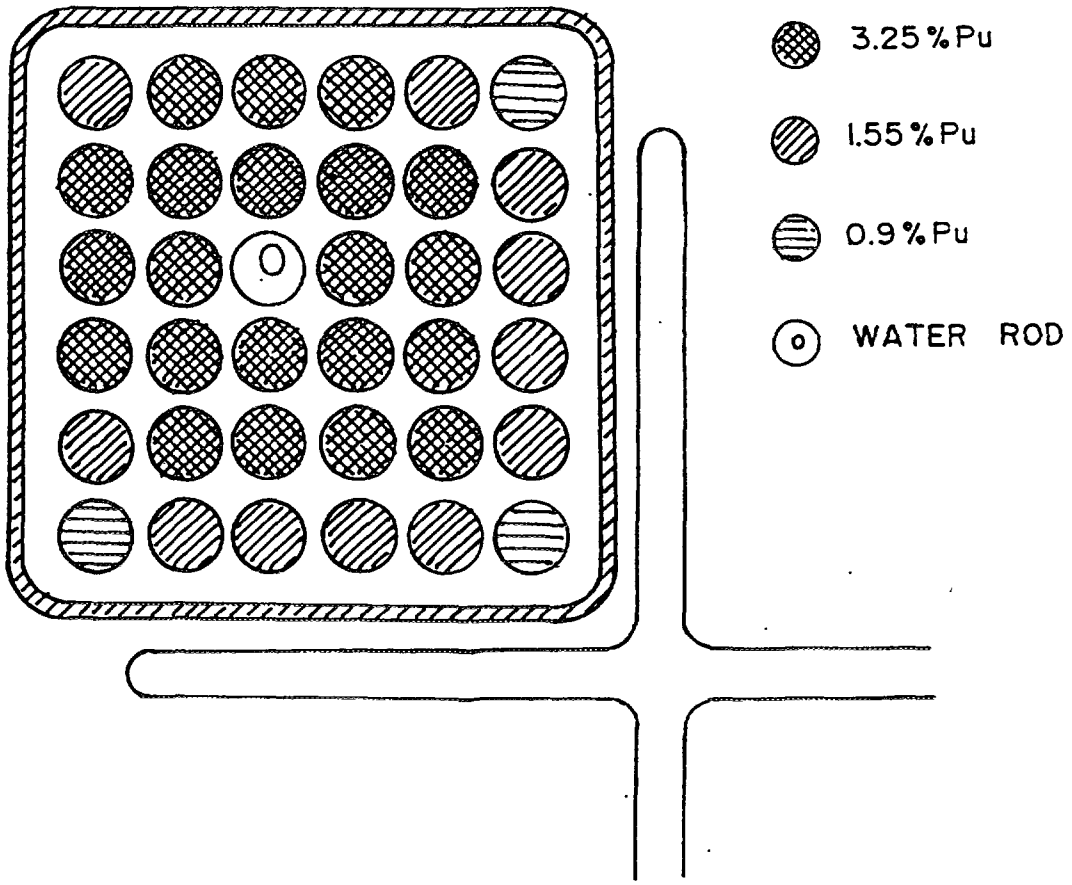

FIG. 2. MOX Assembly for BWR

TABLE II.

ISSUES ADDRESSED FOR Pu RECYCLE IN TAPS.

1.	Decrease in Control System Capability
2.	Decrease in delayed neutron fraction
3.	Increase in negative reactivity feedback
4.	Increase in long term decay heat of the spent fuel bundles
5.	Thermal margin criteria (MCHFR)+
6.	Design basis accidents (e.g. control rod drop accident)
7.	Operational transients (e.g generator lockout)

+ Minimum Critical Heat Flux Ratio

fuel for power reactors. Table-IV gives the summary of these irradiation experiments. Several fuel characteristics which are of interest for extended burn-up or economics such as higher grain size, controlled porosity, annular pellets etc. have been under study. These fuel clusters were fabricated at our Radiometallurgy Division.[3]

Based on this initial work, a flow-sheet for MOX fuel manufacture was selected, as shown in Fig.3.

TABLE III.

MOX FUEL ASSEMBLY DESIGN FEATURES.

1.	MOX fuel rod rating	-	82 w/cm^2
2.	Peak Assembly burn-up	-	25,000 MWd/Te
3.	Local peaking factor	-	1.172
4.	Axial peaking factor	-	1.273
5.	Peak pellet burn-up	-	37,200 MWd/Te

TABLE IV

EXPERIMENTAL IRRADIATIONS OF MOX FUEL (BWR) TYPE CLUSTERS IN CIRUS

Fuel Composition : UO_2 - 4% PuO_2

Sr. No.	Designation	Max.linear Rating W/cm	Burn-up MWD/Te	Remarks
1.	AC-2	414	16,265	Standard
2.	AC-3	490	16,000	Standard
3.	AC-4	490	2,000	Limited burn-up planned. Design variables studied include fuel clad gap, annular pellets, LTS, grain size and Pu cluster size.

A manufacturing plant for MOX fuel was planned and constructed at Tarapur. The plant has 2 fabrication lines, in parallel, and the initial work has commenced on one of these. To start with, work has been taken up on 10 Kg batch scale for the mechanical milling of starting powders - UO_2 and PuO_2 using an attritor. The attritor mill gives excellent homogeneity in short times and is very convenient for operation in glove-boxes. Sintering is done on 30 Kg scale in a batch sintering furnace. Although we have equipped the plant with continuous sintering systems, we have found the demands on services of batch sintering furnace much easier to meet and for the initial work, we have utilised the latter. The centreless grinder used for the sintered pellet diameter control is fitted with a composite diamond grinding wheel, which has a fairly long life, and the cutting is achieved in one pass. The coolant recirculation line has a hydroclone for the separation of the sludge and clarification of coolant. TIG welding is practiced for welding the second end-plug to tubes loaded with pellets and internal components, helium gas being filled in the annulus to a pressure of 2.5 bars. The rods are checked for contamination followed by quality control checks and are configured into fuel assemblies.

Based on satisfactory irradiation performance of the initial fuel assemblies from the plant, scaling up of production operations is in hand to recycle Pu within the allowed limits of loading in the reactors.

5. QUALITY CONTROL DURING MOX FUEL MANUFACTURE

Prior to the manufacturing campaign of the BWR-MOX, a detailed quality control(Q.C) plan was formulated to meet the stringent specifications of MOX fuel. The Q.C plan was finalised taking into account various aspects like sample size, and accuracy required. Quality control facilities were set up to carry out the various checks. The Q.C actions are subjected to Quality Surveillance (Q.S) by an independent agency which performs this function for all nuclear fuel manufacturing activities in India.

The Zircaloy hardware and UO_2 powder are supplied by the Nuclear Fuel Complex (NFC), Hyderabad which manufactures NU/LEU fuel for all Indian power reactors. PuO_2 powder prepared by the oxalate route is received from the Power Reactor Fuel Reprocessing Plant (PREFRE) at Tarapur. The documents received are scrutinised for compliance with specifications for starting materials prior to start of manufacture.

During the MOX fuel fabrication programme, two techniques of Q.C were developed and adopted. A quick check of the blends for Pu content has been carried out using a Neutron Well Coincidence Counter (NWCC). This technique has the advantages of being fast and representative of bulk material compared to the X-ray Fluorescence (XRF) Technique originally planned to be employed. During this campaign, a large number of samples were analysed for Pu content and a good control correlation was obtained with the chemical analyses carried out.

Use has been made of a passive gamma scanner to detect faulty mix-up of pellets of different enrichments in the three types of fuel rods. Further, a gamma-auto-radiography technique (GAR) has been utilised to detect mix-up of pellets of different enrichments and the detection of PuO_2 agglomerates present in the peripheral zone of the cylindrical surfaces of pellets. The standard Q.C check for ensuring plutonium homogeneity in the pellets is alpha-autoradiography of a sample pellet from every blended batch. However, GAR enables detection of PuO_2 agglomerates lying at the periphery of all the pellets in the finished rod, which is vital informaltion as Pu clusters close to the clad inner surface can be deleterious to the fuel rod performance. The GAR technique has been useful in changing some of the manufacturing procedures to improve the quality of the fuel. Figs.4 (a) & (b) are gamma-auto-radiographs showing PuO_2 agglomerates and presence of UO_2 pellet in a MOX fuel rod. Microdensitometric analysis of the gamma-auto-radiographs would yield quantitative information on the plutonium enrichment in the clusters. Both these techniques have been adopted as regular Q.C checks in the manufacturing activity.

5.1 Fuel Characteristics

Inspection of the pellets for density and dimensions was carried out as per MIL-STD 105D special level IV. All the pellets were also examined for physical integrity. The PuO_2 cluster size of sample pellets, examined by alpha-auto radiography, did not exceed 100 microns. Microstructural evaluation of pellets was done in a separate metallography line. The chemical characteristics like Pu and heavy metal contents, O/M ratio, impurities and hydrogen content have been achieved well within the specifications and histograms of some of characteristics are presented in Figs. 5(a) to (d). Dissolution test is performed on all pellet batches to ensure compliance of reprocessing requirements.

For the fabrication of the fuel rods, qualification of the TIG welding machine and its operation were carried out as per the QC plan. The welded fuel rods were subjected to contamination check, helium leak testing, visual examination, metrology, X-radiography, gamma scanning and gamma auto-radiography. The fuel assemblies were constituted from the accepted rods and subjected to various QC checks prior to shipping to TAPS.

6. CONCLUSIONS

The work carried out on MOX fuel development so far has been encouraging and it is planned to exploit the potential of plutonium utilisation initially in the BWRs, to be followed by the PHWRs until

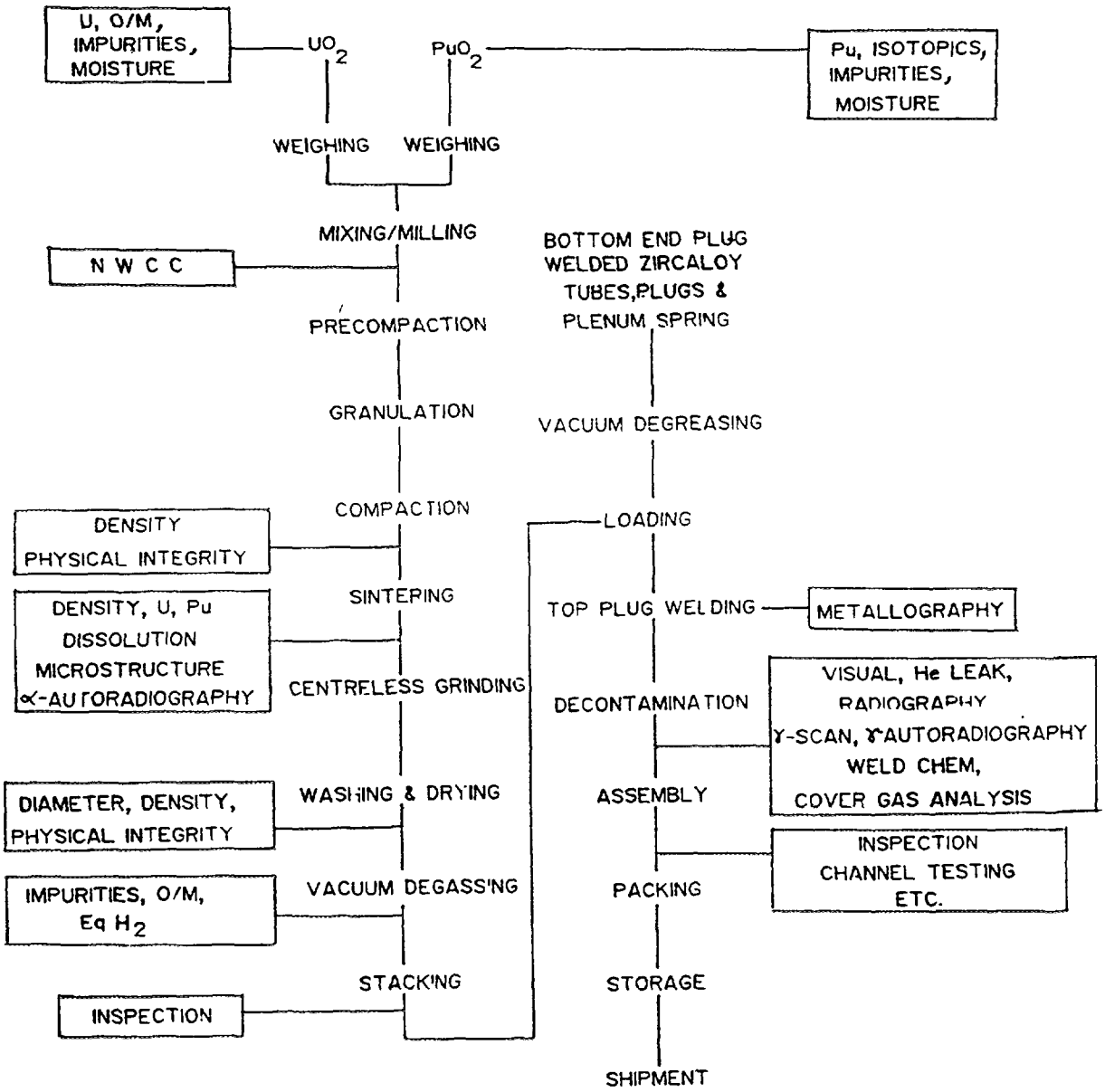

FIG. 3. *Flow Sheet for Fabrication of MOX Fuel with QC Points*

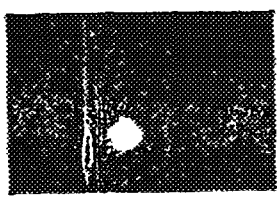

FIG. 4a *Gamma Autoradiograph Showing surface PuO_2 Agglomerate*

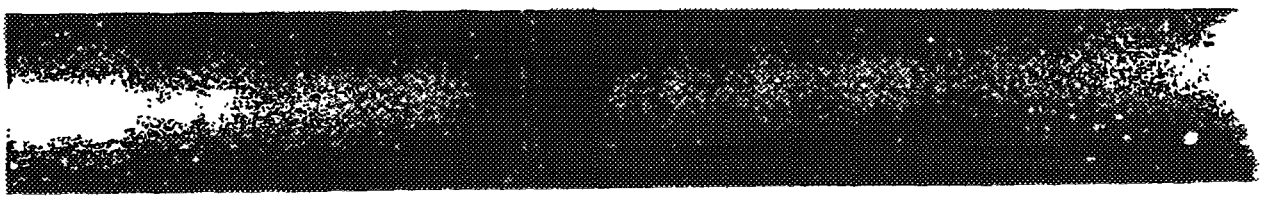

FIG. 4b *Presence of UO_2 Pellet in a MOX Rod*

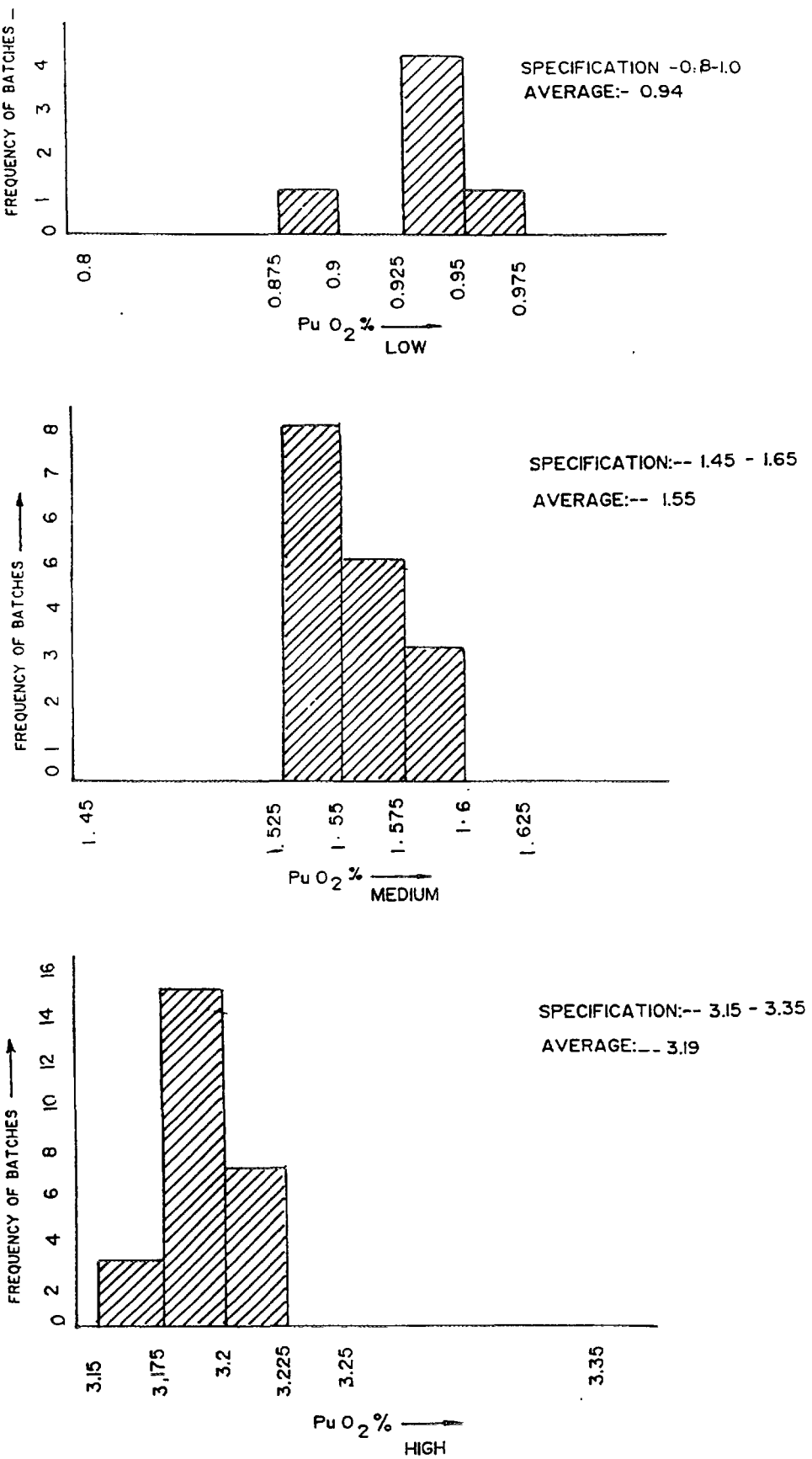

FIG. 5a *Percentage of PuO_2 in MOX Pellets*

the larger requirements of FBRs are to be met. Progressive enhancement of plutonium quantities in these two reactor types has been planned. These developments would run parallel to the fast breeder development programme and the fuel manufacturing technological innovations realised would greatly facilitate the breeder fuel manufacturing work.

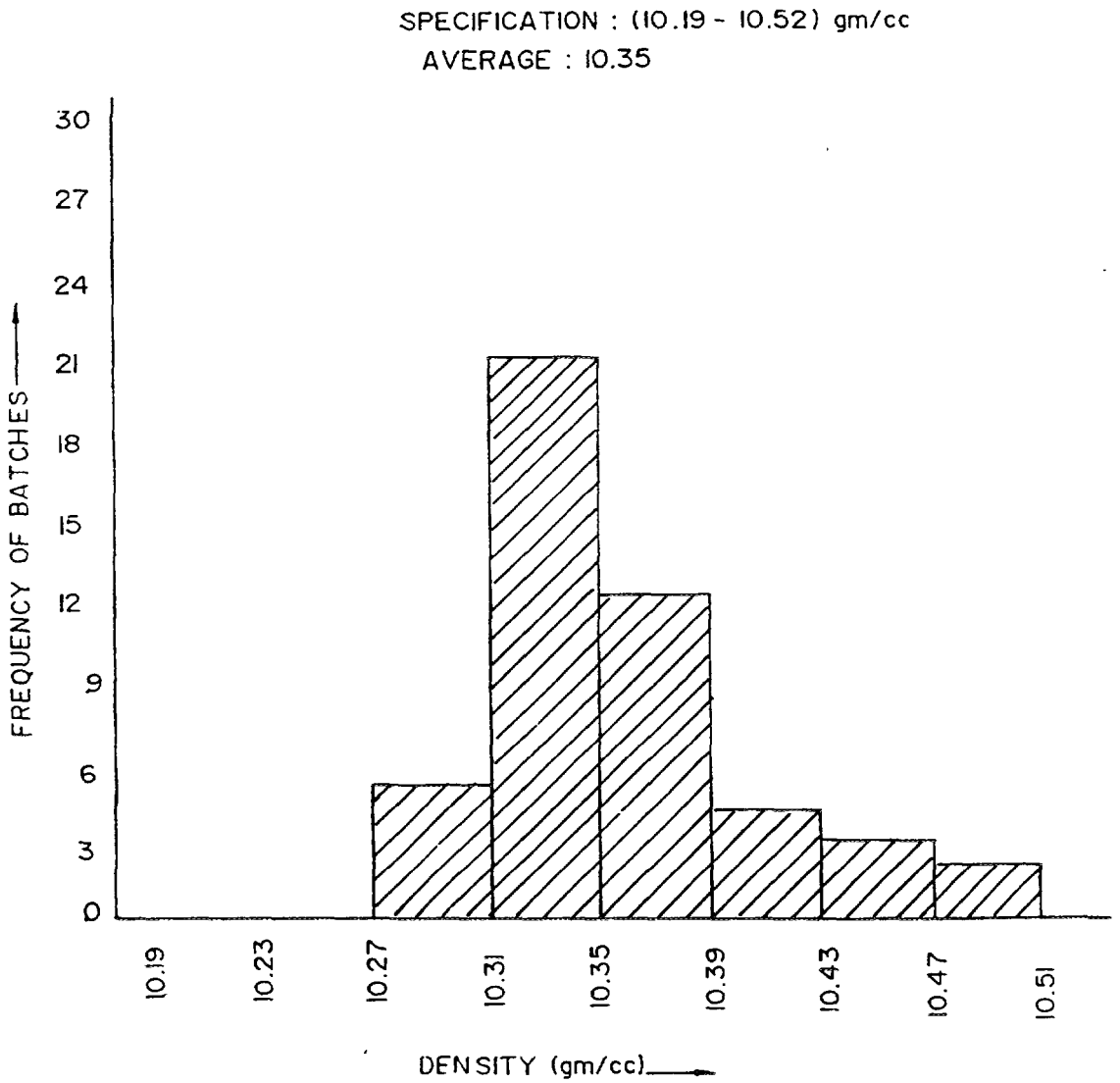

FIG. 5b Density of MOX (BWR) Fuel Pellets

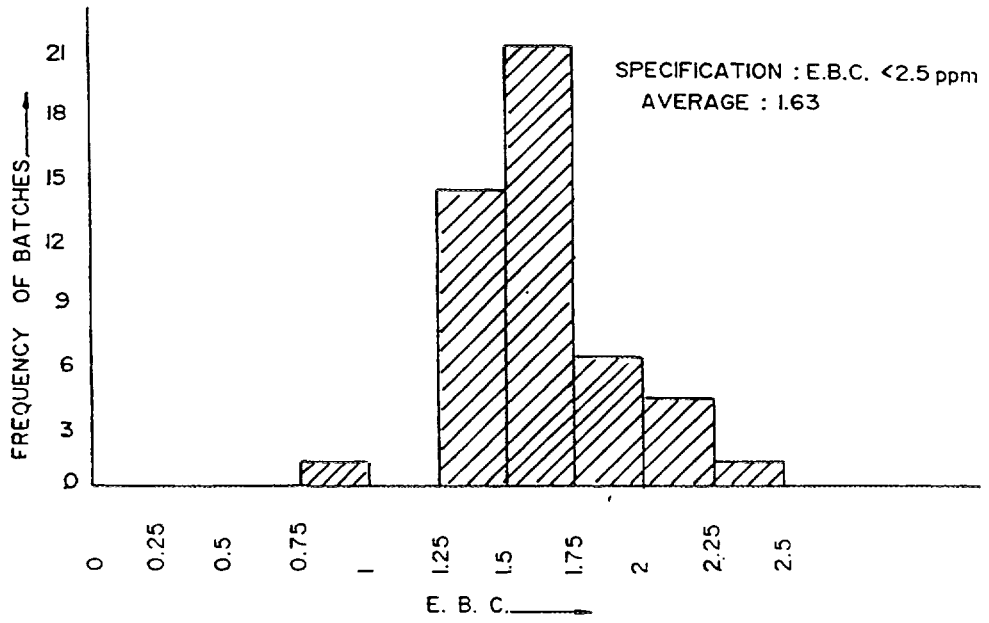

FIG. 5c Equivalent Boron Content

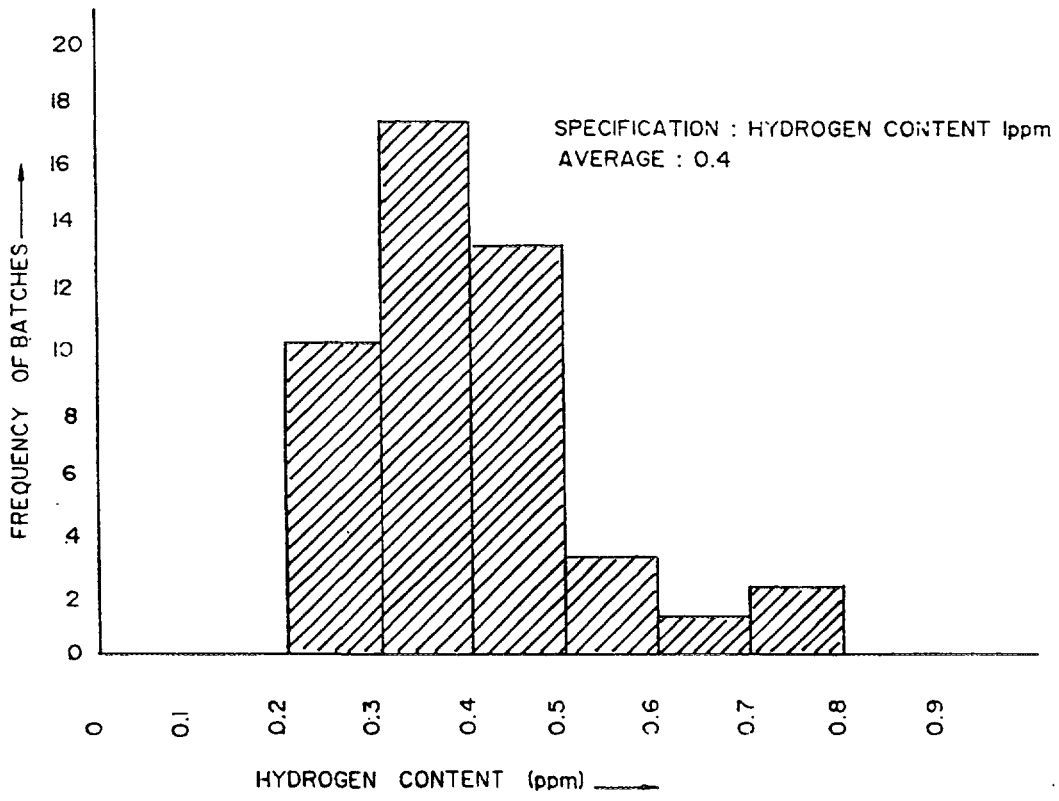

FIG. 5d Hydrogen Content in MOX Pellets

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BNFL SUPPLY OF MOX FUEL ASSEMBLIES TO THE BEZNAU 1 PWR OF NOK

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Abstract

The Swiss utility NOK has been using MOX fuel in its Beznau 1 and 2 reactors since 1978, and since that time it has irradiated more than 150 MOX fuel assemblies produced by Westinghouse, Belgonucleaire (COMMOX) and Siemens. In 1991 NOK decided to support BNFL on the provision of the MOX Demonstration Facility (MDF) at Sellafield by becoming their first customer. Following the successful commissioning of the plant, BNFL, under a sub-contract to Westinghouse, produced their first LWR MOX fuel assemblies in mid-1994 for loading into the Beznau 1 reactor. This paper considers the recycle philosophy adopted by NOK and their approach to utilising MOX fuel made by a new supplier in their reactors. The paper considers the fabrication process used by BNFL in the MDF with particular attention being given to the Short Binderless Route used to prepare the MOX press feed material. It also reviews the quality characteristics of the pelleted fuel made for the NOK fuel assemblies and provides data on the predicted performance of the fuel in reactor. After reviewing the irradiation conditions in the reactor, details are provided of a planned Post Irradiation Examination programme that is scheduled to take place in 1997-98.

1. INTRODUCTION

Nordostschweizerische Kraftwerke (NOK) is a Swiss utility supplying the north east corner of Switzerland with electric power from a mixture of hydroelectric and nuclear sources. It owns and has operated two Westinghouse PWRs of 350 MW(e) at the Beznau site on the river Aare since 1969 with very high reliability. Since the beginning Westinghouse has supplied Beznau I (KKB-l) with uranium fuel assemblies and in 1978 supplied the first four MOX assemblies to be loaded by NOK.

Since 1978 NOK has continually built up its usage of MOX based on its reprocessing contracts with COGEMA and BNFL. Initially due to the slow build up of its own Pu coming from La Hague, NOK began by using Pu loaned from other parties, for return when NOK's own material became available. In this way more flexible management of the recycle process has been possible which has allowed NOK lo build up a wealth of experience in the use of MOX assemblies. The approach also enabled the Swiss licensing authorities to gain early experience in the licensing and handling aspects of MOX assemblies and as a result generate confidence in the use of MOX in the Beznau plants, allowing for example up to 48 (from a total of 121, ie 40%) MOX assemblies to be in the core at any one time. Later, based on the experience in KKB-l, MOX was also introduced to Beznau 2 (KKB-2) under the same conditions; then as now, KKB-2 fuel was supplied by Siemens.

In 1989 BNFL embarked upon a strategy aimed at becoming one of the world's leading MOX fuel suppliers and thereby provide services which give its customers the opportunity to close the fuel cycle. The strategy included constructing at Sellafield a small-scale demonstration fabrication plant - the MOX Demonstration Facility (MDF) and later to construct a larger-scale fabrication plant - the Sellafield MOX Plant (SMP). In 1991 NOK decided to support BNFL in developing this strategy by becoming their first customer for fuel from the MDF. Following the successful commissioning of the MDF in 1993, BNFL, under a sub-contract to Westinghouse, produced their first LWR MOX fuel assemblies which were loaded into KKB-l reactor in mid 1994.

This paper considers the recycle philosophy adopted by NOK and the approach to utilising MOX fuel produced by a new supplier in their reactors. The paper also considers the fabrication process used by BNFL and l:he quality characteristics of MOX fuel made in the MDF. Finally the paper reviews the expected performance of the fuel and summarises plans for a PostIrradiation Examination Programme which will be undertaken to support validation of the fuel performance codes and provide the first data on the performance of BNFL's MOX fuel in a power reactor.

2. NOK'S RECYCLE PHILOSOPHY AND THE APPROACH TO USING A NEW SUPPLIER

Due to its early entry into the recycle business NOK adopted a philosophy of collaboration with its various fuel vendors. Figure 1 shows how a number of vendors have been involved in some or all of the stages of MOX fuel fabrication for the utility. Following the initial supply of four MOX fuel assemblies by Westinghouse (with the pellets and rods supplied from Cheswick, USA and the assemblies being manufactured in FBFC, Belgium) NOK moved to Siemens (Hanau) as a MOX supplier, then Belgonucleaire with Westinghouse and even later COMMOX involving both Westinghouse and ABB. These sometimes complex supply arrangements resulted in the delivery and irradiation of 152 MOX fuel assemblies to Beznau to date with about two thirds of the total being used in KKB- 1.

In keeping with its philosophy of collaboration and its strategy of having as wide a source of supply as possible for its fuel cycle products and services, NOK supported BNFL in the commissioning and operation of the MOX Demonstration Facility. Under carefully defined contractual terms and conditions, which included some flexibility in the delivery of the first fuel assemblies to allow for proper commissioning and qualification of the plant and process, BNFL worked closely and successfully with NOK, Westinghouse and NOK's expert consultant to develop a MOX fuel design and produce four fuel assemblies which were subsequently loaded into KKB- 1.

Because of NOK's MOX fuel utilisation, few special measures were required to license the BNFL fuel in KKB-l. All handling procedures were in place and fully proven. On arrival at the reactor site the assemblies are stored in the standard dry fuel store for uranium assemblies in individual locked and IAEA sealed storage channels. No special precautions for radiation protection are taken, over and above those imposed by routine radiation measurements on receipt and during handling. Operator proximity to the assemblies and handling times are adjusted accordingly. However the frequency of IAEA inspections is raised from three monthly to monthly during the time the MOX assemblies are in the fuel store. Movement to the transfer pool takes place at the time of reactor reloading. Unlike uranium fuel, MOX fuel assemblies are loaded at the next available fuel reload and do not form part of the strategic store of (uranium) fuel held at the plant.

NOK's policy is to operate the MOX assemblies under similar conditions of power and burn-up as the uranium assemblies loaded in the reactor. Core loading evaluation needs to take into account the possible increase in assembly peaking, slower reactivity burn out and slightly lower control rod worths although experience has shown that these can be accommodated within the normal reload planning without special operational or equipment modifications. In some more extreme loading patterns, the available neutronics codes are not able to accurately predict control rod worths and this has to be accommodated in the design. Unlike uranium assemblies the decay of ^{241}Pu and consequent build up of americium needs to be taken into consideration when defining the reactivity on the date the fuel is loaded into reactor.

To prove that the fuel provided by BNFL could meet the design performance limits it was necessary to evaluate and predict the likely performance of the MOX fuel which was to be produced by a new fabrication process and thus had unproven characteristics. To obtain the appropriate information on the fuel rod behaviour the BNFL fuel performance code ENIGMA was used. After completing an extensive joint BNFL and NOK programme which involved NOK in employing an experienced consultant, the BNFL MOX assemblies were cleared to reach a peak burn-up of 50 GWd/t on five annual irradiation cycles.

Plant	Beznau I	Beznau I	Beznau II	Beznau I	Beznau II
Recycling	1978 - 1981	1988 - 1997	1984 - 1995	1994 - 1999	1996 - 2005
Matrix-material	Natural Uranium	Tails Uranium	Natural Uranium	Tails Uranium	Tails Uranium
MOX-F.Assy. Manufacturer	WH (BN, FBFC)	COMMOX (WH, ABB, FBFC)	SIEMENS	WH (BNFL)	SIEMENS (BNFL)
Max. Assy Bu MWD/T	30 000	43 000	36 000	38 000**	42 000**
Enrichment of surrounding U-F Assay.	3.30%	3.25%	3.40% - 4.00%	3.25% - 4.00%	4.00%

** planned burnup

FIG. 1. Overview of using MOX Fuel in the Beznau Reactors

3. THE MOX FUEL ASSEMBLY CHARACTERISTICS AND APPROACH TO LICENSING THE DESIGN

3.1 FUEL ROD DESIGN

The MOX fuel rod has been designed using a MOX version of the ENIGMA fuel performance code[1]. The ENIGMA code was developed jointly by Nuclear Electric and BNFL during the 1980's for the assessment of UO_2 fuel performance. The code contains all the submodels necessary for such analyses and in sufficient detail for design and licensing evaluations. It has been independently verified and extensively validated against a data base of around 350 UO_2 fuel rods from a range of international projects and commercial irradiations. In recent years BNFL have extended the code's capability to analyse MOX and gadolinia-doped and niobia-doped uranium fuels. The MOX version of the code contains modifications to just 3 sub-models; these are: fuel thermal conductivity, fuel creep and radial power depression in the pellet. In addition a new model has been included to account for the enhanced helium release in MOX fuel. The MOX version has been validated against data from the PRIMO 1 international project and Halden projects.

The design and licensing methodology adopted was broadly in line with standard USNRC approved reload methodology. Thus the design and licensing criteria applied in the evaluation of MOX fuel were the same as those applied in the evaluation of UO_2 fuel. However because at the time of this first demonstration BNFL had no substantive manufacturing experience with MOX fuel, the fuel parameters were allowed to vary over the full range of manufacturing tolerances given on the drawings or in the product specifications in uncertainty analyses. Also because of the small data base available to BNFL on MOX fuel compared with that available on UO_2 fuel, the calculational uncertainties applied in the evaluations were larger than can be justified for UO_2 fuel. Nevertheless BNFL were able to satisfy the traditional approach to licensing by showing that all the design and licensing criteria were met for the anticipated fuel duty at Beznau Unit 1 when uncertainties were included at the 95% probability level.

Because the above approach inevitably lead to reduced margins to design and licensing criteria for MOX fuel relative to UO_2 fuel, and therefore a theoretical increase in failure probability for MOX fuel relative to UO_2 fuel, NOK requested a demonstration that the failure rate of BNFL MOX fuel would be no higher than the current experience in PWR's world-wide of approximately 1 failure in 100,000 fuel rods. BNFL therefore agreed to carry out additional calculations similar to those described above but including uncertainties at the 99.999% level rather than the 95% level. A joint NOK/BNFL review of all design and licensing criteria highlighted that the criterion most likely to be challenged by this treatment of uncertainties was that of rod internal pressure. Thus further work concentrated on rod internal pressure and successfully demonstrated that the existing criterion, of no fuel/clad gap re-opening, was satisfied even when uncertainties were included at the 99.999% level.

3.2 NUCLEAR DESIGN

With regard to the nuclear design of the MOX assemblies, the principle design issue has been in allowing for isotopic variations in the plutonium feed material. Such variations can affect two important performance characteristics of a fuel assembly - the reactivity of assemblies over their life-time in reactor (the life-time averaged reactivity or LAR) and the within-assembly power peaking factors. By recourse to the principles of reactivity equivalence, however, BNFL has been able to account for both of these effects [2]. Reactivity equivalence, expressed through a simple formula, relates variations in isotopic composition to plutonium concentration changes such that the LAR of MOX assemblies made is kept constant.

Using an equivalence formula as described in Reference 2, the assembly average plutonium concentration was fixed so as to give an LAR equivalent to an existing 3.25 w/o enriched UO_2 assembly in the KKB-1 core. Low, medium and high plutonium concentration zones within the assemblies were then fixed so as to ensure acceptable power peaking. Additional peaking issues caused by isotopic variability in the plutonium in each zone were accommodated within the engineering hot channel uncertainty factor

FQE. The licensing limit for FQE was increased from 3% to 4% in order to accommodate the additional uncertainties arising from the use of MOX fuel.

4. FABRICATION PROCESS USED BY BNFL

The flowsheet adopted by BNFL in the MDF, summarised in Figure 2, is basically the same as that used by other MOX fuel manufacturers. The exception to this is the use of the Short Binderless Route for blending and conditioning of the MOX powder and preparation of the press feed.

The MDF consist of four main areas of plant:

- Fuel pellet production
- Fuel rod production
- Fuel rod inspection
- Fuel assembly manufacture and inspection

The plant is co-located with other MOX support facilities including a comprehensive ceramography and metallography facility for carrying out quality control of fuel pellets and fuel rod weld samples, and development facilities for plant trouble-shooting and process optimisation.

The heart of the process is the pelleting plant where UO_2 and PuO_2 feed materials are weighed out in the correct proportions and processed to press feed by the Short Binderless Route. This Short Binderless Route process uses a high energy attritor mill to blend the feed powders and a spheroidiser to condition the powder before it is used as a feed to a pelleting press. At the milling stage zinc stearate lubricant and Conpor pore former are added; the latter is used to control the pellet density and ensure that the characteristics of the MOX pellets are similar to those of UO_2 pellets produced by BNFL from IDR-UO_2 powder. The milling stage is undertaken for a period of up to 60 minutes and spheroidising is completed within a similar time frame. Using this process 25 kg batches of press feed of consistent quality can be manufactured during a period of about two hours. In MDF the press feed granules are pressed to green pellets which are transferred by a cushion transfer conveyor to the furnace boat load station. Here the pellets are carefully loaded by a 'pick and place' machine into sinter furnace boats which are then charged to the furnace. Pellets are then sintered at 1650°C over a cycle time of 24h before being discharged from the furnace for dry grinding and subsequent inspection. After inspection pellets of acceptable quality are loaded into an in-line pellet store pending loading into fuel rods.

Fully inspected pellets are manually loaded, in 1m long sub-stacks into pre-dried fuel cans in which the bottom end plug has been welded and inspected. Each pellet sub-stack is weighed and when the complete fuel column has been loaded into the rod it is processed through various work stations to have the spring and top end plug inserted and welded before final pressurisation with helium and eventual seal welding. The surface of the rod is then checked for contamination before it is transferred to the rod inspection area. Each fuel rod is uniquely identified with a bar code which is entered into a computer based traceability system at each work station in the rod production and inspection areas.

The fuel rod inspection area contains shielded work stations to inspect and check that the rods meet the specification requirements for:

- Leak tightness.
- Conformance with the weld acceptance criteria.
- Dimensions and straightness.
- Rod enrichment.
- Surface finish.

Certified fuel rods are transferred to the rod store where they are loaded into magazines in the same location as in the finished fuel assembly; rejected rods are passed back to the rod fabrication plant for reworking or breakdown.

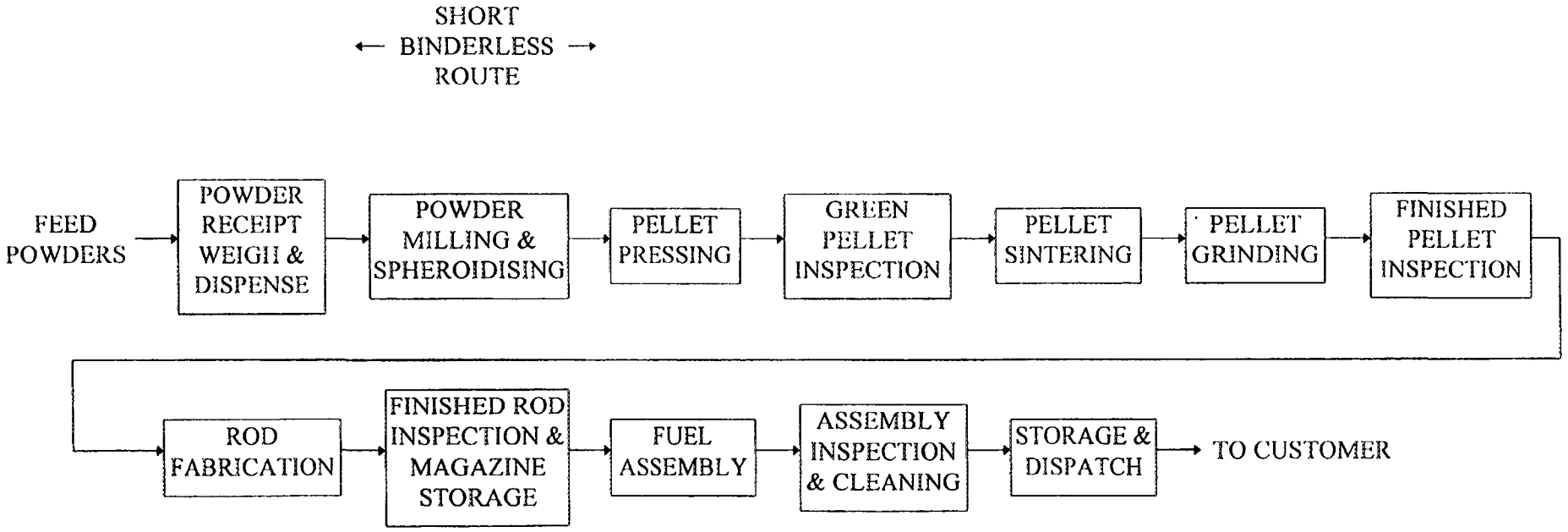

FIG. 2. The MDF Flowsheet

The MDF fuel assembly and inspection area has been designed for PWR fuel assemblies although it could be modified to accommodate BWR designs. In this part of the plant certified magazines are removed from the rod store and brought adjacent with a rod loading machine which transfers the rods into the fuel assembly skeleton. After fitting the top and bottom nozzles assemblies are then inspected to check they meet the specification requirements for:

- Dimensional envelope
- Channel spacings
- Cleanliness
- Control rod withdrawal force
- Surface finish

In this area of the plant all fabrication processes and inspection work is carried out inside heavily shielded enclosures to minimise dose uptake to plant operations personnel.

5. QUALITY CHARACTERISTICS OF BNFL MOX FUEL PELLETS

Following the commissioning of the MDF, plutonium was introduced into the plant in October 1993. The plant and processes were then qualified to meet NOK and Westinghouse acceptance criteria. The following paragraphs summarise the data obtained on the key quality characteristics during the fabrication of several tonnes of MOX fuel pellets in the plant. The MOX pellets were produced against a BNFL MOX pellet specification developed in conjunction with NOK, Westinghouse and NOK's experienced consultant. The details of this specification are confidential but the following comments are made about the pellet quality.

(i) No difficulties have been experienced controlling the pellet dimensions, the density, surface finish or thermal stability of the fuels made in MDF. The standard deviation on pellet diameter is 0.003 mm and on geometric density is 0.026 g/cc. The surface roughness of pellets produced in the plant averages 0.58 μRa with a standard deviation of 0.092 μRa. The thermal stability of the MOX pellets is well within the specification limits set by BNFL for UO_2 fuel.

(ii) The key chemical characteristics of the fuel pellets are summarised in Table 1. The hydrogen content of the pellets produced was low and tests showed that pellets produced do not pick up hydrogen or moisture when stored in air. This behaviour is similar to that of IDR-UO_2 pellets produced by BNFL Fuel Division at Springfields. The oxygen to metal ratio (O/M ratio) of the fuel produced was consistently close to 2.000.

No difficulties were experienced controlling the fissile material content of the fuel within the specified enrichment tolerances. This is an important observation as BNFL is the only MOX producer who directly produces fuel of the correct enrichment without going through a master blend process, and thus supports the application of the process in the larger-scale Sellafield MOX Plant.

(iii) The microstructural properties of the pellets produced are excellent and consistent. The grain size of the fuel is consistent throughout the pellets and averages 7.4 μm with a standard deviation of 0.6 μm. The pore size distribution is frequently assessed as part of the specification requirements and shows that it is consistent throughout the pellets and between the lots examined. For pores with a diameter greater than 5 μm the median pore size has never exceeded 15.4 μm during the production to date.

The homogeneity of the fuel pellets produced by the Short Binderless Route is extremely good when measured by colour autoradiography. To substantiate this claim the Pu/(U+Pu) ratio has been assessed using Electron Probe Micro Analysis. The data available, summarised in Figure 3, show that in pellets with a mean Pu/(U+Pu) ratio of 5.5% the highest plutonium rich region found in the pellets had a Pu/(U+Pu) ratio of 32%.

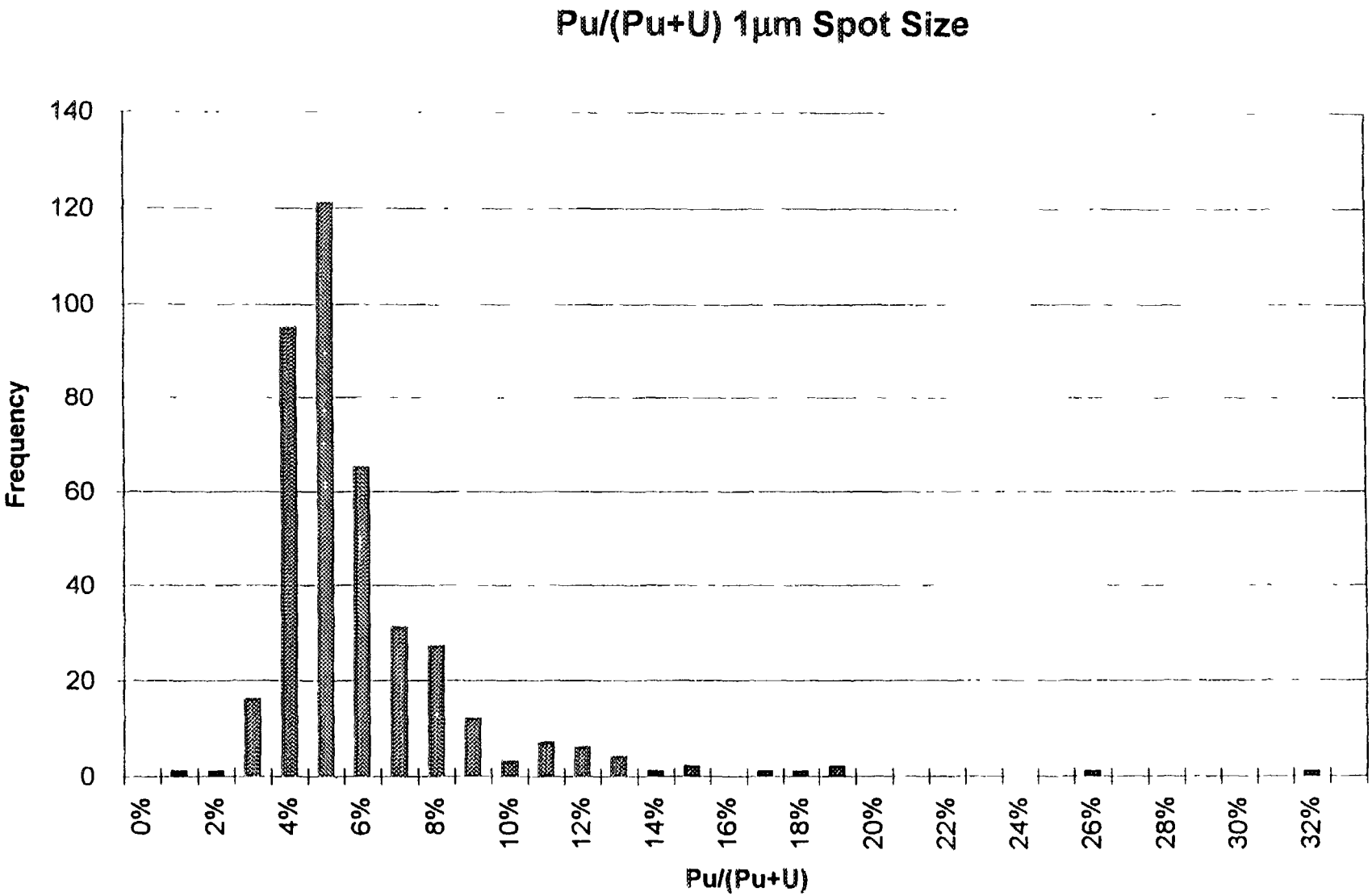

FIG. 3. *Histogram representing the homogeneity of SBR MOX pellets measured by EPMA*

6. LESSONS LEARNT DURING THE EARLY STAGES OF OPERATING MDF

Although the fabrication plant and processes used by BNFL are now becoming well established the need to produce good quality product to a closely defined schedule proved demanding on occasions for both the customer and the supplier. During plant commissioning and qualification a number of valuable lessons were learnt before BNFL could ensure that the plant was capable of producing fuel which met the requirements of a reliable fuel manufacturer matching previous NOK experience. Some of the important lessons are outlined below.

(i) Despite all plant systems and QA procedures being in place prior to the start of plant commissioning it was necessary to revise the systems for actual production; to meet this requirement it was essential that a structured approach to revising and implementing, the new system was available.

(ii) The need for a comprehensive development programme to test the key parts of the plant design was emphasised during the early stages of commissioning and operating the plant. Although no difficulties were experienced scaling up the Short Binderless Route process, the plant commissioning period might have been shortened if some engineering aspects of the plant design had been more fully proven.

(iii) The successful operation of a MOX fabrication plant needs the skills of both the fuel fabrication plant operators and those with plutonium handling expertise. Issues associated with pellet pressing and pellet grinding were addressed by experts from BNFL Fuel Division who were experienced with UO_2 fuel production.

(iv) Advanced inspection techniques are employed in MDF to minimise operator dose. Time must be made available for operators and inspectors (plant and customer) to become familiar with and trained in using these techniques.

(v) When developing new inspection techniques care should be taken not to develop methods which highlight differences not identified using conventional methods. For example a high magnification camera imaging system used to visually examine the surface of rods and assemblies may identify detailed features which are of no consequence to the performance of the fuel and give unnecessary concern to the inspector. The plant operators need to be able to respond quickly to these 'non' problems to allay customers concerns.

(vi) Where fully automated systems are developed either fall back systems need to be available or alternative approaches developed so that plant throughput is not restricted.

7. POST-IRRADIATION EXAMINATION PROGRAMME

Due to the lack of a data base on the performance of SBR fuel, BNFL has a number of initiatives in place to obtain fuel performance data. These include carrying out a number of test irradiations and participation in collaborative schemes with utilities and vendors. To this end BNFL and NOK have agreed to collaborate on a programme aimed at obtaining data on the performance of SBR fuel pellets loaded into the first four fuel assemblies produced in MDF.

The scope of BNFL/NOK programme includes carrying out special characterisation work on pellets and rods loaded into one of l.he group of four assemblies loaded into KKB- 1 in mid 1994. The assembly containing the eight rods will be removed from the reactor in mid-1997 and after cooling some of the rods will be subjected to Post-Irradiation Examination (PIE). At discharge the high enrichment rods should have reached a peak pellet burn-up of 36 GWd/te. The assembly average linear rating of the fuel during irradiation will be 183 W/cm.

The PIE programme will include the following:

- Non-destructive testing
- Gas release
- Pellet ceramography
- Fuel EPMA/SEM examination
- Burn-up analysis

In addition tests will be carried out to establish the level of improvement in fuel solubility that results from irradiating SBR fuel. This data is important to BNFL who, as one of the world's leading reprocessing companies, fully intend to reprocess MOX as well as UO_2 fuel in their plants at Sellafield. Finally, consideration is being given to developing another programme to obtain information on the properties of unirradiated (using archive samples) and irradiated MOX fuels and carrying out ramp testing and subsequent PIE on refabricated fuel segments selected from the rods.

TABLE I SOME CHEMICAL CHARACTERISTICS OF SBR MOX PELLETS

Characteristics	Mean Value	Standard Deviation of Mean
O/M ratio	2.000	0.000
H_2 (ppm)	0.25	0.11
Gas content (μl/g)	8.99	5.7
C (ppm)	37.6	21.2
Cl (ppm)	3.0	2.7
F (ppm)	0.8	0.7
Fe (ppm)	66.5	43.7
Mo (ppm)	4.9	4.3
N (ppm)	9.5	8.9
Zn (ppm)	4.1	3.3

8. SUMMARY AND CONCLUSIONS

By continuing to promote their philosophy of collaborating closely with their fuel vendors and service providers NOK have provided vital support to BNFL in the first stage of fulfilling their strategy to become one of the world's leading MOX fuel suppliers. The close involvement of NOK in the commissioning and early stages of operating the MDF have provided assurance that the plant is capable of meeting its requirements for producing good quality MOX fuel. The lessons learnt during the commissioning and operation of the MDF, together with the vast experience commissioning new UO_2 fabrication plants, place BNFL in a sound position to successfully commission and operate the larger-scale SMP now under construction.

ACKNOWLEDGEMENT

The authors would like to thank staff at NOK, BNFL and Westinghouse who have given them their support throughout the duration of the project.

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SESSION 2
MOX FABRICATION AND REPROCESSING

TECHNICAL DESIGN CONSIDERATIONS IN THE PROVISION OF A COMMERCIAL MOX PLANT

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Abstract

The Sellafield MOX Plant (SMP) has a design production target of 120 t/year Heavy Metal of mixed uranium dioxide and plutonium dioxide (MOX) fuel. It will have the capability to produce fuel with fissile enrichments up to 10%. The feed materials are those arising from reprocessing operations on the Sellafield site, although the plant also has the capability to receive and process plutonium from overseas reprocessing plants. The ability to produce 10% enriched fuels, together with the requirement to use high burn-up feed has posed a number of design challenges to prevent excessive powder temperatures within the plant. As no stimulants are available to represent the heat generating nature of plutonium powders, it is difficult to prove equipment design by experiment. Extensive use has therefore been made of finite element analysis techniques. The requirement to process material of low burn-up (i.e. high fissile enrichment) has also impacted on equipment design in order to ensure that criticality limits are not exceeded. This has been achieved where possible by 'safe by geometryþ design and, where appropriate, by high integrity protection systems. SMP has been designed with a high plant availability but at minimum cost. The requirement to minimise cost has meant that high availability must be obtained with the minimum of equipment. This has led to major challenges for equipment designers in terms of both the reliability and also the maintainability of equipment. Extensive use has been made of theoretical modelling techniques which have given confidence that plant throughput can be achieved.

1. INTRODUCTION

The design and construction of the Sellafield MOX Plant (SMP) is being carried out by BNFL Engineering Limited. This paper gives an overview of the capabilities of SMP and outlines some of the technical issues that have been encountered in the design. Examples are used to illustrate the techniques for achieving design solutions to the technical issues.

2. PLANT OUTLINE

The basic plant requirements as defined by the client were;

- A design throughput of 120 t/year Heavy Metal.
- The plant must be capable of handling any plutonium dioxide material likely to arise from reprocessing operations at Sellafield site, together with the ability to receive plutonium dioxide from overseas reprocessors. Thus there is a wide variation in the composition of plutonium dioxide that the plant can receive.

Initially the plant will utilise depleted or natural uranium dioxide as its feed but has the capability to use material arising from oxide reprocessing if required.

In terms of products SMP will be capable of producing pellet enrichments up to a maximum of 10% fissile plutonium for a wide range of pellet sizes. Fuel rod dimensions and assembly configurations to suit the majority of light water reactor designs can be manufactured. The plant product is complete PWR and BWR fuel assemblies.

The plant is located adjacent to the THORP reprocessing plant at Sellafield to maximise the use of existing services and infrastructure. It also enables direct transfer of material from THORP. The existing site services for analysis and waste treatment and management are also employed.

2.1 FLOWSHEET

BNFL has wide experience in the use of a binderless route for the production of uranium fuels. For SMP a 'short' binderless route has been developed by BNFL which incorporates high energy milling. This has the following benefits;

- Reduced milling time allows powders to be milled at the target enrichment.
- Elimination of the pre-compaction stage reduces equipment requirements
- Equipment size can be reduced.

The use of attritor milling, together with the elimination of a pre-compaction stage, allows the equipment to be linked together thus eliminating the need for make and break connections every time a batch is transferred between process stages. This reduces the potential for contamination spreading within glovebox containment and hence reduces total background radiation levels. The ability to mill all powder at the target enrichment also leads to an extremely homogeneous product.

An outline flowsheet is shown in Figure 1.

2.2 FLOWSHEET DEVELOPMENT

The short binderless route has been proved in the MOX Demonstration Facility (MDF), which is also located at Sellafield. This plant has a design capacity of 8 t/year and has successfully produced fuel which is currently loaded into reactors.

Extensive development work has been undertaken to demonstrate that the required scale-up to 120 t/year required by SMP is achievable. Initial development has been carried out, using uranium, on individual equipment items. Further work is being carried out on a full-scale, integrated test facility to define the operating window prior to plant testing and commissioning.

Particular emphasis is being placed on the ability to re-incorporate production scrap directly into the process as MOX rather than by the oxidation route that has been used previously on BNFL uranium facilities. This too is being demonstrated, both in the uranium facility and also, at a reduced scale, using MOX material.

2.3 ENGINEERING DESIGN DEVELOPMENT

With a high throughput alpha active plant such as SMP and with the overriding requirement to minimise operator dose there is the need for a high degree of remote equipment operation. This has largely been achieved by the use of automated production equipment. Such equipment is not readily available and a number of process systems, while using standard engineering techniques, incorporate some novel design. BNFL Engineering Ltd. has substantially completed an engineering development programme to demonstrate key features of the design.

Particular emphasis is being placed on remote welding equipment for fuel rods and also automated inspection techniques, including the use of automated real time CCTV imaging and laser surface inspection.

In order to minimise maintenance radiation doses, modular design has been incorporated.

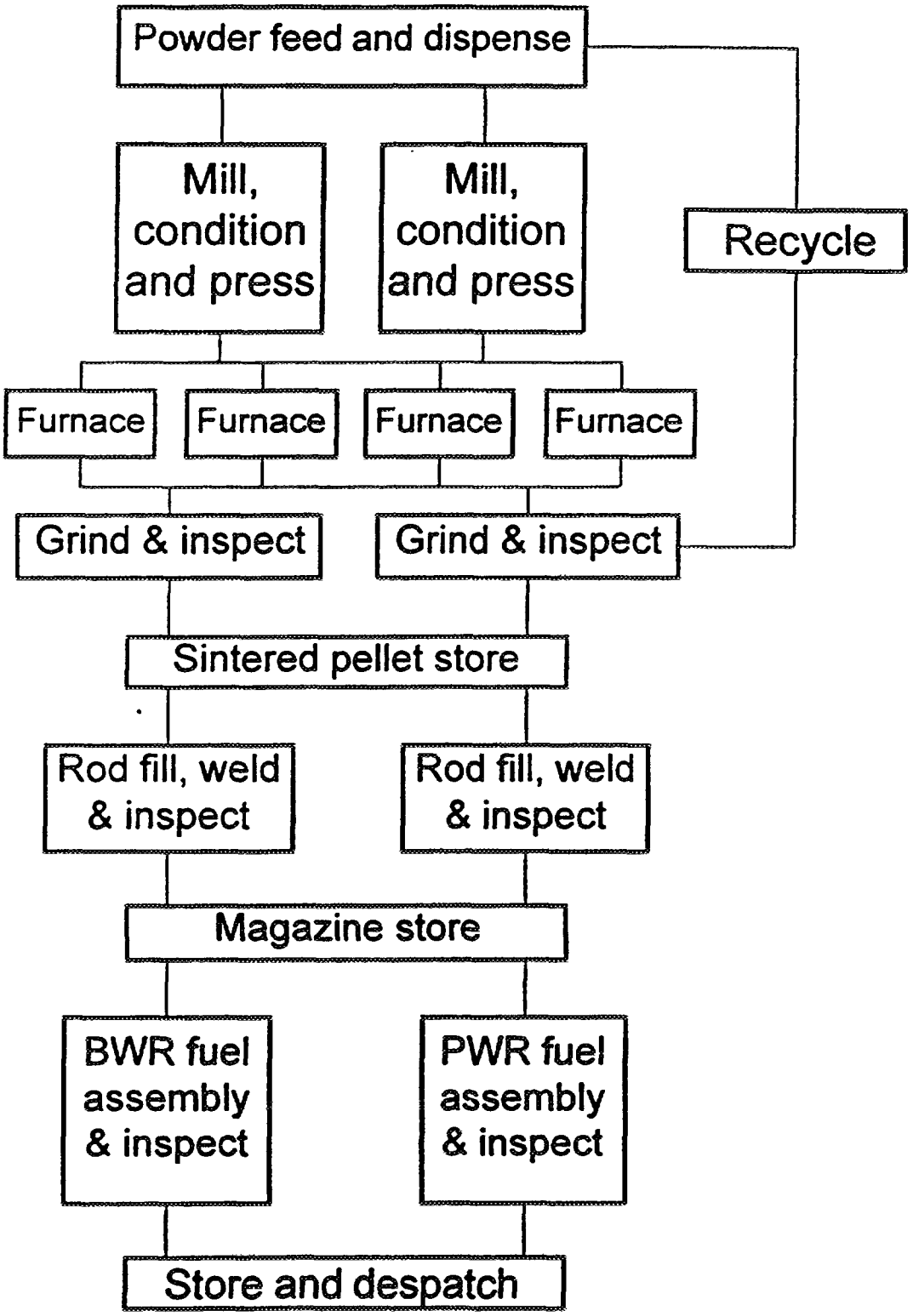

FIG. 1. SMP Process Flow Diagram

3. DESIGN ANALYSIS

With both areas of development mentioned above extensive use is made of simulant materials to replicate the behaviour of MOX materials.

For flowsheet development uranium dioxide has sufficiently close properties to MOX for it to be used as a representative simulant. Likewise, for engineering development, extensive use has been made of uranium pellets and other simulants, such as steel pellets, which replicate the parameters under investigation.

Such simulants allow development work to be undertaken without the requirements for alpha active facilities which would be required for work with MOX materials.

There are however some properties of plutonium dioxide that are difficult to simulate. One such is its heat generating nature. MDF has been used to process material arising from relatively low burn up fuel, typically 30 GWd/t. SMP however has to process material of between 30 and 55 GWd/t. This leads to a ten-fold increase in the heat generated within MOX powder in SMP compared to that experienced in MDF.

The usual method to remove heat from process equipment would be to employ a heat transfer medium and forced convection to carry the heat away from the equipment. In the case of SMP the use of water is excluded by criticality safety considerations. The alternative of high gas flows through containment systems is not favoured due to the potential to spread contamination throughout the plant. Further problems are encountered due to the poor conductivity of MOX powders. One approach would be to agitate powders to improve conductivity of the bulk powder. However in a plant that goes to some length to agglomerate powders to improve the flow properties high agitation levels cannot be tolerated as they may destroy the required powder characteristics.

Because no simulant is readily available to investigate the impact of this heat generation rate on plant and equipment, BNFL Engineering Ltd. has adopted a theoretical approach to its development work, making extensive use of computer analysis techniques to predict temperatures within equipment and ensure that excessive temperatures are not generated.

Finite element analysis has been used to model particular items of equipment. This involves the production of a model net in either two or three dimensions as appropriate. The mesh represents both the geometry and the material. The thermal properties of the material are applied and either steady state or dynamic analysis performed. Generally the process equipment contains heat generating powder that is resident for varying lengths of time. This makes the model, by definition, dynamic. However to allow for fault conditions where powder may be held up in equipment, steady state analysis is carried out.

For example the plant contains a number of hoppers which, for reasons of criticality prevention, are designed as slab vessels. Heat transfer assessments made of the initial design of hopper indicated that unacceptably high temperatures might be experienced during fault conditions. Further analysis was carried out to determine the optimum hopper dimensions to allow heat to be successfully dissipated and the hopper redesigned.

Analysis has also been carried out using computational fluid dynamic (CFD) techniques to demonstrate that natural convection will give sufficient heat flow to maintain equipment at acceptable temperatures.

CFD analysis was used to confirm the design of a store for boats of fuel pellets. Initial designs had shown an unacceptable temperature build-up within the store due to poor gas flow. CFD analysis was used to optimise the flow and achieve acceptable temperatures.

A major problem with the use of such computer modelling is validation of the results. To this end some development work has been performed to model the plant equipment. This has shown that the model predicts similar equipment temperatures to those found experimentally.

4. EQUIPMENT RELIABILITY AND THROUGHPUT

Equipment development by its nature tends to investigate pieces of equipment in isolation. Generally it is not cost effective to perform extensive development on large integrated sections of plant prior to plant commissioning. An alternative method is therefore required for demonstrating plant capability and exploring the sensitivity of throughput to variations in operating parameters and assumptions.

The requirement to minimise overall radiation dose within a plant such as SMP leads to the requirement to minimise in-plant inventory and, therefore, buffer storage. A consequence of this is that all equipment becomes closely matched in terms of its throughput.

To carry out this work, BNFL Engineering Ltd produced an operations research model of the plant. The model reflects the plant flowsheet and operational sequences and incorporates data and assumptions on equipment failure rates and repair times. Random number techniques are applied to the frequency and duration of failures to allow investigations of the expected throughput of the plant and identification of any bottlenecks. The model is currently being used to determine the optimum operating regime to maximise throughput for given plant order schedules.

Data on expected equipment availability has been obtained by comparing SMP equipment with similar equipment in use on uranium fuel plants with appropriate account taken of the alpha active environment in which SMP equipment will be operating.

During initial plant design the capacity of the plant blender was investigated. Theoretical modelling allowed the blender capacity to be optimised so as to maintain overall throughput while minimising sampling costs.

5. SAFETY CONSIDERATIONS

As with any facility safety is considered of paramount importance and full account of safety requirements must be taken in the plant design. The main hazards identified in the plant were dose uptake, criticality and loss of containment. Detailed dose uptake assessments, including the use of computer modelling to calculate expected dose rates, have been carried out to assess proposed plant design and identify areas of concern. Particular use of these techniques has been to determine equipment shielding requirements and the information has then been used to allow design improvements to be carried out.

With a highly automated plant such as SMP there is the potential that increased plant complexity will lead to higher maintenance doses. Detailed evaluation of equipment design at an early stage has been carried out and the findings incorporated into plant design. This will confirm that equipment is readily maintainable and that dose uptake targets are achieved.

The plant has been analysed to identify criticality hazards. The small size of process equipment within the powder processing area made possible by the use of the short binderless route has enabled all the powder processing area to be designed as inherently safe by shape.

For downstream plant areas simple but high integrity protection systems, which are independent of the plant control system, have been engineered.

6. SUMMARY

The Sellafield MOX plant has represented a major challenge to the design team of BNFL Engineering Ltd and is a significant scale up of the MOX Demonstration Facility.

In order to demonstrate that scale up is achievable an extensive development programme has been carried out. This programme encompasses both traditional laboratory and pilot plant scale equipment and the use of computer analysis techniques.

In the safety area a systematic approach, based on design analysis at an early stage and feedback into detailed design, ensures that the plant design complies with all BNFL's safety criteria.

MOX FUEL FABRICATION EXPERIENCE AT BELGONUCLEAIRE

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Abstract

The paper reviews the status of MOX fuel fabrication for Light Water Reactors (LWRs) at the BELGONUCLEAIRE Plant at Dessel (Belgium), as well as the irradiation experience accumulated so far. Since 1986, about 240 tons of MOX fuel rods have been fabricated and loaded in commercial reactors in France, Switzerland, Germany and from 1995 on, in Belgium. Their excellent in-reactor performance up to 50 GWd/tHM has been demonstrated through surveillance programmes as well as specific experimental tests, the main results of which will be reviewed. These fabrications were achieved according to the MIMAS (MIcronized MASter blend) process developed at BELGONUCLEAIRE. Its main features and advantages will be presented, as well as the efforts which are scheduled in order to further improve the fabrication records.

1. INTRODUCTION

Although the BELGONUCLEAIRE (BN) MOX plant went first in operation in 1973 already, the industrial production started in 1986, using the new fabrication process MIMAS (MIcronized MASter blend) described hereafter. The evolution of BN-Dessel as well as the various processes which were experienced earlier have been extensively described elsewhere [1].

The main achievements reached over 9 years (1986-1994) of operation of the plant are presented in the next chapter, with emphasis on the production experience gained so far and on technical aspects having an influence on the fabrication capacities and hence on economics.

The MOX fuel produced in the BN-Dessel plant is being irradiated since several years in LWRs in France, Germany, Switzerland and Belgium where the first loading occurred in 1995. The fuel behaviour is being monitored by surveillance programmes in France and Switzerland. In parallel, experiments are carried out on a smaller scale in order to investigate particular aspects of the fuel behaviour up to high burnups. A summary on such programmes and typical results are provided in chapter 3.

Finally, chapter 4 looks into the future by reviewing the main targets and improvements to be reached, as well as the positioning in the future MOX production market of the Dessel medium size flexible MOX fabrication plant.

2. MOX FABRICATION : 1986-1994

2.1. THE MIMAS PROCESS

The MIMAS (for MIcronized MASter blend) process was developed in the mid-eighties at BN to replace the former process used in the Dessel plant, which consisted in direct blending of UO_2 and PuO_2 powders and was mainly used for the fabrication of the first core of the German Fast Breeder Reactor SNR-300. The main reasons for switching to the new process were the requirement to produce soluble fuel, for further reprocessing, and also the necessity to better adapt the fabrication to the specific conditions of LWR fuel specifications.

Basically, MIMAS MOX pellets are made of a solid solution of UO_2 and PuO_2, soluble in nitric acid, and homogeneously dispersed in a UO_2 matrix. This result is obtained through two blending steps (Fig. 1): the primary (or master) blend obtained by ball-milling and the secondary (or final) blend.

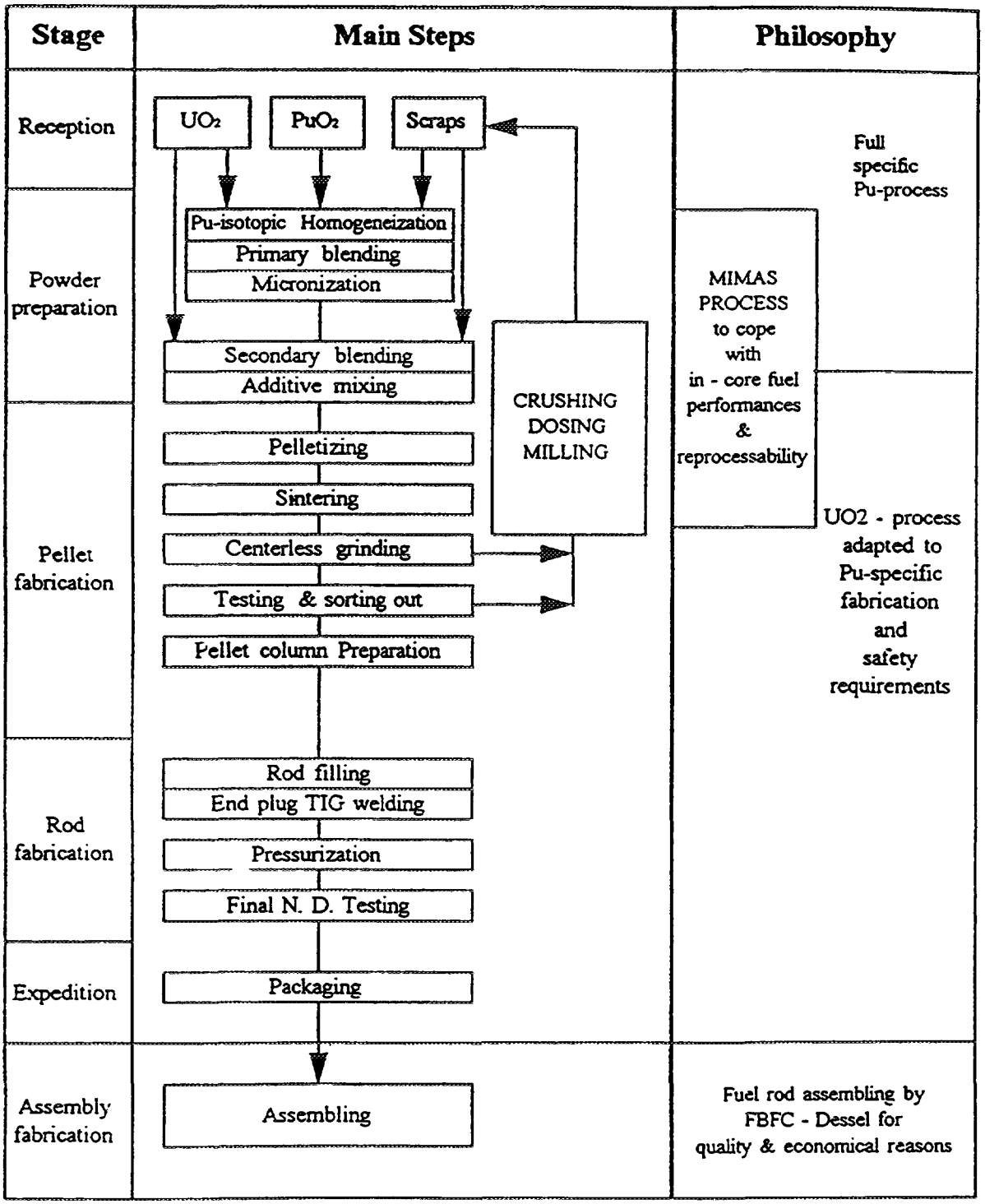


FIG. 1. MIMAS Process

Thanks to this double blending, the MIMAS process can lead to excellent isotopic homogeneity of the Pu in the product, even with Pu of various origins (LWRs, GCRs), various forms (PuO_2 powder as produced by European reprocessors or, if required, UO_2-PuO_2 mixture produced in Japan) and various batch sizes. It should be noted that the UO_2 powders used up to now are free-flowing, either of the AUC type or of the ADU type developed by COGEMA and recently qualified at the BN-Dessel plant [2].

The homogeneity of the product, within acceptable specification limits, is of prime importance to the reload designer to reduce uncertainties in MOX assembly design and operation.

Another characteristic of the MIMAS process is the dry recycling of the scrap (rejected pellets, grinding powder) which is re-introduced at the primary or at the secondary blending step. Owing to the large flexibility in terms of homogeneisation, scrap of previous fabrication campaigns or of different Pu contents can be re-utilized.

The sintering conditions have been adapted to the conditions of the plant furnaces and to the requirements of the customers (solubility, density, hydrogen content, ...).

The remainder of the process is close to the UO_2 production process. Only the technology is different, for alpha-toxicity and criticality prevention reasons.

2.2. MOX FABRICATION TECHNOLOGY

BN has developed a specific technology to produce MOX fuel with industrial quality level, in accordance with specific safety and safeguards requirements imposed by the presence of plutonium.

The main items of the technology developed and used by BN are related to [3]:

* powder preparation : blending and milling equipment, isotopic homogeneisation, scrap recycling, dust collection, reduced fissile material holdup;
* sintering under moistered hydrogen/argon atmosphere;
* dry pellet grinding;
* contamination-free rod loading and welding ;
* adapted non-destructive testing.
* fissile material inventory, real time followup and book-keeping.

This technology would be without any industrial future if it would not lead to a good economy of the fabrication. The fact that the capacity of 35 tHM/year has been achieved, on an average, over the last five years at competitive fabrication prices demonstrates the success of the process as well.

2.3. MIMAS PRODUCTION RECORDS

Since 1986, a cumulative tonnage of 240 tHM of MOX fuel rods fabricated following the MIMAS process has been achieved (Fig. 2). During the five last years, thus after the 4-year startup phase of the modernization of the plant in the mid-eighties, the yearly production has varied between 30 and 37 tHM. The level achieved each year depends on various factors, like the number of campaigns (i.e. a production for one customer according to one given technical file), the number of parameters (e.g. plutonium enrichments - typically 3 in PWR designs, but up to 6 in BWR designs) or technical factors linked with the specific requirements of the customers.

The fuel manufactured from 1986 till 1994 has been produced for four countries namely France, Switzerland, Germany and Belgium, as shown on Fig. 2.

2.4. FIELDS OF APPLICATION OF THE FABRICATION PROCESS

From 1986 till end of 1994, BN produced commercial MIMAS MOX reloads as well as experimental test fuel, both for PWRs and BWRs. A broad range of applications, as far as important parameters like the UO_2 type, Pu and Am content, fissile Pu content in the MOX fuel or diameter of the fuel rod are concerned, has been demonstrated both on fabrication and irradiation behaviour points of view.

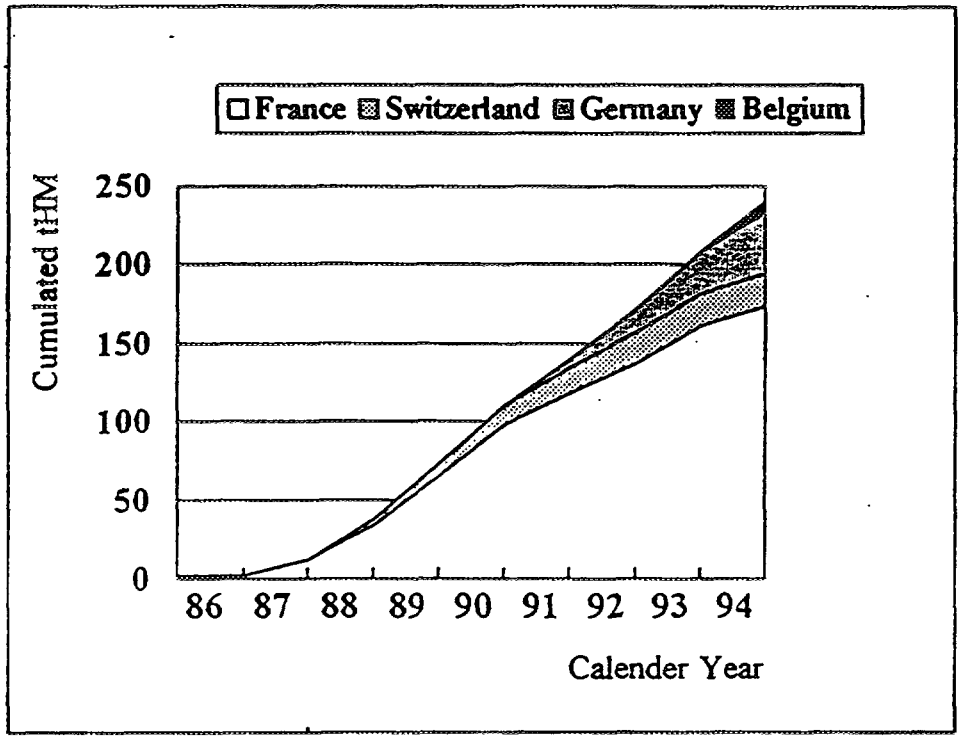


FIG. 2. Cumulated MIMAS Production at BELGONUCLEAIRE

Table I gives the levels of a series of such parameters which have been reached so far with MIMAS fuel in commercial fabrications.

The BN past experience (FBRs and LWRs fuels fabrication) on the former direct blending process shows that this range of applications can be extended (e.g. to higher Pu or Am contents). However, this still needs to be demonstrated for the present process, taking into account the various operational constraints regarding e.g. radiation doses to the personnel. These aspects are covered in chapter 4.

2.5. FUEL FABRICATION COST CONSIDERATIONS

The factors described hereabove - affecting the plant production - have a direct impact on the cost of the product. Indeed, the yearly plant costs are almost completely fixed costs like e.g. personnel and investment. Only a small fraction related to materials or waste treatments are influenced by the production rate. Consequently, the production cost of a given quantity of MOX fuel is almost equal to the cost corresponding to the time required to produce this quantity. Each day during which no fuel pellets or fuel rods are produced (e.g. because the pellets Pu content is changed, which requires a cleaning of the line) penalizes the cost of the MOX unit quantity. Moreover, although the product quality has to remain at the same level as the UO_2 quality, the specifications must be adapted to the specific technology linked with plutonium handling. If this is not the case, a reduction of the fuel pellets/rods production rate is expected, leading to large penalties on the production cost.

Reports (e.g. [13]) have outlined the technical and economical feasibility of the MOX fuel in LWRs.i

TABLE I MIMAS FABRICATION: FIELDS OF APPLICATION
Status as of 31.12.94.

* Cumulated MOX tonnage	240 tHM
* Fuel rod types	14 x 14,16 x 16,17 x 17 (PWR), 9 x 9 (BWR)
* MOX fuel designers	FRAGEMA, SIEMENS, BN
* 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 Pu tot. + U)	2 to 8 %
* Am content (over Pu tot.)	up to17,000 ppm
* Fissile Pu content over Pu tot.	66 to 75 %
* Pu tot. content in primary blend (over U + Pu tot.)	20 to 40 %
* Pu 238 content (over Pu tot.)	up to 1.7 %
* UO_2 material	free-flowing AUC and TU2

3. BELGONUCLEAIRE MIMAS MOX FUEL IRRADIATION EXPERIENCE

3.1. RELATIONSHIP BETWEEN FABRICATION PROCESS AND EXPECTED IRRADIATION BEHAVIOUR

The MIMAS process produces a MOX pellet made of master blend of typically 30% PuO_2 distributed in the UO_2 matrix in agglomerates of a size which is typically 30 to 100 micron homogeneously dispersed in the pellet.

The MIMAS MOX fuel can thus not be considered as a "new compound" as if it were a complete solid solution of UO_2 and PuO_2. Its behaviour is very close to that of UO_2, regarding thermal and mechanical aspects. Only minor differences might be pointed out, which have to be accounted for in the MOX fuel design:

* the neutronic behaviour of MOX differs from UO_2 in that way that the former has a larger flux depression in the pellet and, for same fuel burnups, an higher linear power at end-of-life, leading to differences in the fuel central temperatures and hence in fission gas release. These aspects are now taken into account in the fuel rod design and core design analyses;

* particular attention is still devoted to chemical properties of MOX fuel, as a result of its potential for chemical reduction and thus oxygen release;

* the irradiation-induced properties such as densification and swelling, as well as thermal conductivity, are controlled by the dominant phase in the pellet, namely UO_2 in the MIMAS fuel, and by the structural features resulting from the fabrication route. The knowledge of the latter is hence of great importance for interpreting the fuel behaviour.

We shall refer hereunder to three particular publications which have reviewed postirradiation results on MIMAS fuels, both for PWR [4,5] and for BWR [4, 6] applications.

3.2. IRRADIATION EXPERIENCE

By end of 1994, about 130,000 MIMAS fuel rods have been manufactured at BN, out of which more than 90% are now under commercial irradiation or have already been definitively unloaded. In addition, about 50 fuel rods or segments (Table II) have been manufactured by BN for experimental irradiation purposes.

Fig. 3 shows the present statistics of commercial MIMAS fuel irradiation in terms of fuel assemblies versus burnup. It shows that a burnup of about 50 GWd/tHM (peak pellet) has already been achieved by MIMAS fuel in commercial reactors (BEZNAU-1 and GRAVELINES 4 PWRs).

There are two major sources of postirradiation results (surveillance or experimental programmes) on BN's MIMAS fuel today:

* the French (EDF, FRAMATOME, CEA) programme on MIMAS fuel irradiated in the ST.LAURENT B1 + B2 and GRAVELINES 4 reactors (EDF 900 MWe) [5];

* BN's international programmes which are co-sponsored by several other organizations (utilities, fuel vendors, laboratories) from Europe and Japan, requiring such new data base : these programmes are "PRIMO" for PWR 17x17 fuel from BR3 [7], "FIGARO" and "NOK-MOX" for PWR 14x14 fuel from BEZNAU-1 [8] and "DOMO" for BWR 8x8 fuel from DODEWAARD [6].

The main published conclusions of those programmes are summarized hereunder.

TABLE II MIMAS FUEL - *Experimental Programmes* -

FUEL TYPE	REACTOR	BURNUP GWd/tHM	STATUS	PROGRAMME
17 x 17 rods	BR3	41	Examination completed	PRIMO
17 x 17 rods	BR3 & BR2	60	Under examination	Bilateral
14 x 14 rods	BEZNAU-1	50	Under examination	FIGARO, NOK MOX
14 x 14 segments	BEZNAU-1	50	Extension of irradiation under review	GEMINI (not started yet)
8 x 8 segments	DODEWAARD	58	Under examination	DOMO

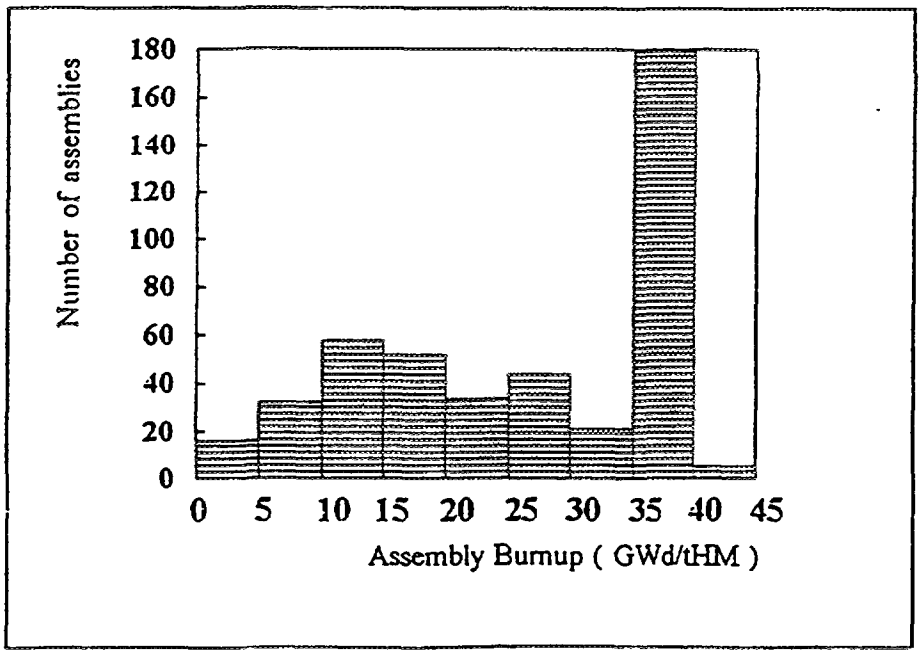


FIG. 3. MIMAS Commercial Irradiation Experience

3.3. TYPICAL RESULTS OF THE MIMAS IRRADIATION BEHAVIOUR

* *Failure frequency*

Out of all the MIMAS fuel irradiated in commercial reactors in France (EDF NPPs), Switzerland (BEZNAU) or Germany (PHILIPPSBURG, BROKDORF, UNTERWESER, GRAFENRHEINFELD) or under experimental conditions in The Netherlands (DODEWAARD) or Belgium (BR3), no failed MOX assembly could be detected, except one in the BEZNAU-1 core : two failed rods were identified in this assembly and the investigations performed indicated that these failures were attributable to fretting by debris carried out by the coolant.

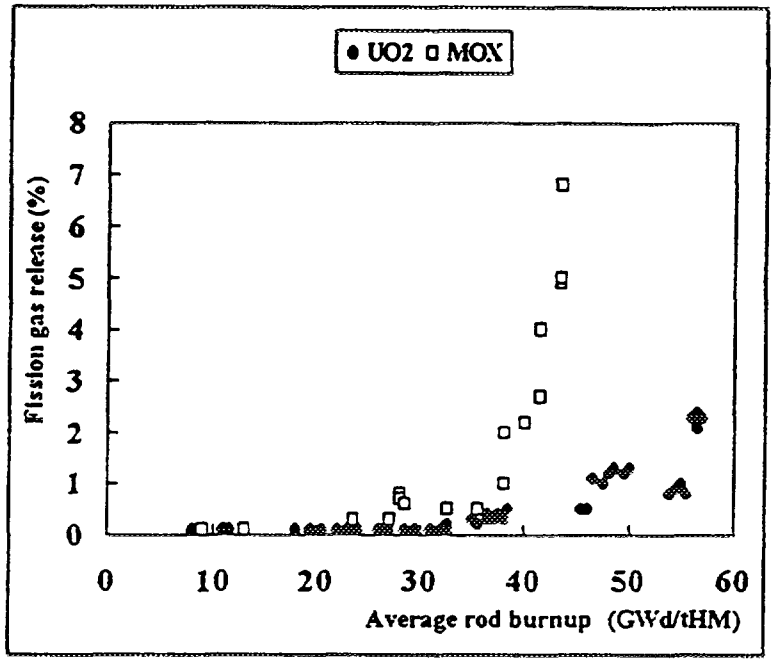
* *Fuel thermo-mechanical properties*

Most of all examinations (fuel rod length changes, diameter profiles, densification, ...) performed recently on MIMAS fuel confirm our previous statement [3] that the mechanical and thermal behaviour of MIMAS fuel is very similar to that of UO_2 fuel. Differences are of the same order as between two kinds of UO_2 powders (AUC and ADU or IDR).

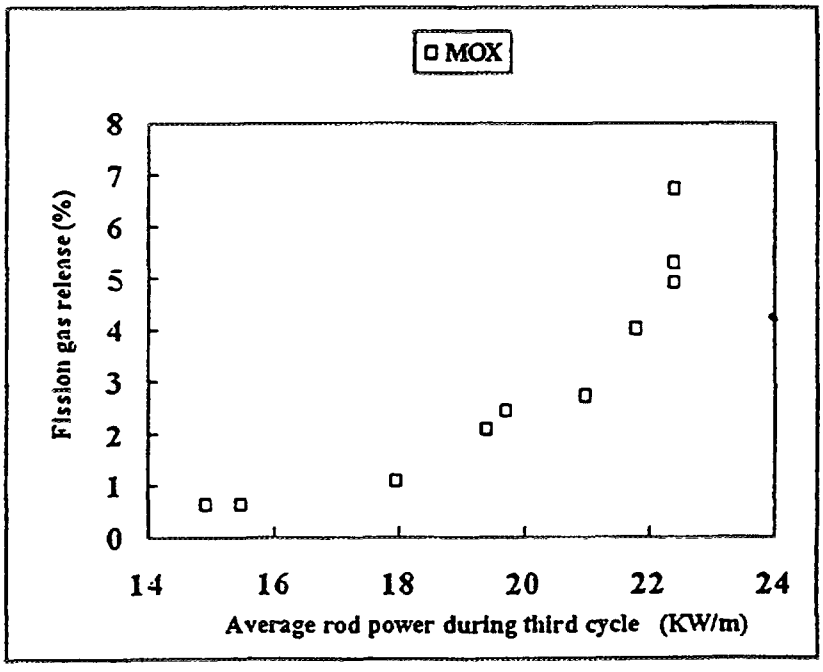
Because it has been the subject to some controversy in the literature, let us emphasize one important fuel behaviour, namely fission gas release which was generally reported to be higher in MOX than in UO_2 fuel.

* *Data on 17×17 MIMAS fuel rods irradiated at low power :*

Figs. 4-a and 4-b [5] show a strong increase of FGR of MOX fuel with burnup (4-a), explained (4-b) by the influence of the linear power achieved during the third irradiation cycle. In comparison, the 17×17 UO_2 fuel FGR does not exceed 2.5% (Fig. 4-a) even at high burnup (up to 60 GWd/tHM), mainly due to the typically low powers reached in UO_2 17x17 PWR assemblies during the last irradiation cycles. A similar trend has been deduced from results obtained on BR 3 fuel rods.



(a)



(b)

FIG. 4. PWR 17 x 17 Fission Gas Release of MIMAS Fuel

* *Data on 8x8 MIMAS fuel segments at high power:*

Fig. 5 [6] compares FGR results on MIMAS and UO_2 fuel segments irradiated at powers in the range 30-40 kW/m up to about 60 GWd/tHM. No difference between the two types of fuel can be found, if one takes into account that the UO_2 fuel power levels are 5 to 10% lower than in the MOX fuel segments.

The mechanical resistance of MIMAS fuel rods is good, as has been proven by power ramps performed in the BR2 (MTR -Mol, Belgium), OSIRIS (MTR-Saclay, France) and in R2 (MTR-Studsvik, Sweden), at peak power levels of up to 48 kW/m [7, 14] for PWR fuel rods and staircase power ramps up to 60 kW/m on BWR fuel segments [6]. No failure has been observed, confirming the excellent results obtained previously on other MOX fuel types [9].

* *Further examination programmes are underway or planned, with following objectives :*

  * determination of central fuel temperature and fuel rod internal pressure versus linear power for high burnup rods (50 GWd/tHM peak burnup) pre-irradiated at NOK-BEZNAU 1 (FIGARO International Programme [8]) ;
  * surveillance programme (non-destructive examination and punctures) on similar rods on a statistical basis for licensing applications (NOK MOX Programme [8]);
  * PCI behaviour under fast ramp tests on segmented fuel irradiated in BEZNAU-1 at high burnup (up to 55 GWd/tHM) : planned GEMINI International Programme [8];
  * measurements on particular material properties like diffusivity, heat capacity or melting temperatures of high burnup BWR and PWR MIMAS fuels.

The results are useful to increase the MOX data base to a level which allows a proper design of MOX fuel as for UO_2 fuel and to avoid penalizing the former in terms of burnup or specific operation modes.

4. THE MOX FABRICATION: BEYOND 2000

4.1. TARGETS AND IMPROVEMENTS TO BE ACHIEVED

In BN's MOX business, the future (let us say the 1995-2005 decade) shall be characterized by the following evolution:

* an increased share of BWR (8x8 or 9x9) fuels and thus a need for even more flexibility requested from our production;
* a trend to increasing the Pu content in the product, for two reasons:
  . a request towards high burnups ;
  . a tendency of fissile Pu content decrease in the powder received from the reprocessors;
* a trend to receiving Pu with higher activity, with an effect on the radiation doses and on the thermal heat dissipated by the powder, both having a direct effect on production efficiency;
* the ICRP-60 recommendations.

To meet the main new constraints imposed by this evolution, adaptation of the MOX plant is necessary in the following directions:

* the decrease of the fissile Pu content and the higher Pu enrichment go along with a more important heat generation in the powders, requiring modification of the powder blending and transfer system ;

* a significant increase is foreseen of the neutron emission by the even isotopes of plutonium (Pu 238, 240 and 242) : this is leading to the massive introduction of neutron shielding in the workshops;

* personal exposure is to be reduced also by remote process control and by mechanization of different process steps.

For the long term perspectives, additional improvements shall probably be requested, depending too on the customer's particular requirements.

Finally, the MOX production experience shall also be improved by permanent analysis and feed-back of the fabrication data and by implementing the necessary qualification programmes for example to demonstrate the fabrication of new products (e.g. 8x8 pellet geometry or new UO_2 powder types).

The trends to be expected after the year 2000 for the main parameters influencing the production are shown on Fig. 6, compared to the present situation.

4.2. POSITIONING OF BN IN THE MARKET

Other papers [10, 11] have analysed the future prospects regarding Pu availability and MOX fabrication market. Their conclusion clearly states that the MOX fabrication demand will stay high for the next ten years at least, despite the expected startup of two large plants, one in France (MELOX) and one in the United Kingdom (SMP).

In this period, the BN plant will keep its position in the market, thanks to its expertise and flexibility : a large part of the plant capacity over ten years is already committed. On Fig. 7 we have plotted - versus time - the expectations (to our best present knowledge) of the quantities of PWR compared to BWR fuels that will have to be manufactured by BN. The PWRs involved are located in Belgium, France, Germany and Switzerland, whereas the BWRs are in Germany and (probably) Japan.

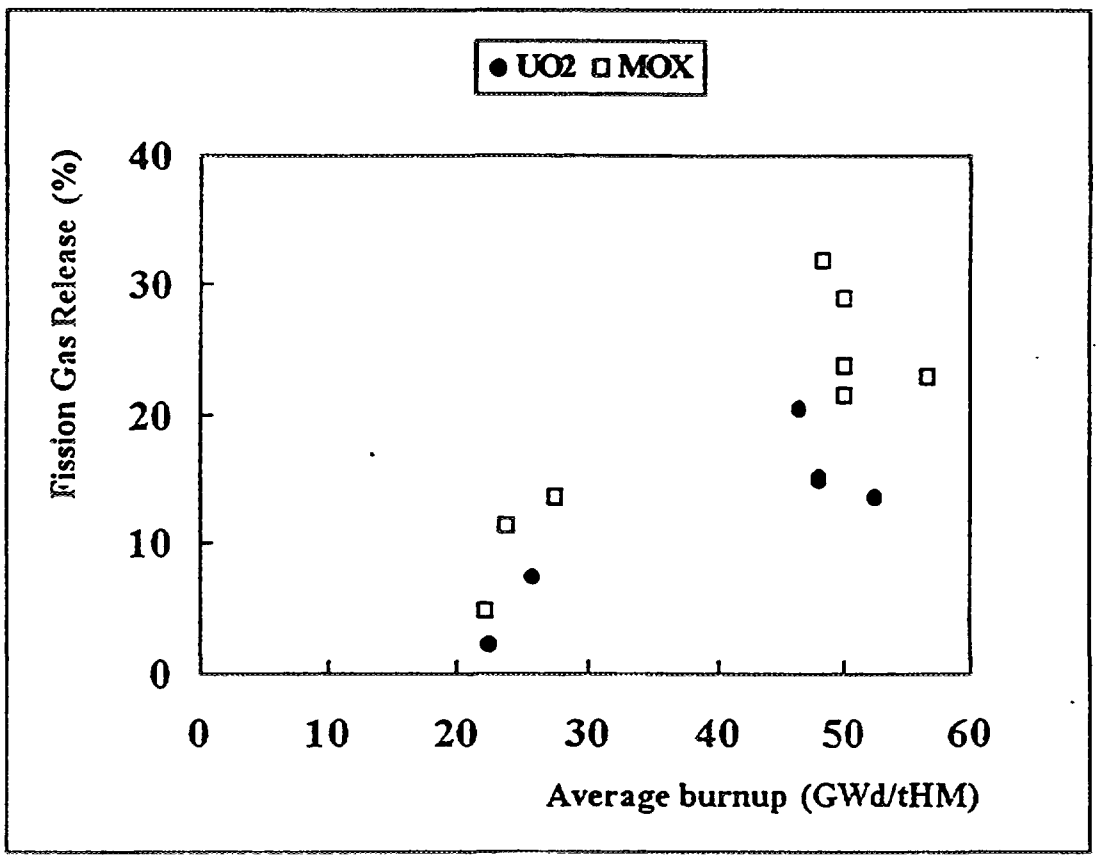


FIG. 5. BWR 8 x 8 Fission Gas Release of MIMAS Fuel

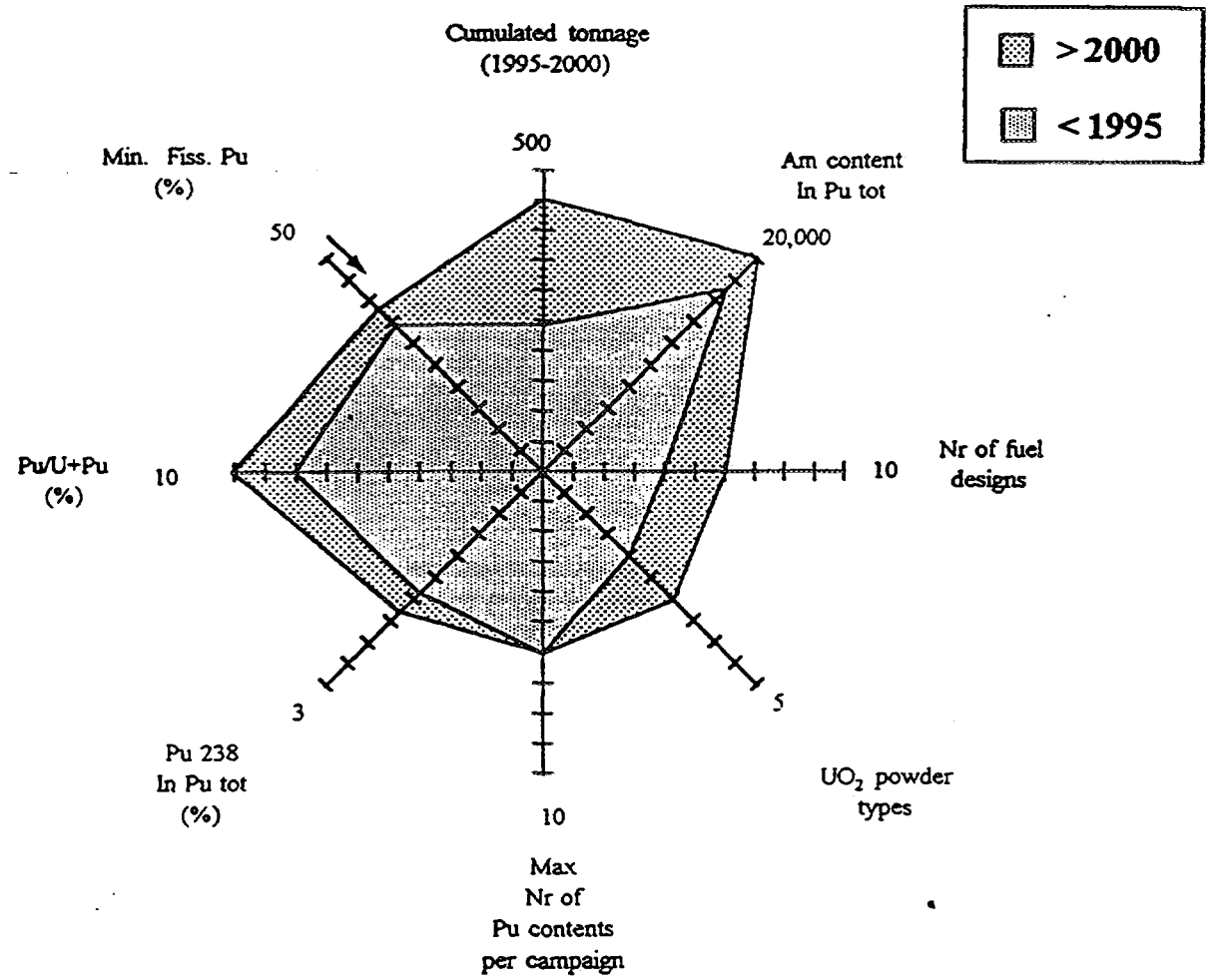


FIG. 6. ***Evolution of the MIMAS Production Field of Experience***

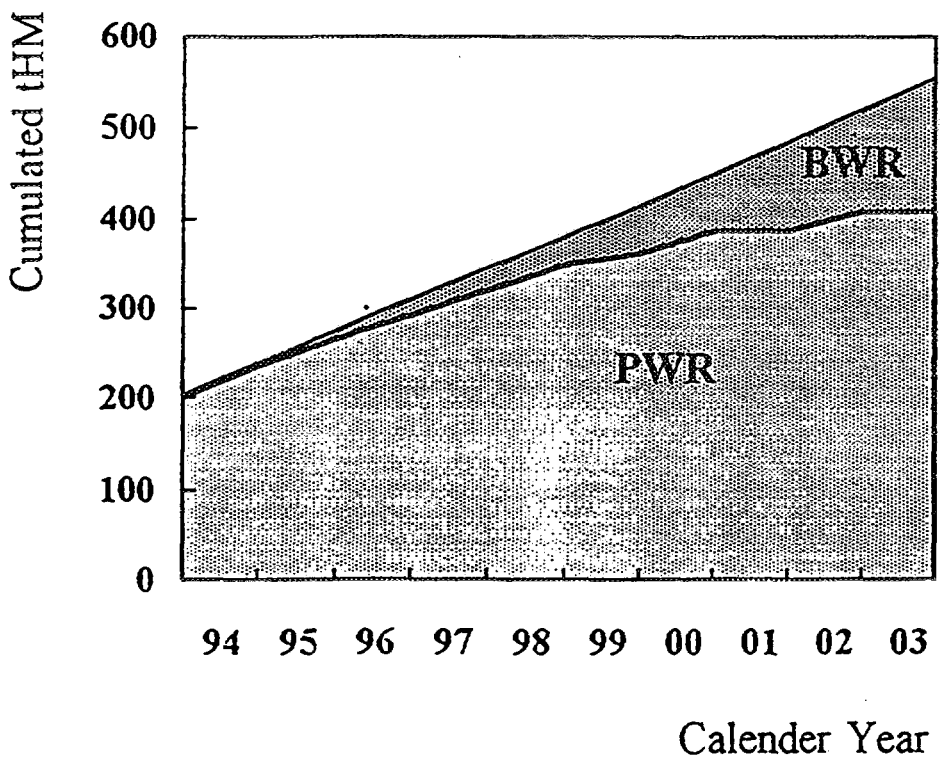


FIG. 7. ***Prospects of MOX Fabrication at BELGONUCLEAIRE***

Beyond 2005, the MOX market and thus BN's position in such market shall depend on the future reprocessing commitments in Europe.

If required, future prospects of the plant can also lay in the transformation into MOX fuel of plutonium originating from nuclear weapons [12]. We see no particular difficulty in using this Pu with high fissile isotopes contents, either in pure form or after mixture with Pu from reprocessing.

5. CONCLUSION

The experience accumulated so far at the BN plant has demonstrated the commercial viability of MOX fabrication. The MIMAS process developed is suited for commercial use considering the particularity of the MOX fabrication technology and the variability of the plutonium as received from reprocessing. The same process has therefore been choosen by COGEMA for the MELOX plant in Marcoule and for CFCa in Cadarache.

The irradiation of MIMAS MOX fuel in commercial reactors as well as experimental tests have also proven its quality and performance (no failure and an ability to behave similarly to UO_2). Due to the neutronic particularities of plutonium and also to minor differences in its other properties, the design has to be adapted and an increased experimental data base is still required. This is being undertaken through international programmes organized by BN on PWR and BWR fuels, and by extensive examinations performed by the French industry on 17x17 PWR fuel.

Finally, the future prospects have been reported. To answer such new requests, the necessary changes are being implemented at the Dessel plant.

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PLUTONIUM RECYCLING AND THE MELOX FABRICATION PLANT

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Abstract

The French decision to turn to industrial plutonium recycling through MOX fuel was taken relatively recently, by the mid-1980s, when recycling of valuable materials emerged as a satisfactory back-end solution for the fuel cycle. The French strategy currently relies on an industrial structure comprising the COGEMA reprocessing plants at La Hague with a total capacity of over 1600 tHM, the existing MOX fabrication facilities at Dessel (BELGONUCLEAIRE PO Plant) and Cadarache (COGEMA Cadarache Plant) with a capacity reaching 65 tHM. In order to implement the French program of recycling plutonium through MOX fuel in 20 to 28 PWR reactors, and to serve foreign utilities as well, it was decided to create the Melox Company (50% Cogema, 50% Framatome), the goal of which is the construction and the operation of the Melox plant. This paper describes the main features of Melox design and presents current status of starting-up activities.

An innovative plant based on well-tried techniques

Several requirements are to be dealt with in order to cope with:

- fuel designer requirements related with in-core behaviour. These include homogeneity of the mixed powders and of their isotopic content, pellet density mastering and stringent dimensional tolerances
- reprocessor requirements concerning pellet solubility.

Moreover, as a result of final customer requirements with regard to new trends in fuel management and flexibility of MOX fuel use, Melox will be able to handle a wide range of basic nuclear materials; aged plutonium and/or coming from high burnup fuel, high plutonium MOX fuel content and uranium being either depleted or coming from reprocessing.

The reference process for the Melox plant is the well mastered MIMAS process which resulted from several steps of process improvements at the Dessel plant since 1984. In this process, the PuO_2 powder is first micronized with a part of the UO_2 powder to form a primary blend of 30% plutonium content. This primary blend is then mechanically diluted by mixing with free flowing UO_2 powder to obtain the specified content of the MOX fuel. These two steps significantly reduce the plutonium-rich spots in the UO_2 matrix.

While coping with the above requirements, special design efforts have been made to reduce foreseeable occupational exposure to a tenth of the current regulatory limit. This implies almost total automation of production and requires extensive optimization studies tailored to each workstation.

Meanwhile, waste minimization has been another major design objective: compared to the factories of the previous generation, a significant decrease of residual plutonium in the waste should be achieved, mainly due to the operation of a large on-site TRU waste incinerator.

A step-by-step commissioning programme

Inactive testing started in early 1993 and provided gradually increasing confidence concerning satisfactory operation of the whole production line, as well as of all its parts. At the end of inactive tests,

special overall tests were performed in order to verify global behaviour in case of general defaults such as lack of auxiliary fluids or power supply.

Active tests were aimed at providing the customers with complete demonstration that fabricated products are of quality and reliably cope with the above described requirements. Before normal operation is reached, the commissioning procedure included functional testing with UO_2 parametrial tests, qualification and validation of processes and generic demonstration programmes with real products.

For cladding, rod inspection and assembly units, commissioning started from the back-end of the production line. This enabled the production of assemblies, using rods coming from the Cogema Cadarache plant, very soon after a general active operation approval by the Regulatory Authorities in August 1994.

The first PuO_2 powder box was opened in mid-March, which started the entire powder and pellet units operation according to the normal process pathway.

Conclusion

The Melox plant can be seen as an outstanding achievement, taking full benefit from previous experience, while introducing several innovative features in terms of radiation protection as well as environmental concerns. It is a major input in the national energy policy, as an important link of the plutonium recycling strategy.

USE OF VIBRO COMPACTED MOX FUEL IN THERMAL REACTORS

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Abstract

The main business drivers for the next generation of reprocessing and fabrication plants will be cost reduction and diversion resistance. Integration of reprocessing plant with refabrication plant provides the scope to reduce powder handling stages prior to pelleting, e.g. by utilising ammonium carbonate co-precipitation or thermal de-nitration to produce a U/Pu Masterblend for pelleting. The pelleting stage must be eliminated to achieve greater cost reduction, as this will also eliminate the requirement for grinding and pellet inspection and significantly reduce reject rates. Gel spheres have been vibro compacted into fuel pins and tested in fast reactors. Although this type of fuel was limited in rating and burn-up compared to annular pelleted fuel, the performance achieved far exceeds any requirements for thermal reactors. Co-finishing routes like gel sphere precipitation offer a measure of diversion resistance in that a pure plutonium stream need never be isolated. The lower smear density of this fuel has a similar effect to annular fuel in reducing the relative amount of ^{238}U available for breeding and so increases the capacity for burning plutonium. Gel sphere routes, being essentially wet and dust free, will reduce dose and, being suitable for remote operation, lend themselves to finishing and fabrication of streams from reprocessing flowsheets offering lower decontamination factors (DFs). Lowering decontamination factors has a negligible effect on dose from fuel and on subsequent reactor performance but allows a substantial reduction in reprocessing costs. Incorporating fission products into fuel also assists in detection and diversion resistance. The acceptance of fuel with residual fission products, and the ability to finish and fabricate it, would provide scope for alternative reprocessing routes, such as pyrochemical methods, previously rejected on the grounds of failing to provide the decontamination attainable in the PUREX process.

1. INTRODUCTION

The UK, France, Japan and Russia have built, or are building, a new generation of large commercial scale reprocessing plants for thermal reactor fuel. All of these plants have, or are planning, associated large commercial scale MOX fabrication plants for utilisation of the extracted plutonium.

Business drivers on the next generation of plant associated with re-cycle will be to reduce costs while further enhancing clean technology, and improving resistance to diversion.

The desire to significantly reduce costs for the fuel cycle as a whole leads us to consider reducing the number of purification stages and relaxing the decontamination requirements of the reprocessing plant. The product stream from such a reprocessing plant is likely to require remote operation to limit dose to operators. Wet finishing routes, such as gel sphere precipitation, avoid dusty and dose intensive operations and are suitable for remote operation. Experience of such routes has been gained in the fast reactor field where the product has been used in the vibro compacted mode.

Improvement in diversion resistance could result in more widespread acceptance of reprocessing options and an increase in business opportunities. The approach in the US to diversion resistance is to leave some fission products in the product and not to separate out a plutonium stream. Such considerations also lead to the adoption of a remote finishing route which avoids dust and dose.

This paper will show that:

- ñ there is considerable experience of the use of vibro MOX in fast reactors,
- ñ vibro MOX for thermal reactors may well satisfy the requirements set by the business drivers for new fuel cycle processes
- ñ there are aspects of vibro MOX fuel which require further consideration if it is to be a serious contender in the fuel cycle for thermal reactors.

2. METHODS OF COST REDUCTION

2.1 INTEGRATION OF FINISHING AND FABRICATION

Integration of finishing into fabrication, eg by producing a mixed U/Pu stream directly by a thermal de-nitration route, would have the potential to reduce combined finishing and fabrication costs due to elimination of some of the milling and conditioning stages and associated equipment. Additional reductions in cost could be obtained by eliminating conventional pellet pressing, eg by adopting vibro compacted granules as pin fill for thermal MOX fuel. The use of vibro fuel also eliminates the requirement for pellet grinding and inspection to meet specifications and hence reduces reject rates and associated costs.

2.2 RELAXED DECONTAMINATION REQUIREMENTS

Specifications for MOX fuels are adapted from those for uranium fuels, which encapsulate the purity obtained in the processing of virgin uranium. From a reactor performance point of view, such tight specifications may well be unnecessary. Calculations show that residual fission products will not have a marked effect on reactor performance in LWRs. Indeed, fuel for reprocessing was still functioning, albeit at reduced output, on the day it was discharged from reactor containing its full fission product inventory. If fission products were decontaminated by a factor of only 100 prior to recycle, rather than by up to 108 under present schemes, then the remaining fission products would be equivalent to those present after only a few days irradiation. The residual impurities in the recycled fuel would reduce attainable burn-up by much less than 1 GWd/t due to neutron absorbers. The effect of residual neutron absorbers can be compensated in MOX at no cost penalty by very slight over-enrichment.

Reducing decontamination factors (DFs) from ca 10^8 to 10^2 allows major reductions in the number of stages and hence cost in the chemical separation area of reprocessing plants. Single cycle flowsheets could easily attain these specifications. Acceptance of lower DFs for thermal fuel would also allow the use of alternative separation technology to Purex, eg molten salts. Reprocessing fast reactor fuel using molten salts to produce fuel with relatively low DF is in use at pilot scale in Russia and has been demonstrated for the Integral Fast Reactor concept (for metallic fuel) in the USA [l].

Finishing and fabrication processes for fuel with relaxed specification should be engineered to be dust free where practical and be maintenance free where possible to avoid penalties in plant and maintenance arising due to higher radiation. Gel sphere production, being wet routes, are essentially dust free, involve no mechanical stages like milling and pressing, generate no dust from pellet grinding, and are suitable for remote operation. Vibro fuel derived from the products of molten salt reprocessing does produce dust but the operation is suitable for remote pin filling and this can be done in cells behind shielding. Dose from fuel in pins is mostly due to neutron radiation, which is inherent in the fissile content of the fuel, and so will not increase markedly with lower DF. Hence, from a generating utilities point of view, dose levels from 'impure' fuel will be similar to those from conventional fuel once inside the pin.

3. METHODS OF IMPROVING DIVERSION RESISTANCE

When a gel sphere route is used it is not necessary to separate a plutonium stream to produce fuel, as a mixed nitrate stream can be blended down to the desired enrichment with uranyl nitrate and can be converted directly to the finished fuel in a single process line. Products with a relaxed specification are easier to detect and offer a degree of diversion resistance due to the increased handling difficulties compared to separated plutonium. Significant wet processing would be necessary to separate out plutonium from any such fuels if it were to be diverted.

An added advantage of vibro packed fuel is a reduction of 20% in the fertile uranium in the pins and this reduces the bred in plutonium by a similar proportion.

Hence, it would appear that there are many advantages in terms of the business drivers in using the vibro fuel routes in the thermal reactor cycle.

4. PAST EXPERIENCE - THE USE OF VIBRO FUEL IN FAST BREEDER REACTORS

4.1 FABRICATION EXPERIENCE

Initially, vibro fuels for fast reactors were produced from dry routes, which utilised granules from crushing high density sintered pellets. The design of vibro fuel was aimed to achieve the same smeared density (averaged over the pin volume) as annular pelleted fuel at 83 % of theoretical density (TD). To achieve this, a three size component, high energy vibration technique was adopted. Smeared densities of 82 to 84 % were regularly achieved using this method, but the high energy compaction technique was slow and limited production rates. To overcome these problems, the target smear density was reduced to 80 %, which allowed two component packing and an infiltration technique to be adopted. A significant improvement in granule manufacture was achieved by omitting the pelleting stage and modifying operation of the Z-blade mixer in the binder route to produce granules in the required size ranges [2].

With a view to simplifying production and reducing potential dose in commercial scale fabrication plants, as well as integrating fabrication into Purex reprocessing, many countries developed variations in gel routes for production of oxide fuels. These gel routes involve mixing the heavy metal nitrate solutions with an organic gelling agent and producing small drops. The organic agent acted as a polymer support for the precipitate and hence an alternative name is polymer supported co-precipitation.

The UK carried out extensive development of an external wet route which is shown in outline in Figure 1.

The wet routes offered a means of producing granules directly from mixed uranium and plutonium nitrate solution. The aqueous feed solution contained uranyl and plutonium nitrate, an organic gelling agent, and a modifying agent that was used to stabilise the feed. Droplets were generated by a vibrating jet, with sphere size being controlled by adjusting the viscosity of the solution. Gelation was carried out in a column of ammonia, and the spheres were washed in a column of water. Drying rates were controlled to avoid cracking during shrinkage. Removal of the organic gelling agent was carried out in a de-bonding stage under carbon dioxide. Over 500 PFR fuel pins were fabricated using the MOX micro-spheres described above.

In 1969, the USSR and the GDR concluded a contract for scientific co-operation in the design, construction, and operation of an automated pilot plant for the re-fabrication of fuel elements in a soviet experimental BOR 60 fast reactor. The plant was based on molten salt reprocessing followed by re-fabrication of vibro fuel rods in a single facility. Following on from successful tests with urania fuel, over 11 000 fuel rods were manufactured and put into the BOR 60 reactor. The mixed oxide for the fuel column was densified using electrodynamic vibration. For the non-spherical granules produced from the molten salt reprocessing, 3 or 4 size ranges were used. Initially, pure granules of plutonia with a particle

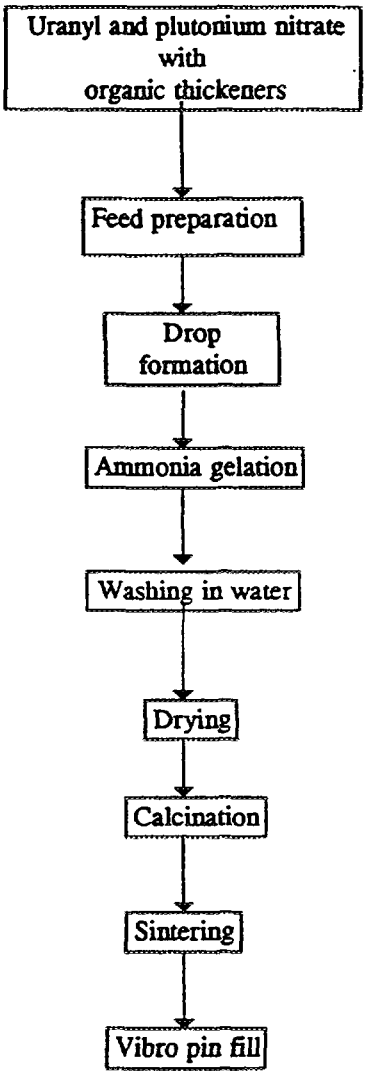

FIGURE 1. A GENERALISED FLOWSHEET FOR VIBRO FUEL FROM GEL SPHERE PRECIPITATION

size range between 25 - 100 um, also produced from the molten salt process, were used. Both the urania and plutonia were close to theoretical density (98%) and had sufficiently good flow properties for vibro-compaction without further conditioning. Experience with vibro-compaction has shown that, in this process, fuel in 'poly-disperse' form is suitable for attainment of a sufficiently high density [3].

4 .2 IRRADIATION EXPERIENCE

Irradiation of 700 non-gel vibro oxide pins in the Dounreay Fast Reactor demonstrated that the performance of vibro fuelled oxide pins, at the linear ratings (nominally 450 W/cm) and target burn-up (7.5% peak burn-up) set for the Prototype Fast Reactor, was comparable to that of annular pelleted pins. Fuel form was not found to be a parameter affecting pin endurance under these conditions and there was confidence that burn-ups of approaching 10 % could be reliably achieved. Several pins were taken to 14 % burn-up and a peak of 21.6 % was achieved.

2 000 non-gel vibro oxide pins were irradiated in the Prototype Fast Reactor at ratings below 450 W/cm (17 failures) and 454 were irradiated at over 600 W/cm (7 failures). 500 gel vibro oxide pins were irradiated in PFR at reference rating (< 450 W/cm), of which there was one confirmed failure at 10.3 % burn-up.

It was recognised that the adoption of pin designs with vibro fuel with linear ratings in excess of 500 W/cm might pose problems associated with fuel temperature and hence fuel column stability and margin to melt. There was a programme of modelling studies and MTR irradiations involving 30 pins, focusing in particular on the thermal behaviour and power-to-melt of vibro compacted fuel. The DFR programme explored high linear rating performance and revealed the onset of a severe form of cladding corrosion not encountered at lower ratings.

The use of vibro compacted fuel of the type used in the UK fast reactors imposed an upper limit on allowable fuel linear ratings of around 450 W/cm, and even when this limit was respected, the reliability of the pins above 10 % burn-up was questionable. These aspects of behaviour, in comparison with the performance of annular pelleted fuel, effectively excluded the use of vibro fuel from the European Fast Reactor concept.

More than 300 vibro compacted MOX fuel assemblies, each containing 37 rods, have been tested in the BOR 60 reactor since 1981 to achieve significant burn-ups at normal power ratings. These assemblies have included mechanical mixtures of urania and plutonia, as well as homogenised fuel. Up to 10% uranium metal has been added to these assemblies as a getter which, it is claimed, practically eliminates the burn-up restriction connected with cladding corrosion [4]. Irradiations have also been carried out in the BN 350 and BN 600 reactors. Higher packing densities, achieved using multi-component fill, can be used to decrease fuel temperature for a given linear rating by reducing void space and increasing thermal conductivity.

5. ASPECTS OF VIBRO MOX FUEL TO BE CONSIDERED IN THEIR APPLICATION TO THERMAL REACTORS

It is apparent from the above that the use of vibro MOX fuel in thermal reactors could satisfy the requirements of the key business drivers set for future fuel cycle facilities. There is considerable experience in the use of vibro MOX in fast reactors, although it is recognised that there are a number of differences between the two reactor systems which could well influence the performance of the fuel. The impact of some of these on the fuel cycle is raised below.

5. 1 FABRICATION

Current thermal reactor fuel pin designs are longer and fatter than fast reactor pins. While the fatter pins will ease filling, the length of the pins will require significant head room for gravity filling. It will also present problems in ensuring adequate homogeneity with in-fill of other sized fractions over a large length.

Zirconium cladding for thermal reactors will be more tolerant to residual chloride from molten salt reprocessing than stainless steel pins in fast reactor, although stainless steel is considered as a possibility for very high burn up thermal reactor applications.

5.2 IRRADIATION AND FUEL PERFORMANCE

The low smeared density of vibro-packed fuel reduces ^{238}U resonance absorptions and increases the lifetime reactivity of fuel for a given fissile content. There is a corresponding penalty on radial peaking, because the reactivity swing with burn-up is increased. This should not be a major concern for MOX fuel, however, because the reactivity with burn-up gradient for MOX is considerably less than that of UO_2 fuel. The higher reactivity swing is caused by the lower in-breeding of fresh ^{239}Pu and the higher burn-out of the plutonium loaded initially, which might be seen as an advantage from a proliferation standpoint.

Limitations on burn-up from thermal creep and pellet-clad mechanical interaction would be expected to be less from vibro fuel compared to pelleted fuel in a thermal flux.

Higher mass ratings, in terms of W/g, are required in vibro fuels to attain a given linear rating. Vibro fuel would run hotter than conventional pelleted fuel due to the higher rating and additional voidage. Higher temperatures will lead to higher fission gas release which is exacerbated by the higher free surface area for release into the pin voidage. These differences should not have a marked effect on fission gas release at the low temperatures in thermal reactors. Consideration has to be given, however, to the effects of accident induced fast transients.

Note that the target ratings and burn up for thermal reactor fuel pins are well below those experienced in fast reactors.

5.3 REPROCESSING

All the vibro MOX from the DFR and PFR irradiations has been reprocessed without problems.

Vibro fuel re-structures when irradiated in fast reactors but is not expected to do so at the lower ratings in thermal reactors which result in much lower fuel temperatures. The higher free surface area would be expected to increase dissolution rates on reprocessing. The use of vibro fuel would be expected to allow the removal of fuel from cladding prior to reprocessing, which is not presently possible with pelleted fuel. Molten salt processes are in use at present to reprocess vibro fuel.

6. CONCLUSIONS

The business drivers set for the next generation of fuel recycle plant require reductions in cost and improvements in resistance to diversion. This paper shows that these can be achieved by a combination of integration of fabrication and finishing routes, selection of processes suitable for remote operation, and acceptance of products with a relaxed specification. All of these lead us to consider the use of vibro fuel in thermal reactors.

The fabrication and use of vibro MOX fuel has been demonstrated in fast reactors and, although it is accepted that a number of aspects require further detailed examination, it is suggested that the concept of vibro MOX in thermal reactors is worth further consideration as part of an advanced recycle facility.

ACKNOWLEDGEMENTS

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CHANGES IN UO_2 POWDER PROPERTIES DURING PROCESSING VIA BNFL'S BINDERLESS ROUTE

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Abstract

The Short Binderless Route (SBR) has been developed for Mixed Oxide fuel production in BNFL's MOX Demonstration Facility (MDF) and the Sellafield MOX Plant (SMP). It is a compact process which enables good homogenisation of the Pu/U mixture and production of free flowing press feed material. The equipment used to achieve this consists of an attritor mill to provide homogenisation and a spheroidiser to provide press feed granules. As for other powder processes, the physical properties of the UO_2 powder can affect the different process stages and consequently a study of some of these effects has been carried out. The aims of the work were to gain a better understanding of the process, to consequently optimise press feed material quality and to also maintain powder hold-up levels in the equipment at a minimum. The paper considers the effects of milling processes on powder morphology and powder surface effects, on the granulation process and also on powder and granule bulk properties such as pour, tap and compaction densities. Results are discussed in terms of powder properties such as powder cohesivity, morphology and particle size. UO_2 powder derived from both the Integrated Dry Route (IDR) and the Ammonium Di-Uranate (ADU) Route are considered. Small (1 kg) scale work has been carried out which has been confirmed by larger (25 kg) scale trials. The work shows that IDR powder with differing morphologies and ADU powder can be successfully processed via the SBR route.

1. INTRODUCTION

It is well established that powders derived using different production routes may exhibit different properties and behaviour even though they have the same chemical composition. For example, uranium dioxide powders derived from dry and wet routes can possess different particle sizes, particle morphologies, specific surface areas and have slight differences in chemical composition, hence they can exhibit different behaviour. Additionally each stage in the processing of fuel can change the properties and hence, the resultant powder behaviour. It is important to understand how the fundamental powder properties can affect the different stages in the processing of nuclear fuels such as granulation, pressing and sintering. In this study three UO_2 powders with different morphologies were initially characterised and then further characterised after processing using the Short Binderless Route (SBR) which has been developed for Mixed Oxide fuel production. The data obtained were used to obtain an understanding of the powder behaviour and support the development of the SBR and its application in BNFL's MOX plants. Both Integrated Dry Route (IDR) and Ammonium Diuranate (ADU) derived UO_2 powders were studied.

2. EXPERIMENTAL

Three uranium dioxide powders, which were derived from powder processes used by BNFL Fuel Division at the Springfields site, were studied. The first two materials were prepared by the Integrated Dry Route (IDR) using different kiln conditions to provide one powder of 'plate like' particle morphology (IDR_P) and one of 'sphere like' particle morphology (IDR_S). The third powder was prepared via the Ammonium Diuranate route (ADU) in the BNFL Springfields Enriched Uranium Residues Recovery Plant (EURRP).

Each powder sample was characterised, before and after milling, using the following techniques. Powder pour densities were determined by measuring the volume of a known mass of material (lOOg) in a tared measuring cylinder. Tap densities were determined by tapping the measuring cylinder until the powder volume remained constant and this volume was recorded. For both measurements the bulk density values were calculated by dividing the mass (g) by the volume (cm^3). The Specific Surface Area of each powder was measured using a gas adsorption technique, iron levels were determined by atomic absorption and particle size was determined by Coulter Counter analysis. The powder morphologies were examined at high magnification using a Scanning Electron Microscope (SEM).

In each milling experiment a 1 kg batch of powder, containing 0.1 wt % zinc stearate lubricant, was milled in a 5.7 Litre attritor mill pot with a steel ball charge. Powder batches were milled up to 40 minutes with samples taken for characterisation, using the techniques listed above, after 20 minutes and 40 minutes. Granules were prepared by tumbling the powder with a further 0.1 wt % zinc stearate lubricant. The granules were compacted to pellets with an Apex press using a 10 mm diameter cylindrical steel die. Green pellet densities were calculated from mensuration results prior to sintering pellets at 1750°C for 5 hours in pure Hydrogen. No pore former was added to the material prior to sintering. Pellet sintered densities were measured using a water immersion technique.

3. RESULTS AND DISCUSSION

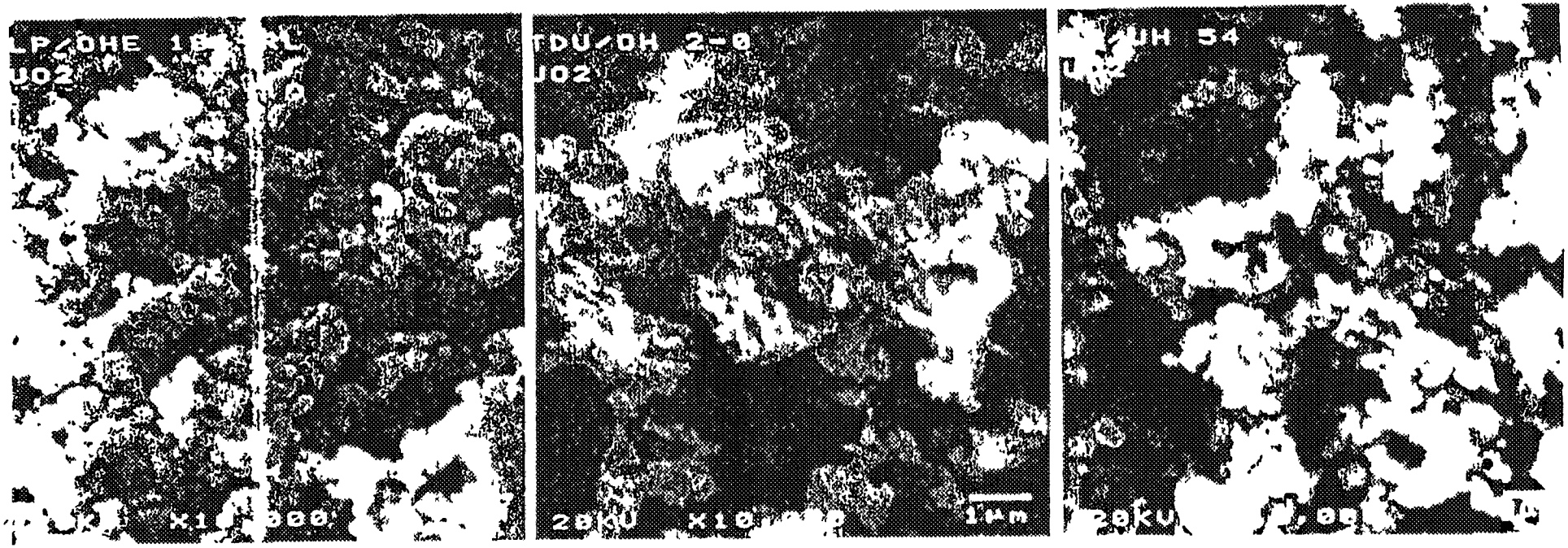
3 .1 EFFECT OF MILLING ON POWDER CHARACTERISTICS

The morphology of each unmilled powder can be seen in Figure 1 a. The IDRs and ADU powders appear similar, each consisting of small (less that 1 μm) rounded particles. The IDR_P powder possessed a definite 'plate-like' morphology with individual particles approximately 1 μm in size.

Table 1 gives results of the Coulter counter analysis results which show that the mean particle sizes for each powder are very similar prior to milling. All of them lie within the range 3.53-3.72 μm. Following milling, in the case of the IDR powders, the particle sizes have apparently increased which is not a very likely occurrence. It could indicate that the newly milled powders have loosely agglomerated during the analysis and given higher results than expected. The SEM micrographs taken after milling and illustrated in Figure lb, indicate a much smaller powder particle size than those recorded by the Coulter Counter analysis. These results could be due to the known tendency of these powders to agglomerate following milling, as demonstrated in spheroidisation.

Table 2 gives results of powder pour and tap densities and granule flow rates. Figures 2 and 3 show pour and tap densities versus milling time. In all cases the powders showed a large increase in pour and tap density following milling for 20 minutes and spheroidisation. In the case of the IDR powders a further slight increase occurred on milling for 40 minutes while the ADU material showed a very slight decrease. The results indicate that extent of milling had an effect on powder and on granule properties. While there was a broad spread in initial pour and tap densities for the three powders there was only a slight difference in the values after milling for 40 minutes.

Pour and tap density is a measure of how powder particles pack together and the properties are affected by parameters such as particle size, shape, and agglomeration. Agglomeration and irregular



IDRs UO_2 IDRp UO_2 ADU UO_2

FIG. 1a SEM Micrographs Unmilled Powders

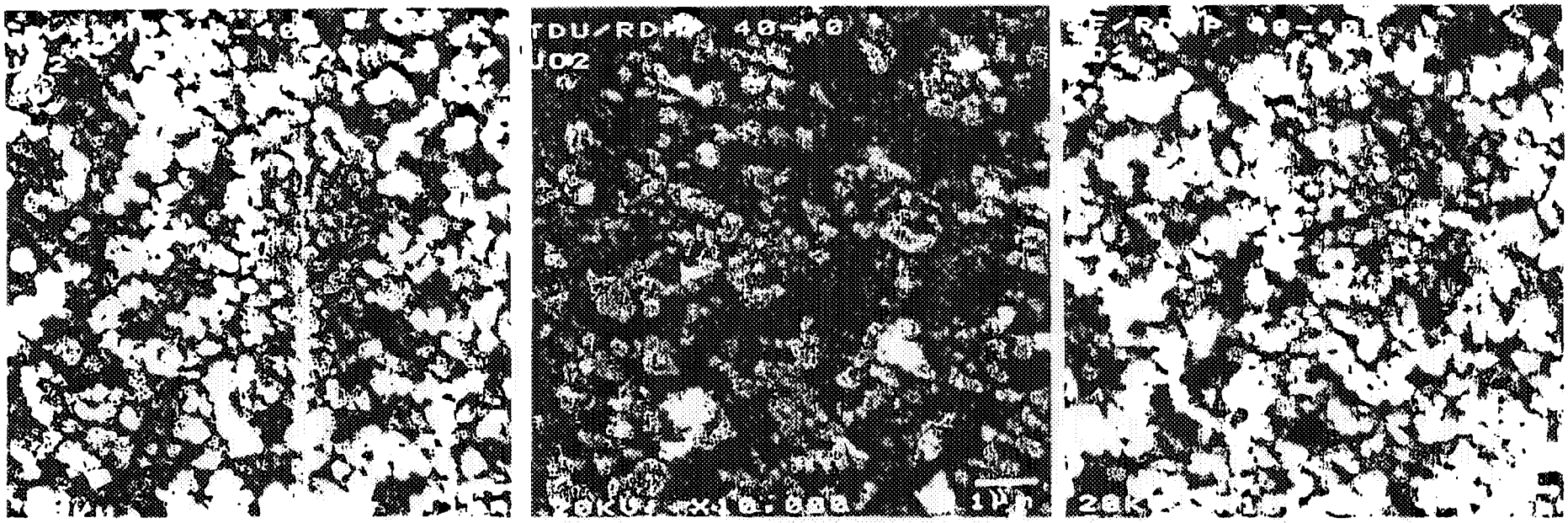


FIG. 1b SEM Micrographs Milled Powders

TABLE 1 Powder Particle Size Analysis by Coulter Counter for Given Milling Time

	Mean Particle Size / μm for given Milling Time / min		
	0	20	40
ADU	3.53	4.29	3.38
IDR_P	3.65	3.90	4.63
IDR_S	3.72	3.63	6.44

TABLE 2 Results of Granulation Trials Pour and Tap Densities and Granule Flow Rate

	Results for given milling time / min								
	0			20			40		
	Pour/ g cm^{-1}	Tap/ g cm^{-1}	Flow/g s^{-1}	Pour/ g cm^{-1}	Tap/ g cm^{-1}	Flow/g s^{-1}	Pour/ g cm^{-1}	Tap/ g cm^{-1}	Flow/g s^{-1}
ADU	1.71	2.43	no flow	3.16	4.04	35.68	3.11	3.71	36.87
IDR_P	0.80	1.39	no flow	3.08	3.66	38.51	3.33	3.88	35.87
IDR_S	1.60	2.03	23.08*	3.11	3.57	38.83	3.22	3.82	41.08

* Flow occurred in only one of three trials

particle shape are detrimental to achieving high packing densities and can affect the pelleting process. It appears that the powder particle characteristics, initially present in the powders, and which can lead to differences in pour and tap densities are minimised after milling suggesting that the milling process has reduced the number of powder agglomerates, in all the powders studied. The SEM micrographs of the milled powders also suggest a less agglomerated structure and show that, in the case of the $IDR_{(P)}$ powder the morphology of the particles is modified with the plates becoming more rounded.

Figure 4 shows the powder specific surface area (SSA) versus milling time, this indicates that for each powder the SSA increases with extent of milling. This also supports the theory that agglomerate breakdown is occurring during the milling process. Throughout the milling cycle the $IDR_{(P)}$ and the ADU powders exhibited very similar SSA values whereas the values for the $IDR_{(S)}$ powder was consistently lower.

As expected, the iron levels increased with prolonged milling however, the level of iron pick-up was well within existing specification levels and is not a problem to the successful application of the SBR in the fabrication plants.

3 .2 EFFECT OF MILLING ON GRANULE CHARACTERISTICS

Table 2 shows pour and tap density and flow results for the granulation trials on unmilled and milled powders. The unmilled powders showed no signs of granulation after tumbling for 1 hour and pour and tap densities were consistent with unmilled material which had not been granulated. Pour densities were, as expected, higher for milled powders after granulation, indicating the fact that granulation had taken place. Only one unmilled powder (IDRs) showed any tendency to flow whereas all milled materials had good flow properties.

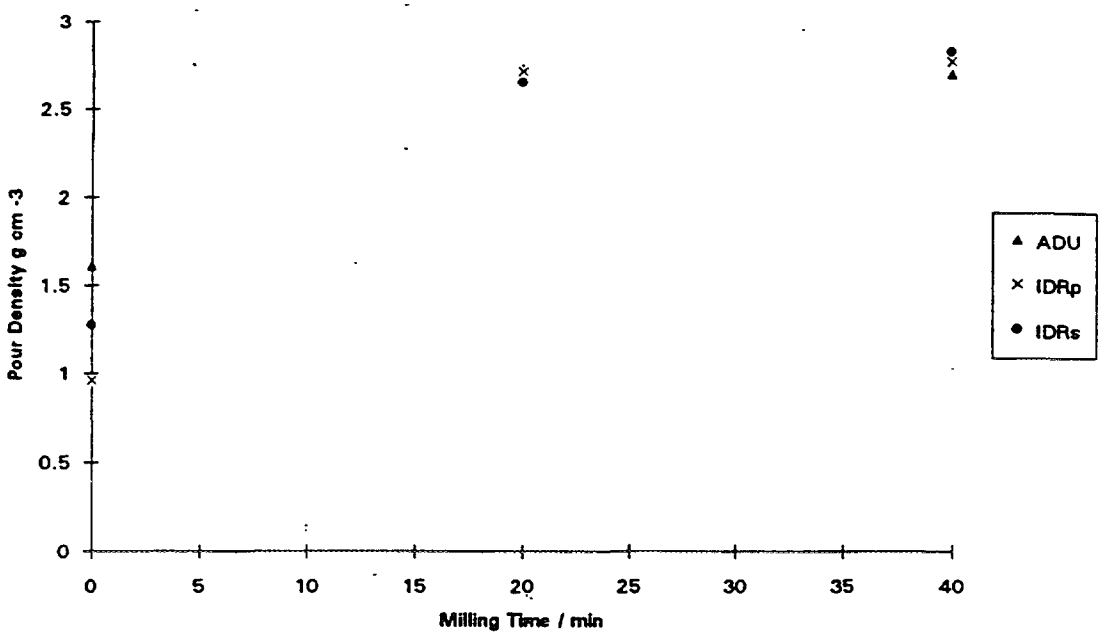


FIG. 2 *Powder Pour Density as a function of milling time*

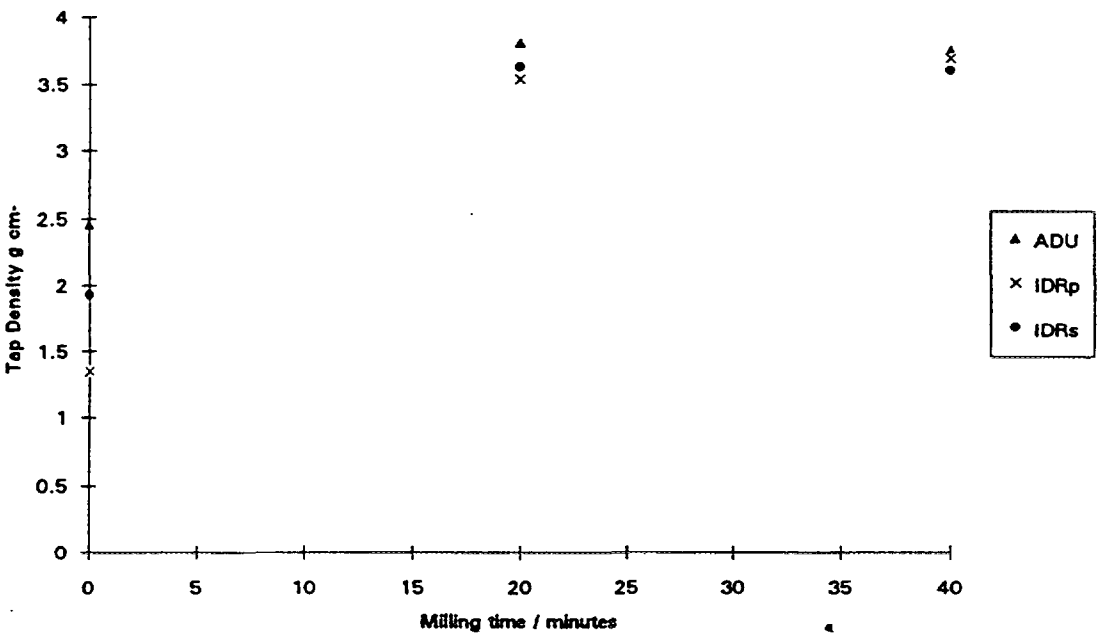


FIG. 3 *Powder Tap Density as a function of milling time*

Granules produced by spheroidisation build up by powder rolling over itself and agglomerating. The powder particles behave as nuclei, and 'pick-up' material via a 'snowball' effect to form larger granules. The granulation results show that the milled powders are the more cohesive and form stable free-flowing granules via the spheroidisation process. This is consistent with the fact that the milled material has a higher surface area and contains newly cleaved surfaces increasing the opportunity for interparticle bonding via Van der Waal's forces.

3.3 EFFECT OF MILLING ON POWDER COMPACT CHARACTERISTICS

Table 3 shows the compaction properties of each powder before and after milling. All powder green densities quoted were for pellets pressed at the same load of 4 tonnes cm^{-2}. The results confirm that the milling operation gives rise to increased green density for the same pressing load. Where granules were pressed at the same load there was a difference of between 0.68 - 0.95 g cm^{-1} in the green density achieved for unmilled powder and powder milled for up to 40 minutes. The greatest difference was shown using the IDRp example confirming the biggest change in powder morphology occurred in this example during milling. It is noticeable that after only 20 minutes milling the green density of pellets produced from each powder was similar.

3.4 EFFECT OF MILLING ON POWDER SINTERING CHARACTERISTICS

The densities of the sintered pellets produced from all compacts are shown in Table 4. For the unmilled powders, the ADU powder exhibited a much lower sintered density than IDR. Milling produced an increase in sintered density for all powders, with the largest increase observed in the ADU powder.

4. PLANT SCALE BEHAVIOUR

Plant scale trials (25kg) have shown that the most important difference between milled and unmilled material is the increased cohesive and adhesive nature of milled material, which, together with changes in ambient humidity may cause powder to adhere to surfaces of equipment. To overcome this, dry inert process gases and very highly polished process surfaces are used in the plant. The phenomena and trends observed in the lkg scale trials have generally been confirmed in large-scale trials.

TABLE 3. The Green Density Achieved for Granule Compaction at 4 Tonnes cm~2 for Given Milling Time

	Green Density / g cm^{-2} for given milling time / min		
	0	20	40
ADU	6.02	6.57	6.70
IDR_P	5.77	6.54	6.72
IDR_S	6.06	6.59	6.73

TABLE 4. Sinter Density Results for Green Compacts Pressed at a Load of 4 Tonnes cm 2

	Sinter Density / g cm^{-2} for given milling time / min		
	0	20	40
ADU	10.56	10.69	10.72
IDR_P	10.71	10.78	10.80
IDR_S	10.75	10.81	10.79

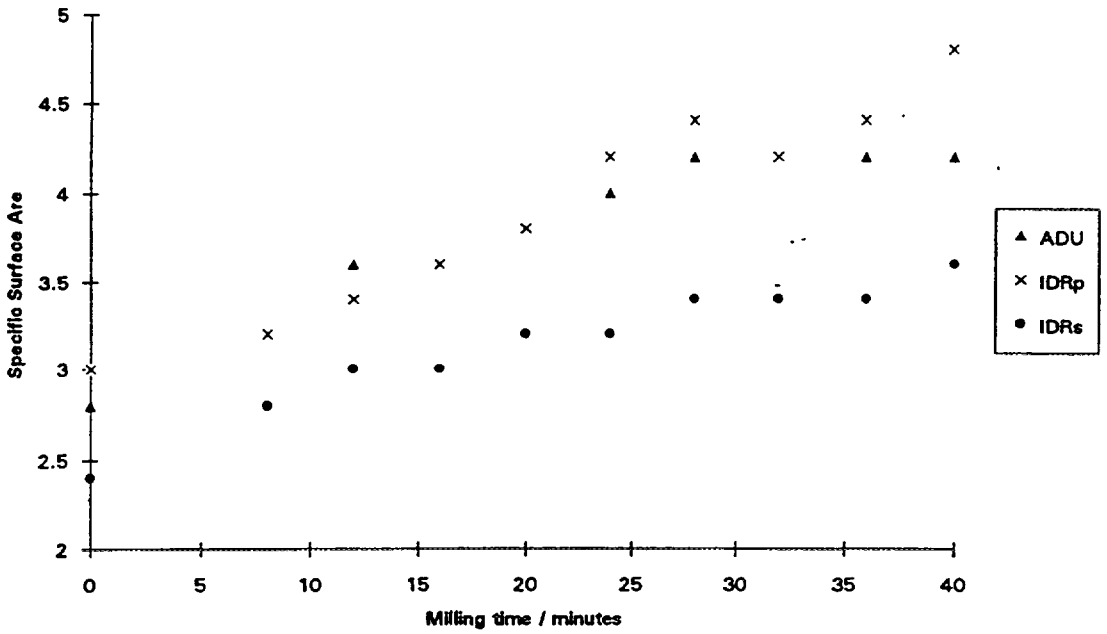


FIG. 4 Change in Powder Specific Surface area with milling time

5. CONCLUSIONS

Experimental studies have demonstrated how the milling of powders affects particle morphologies and subsequent behaviour of powder during pellet fabrication. The properties of powders changed most during the initial phase of milling but the change was less marked after further milling. All powders were successfully processed via the Short Binderless Route, producing good quality green and sintered pellets. The results support the use of ADU derived UO_2 in the SBR process.

MECHANISMS OF DRY BALL MILLING IN MOX FABRICATION

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Abstract

An important stage of MOX fuel pellets fabrication is the blending process of the nuclear powders which is carried out in the French plants in a ball milling and under dry conditions in order to avoid any criticality related difficulties. The milled powders must present a sufficiently intimate distribution of the UO_2 and PuO_2 particles which is necessary to synthesize the homogeneous $(UPu)O_2$ solid solution during the subsequent reactive sintering; the homogeneity of the UO_2/PuO_2 fuels is important in relation both to uniform irradiation performance and to solubility during reprocessing. Our study is related to the mechanisms which take place inside the ball mill as a function of the nature and size of the balls, the rotation speed, the milling time and the mill-to-ball charge-to-powder volume ratios. The milled powders have been characterized with a particular emphasis about the efficiency of the blending in accordance with the duration of the milling which can be one of the limiting parameter for industrial capacity of MOX fuel fabrication. Previous works have indicated the existence of three types of configurations of the balls in a ball mill as a function of the rotation speed (expressed relatively to the critical speed $Vc \quad \sqrt{g/(R \quad r)}$, R being the radius of the mill and r the radius of the balls). In the case of the nuclear oxides, video observations of the motions of the balls through a transparent cap of the cylindrical milling jar indicate that the milling of the powders is mainly achieved by the sliding mechanism with the rolling over becoming significant when the rotation speed is increased. The impact mechanism is almost never present. It can be therefore deduced that the micronization of the mixed UO_2 - PuO_2 powders is carried out by abrasion with the minor mechanism being a "static" crushing of the blended powders between the milling balls. The abrasion mechanism leads to a specific quality of the mixing of the UO_2 - PuO_2 powders which has been observed on green pellets of 60% d.th. by backscattered electron microscopy coupled with image analysis technique. After 4 h of milling, the class of particles larger than 13 μm has completely disappeared. These experimental data will help to establish a model describing the mechanisms of UO_2 - PuO_2 dry ball milling and have already made easier the development of the industrial milling at the new COGEMA MELOX plant.

I. INTRODUCTION

The mixing of nuclear oxides involved in the fabrication of MOX fuels has to be carried out under dry conditions in a ball mill in order to avoid criticality related difficulties. The milled powders must present a very intimate distribution of the UO_2 and PuO_2 particles which is necessary to synthesize the homogeneous $(UPu)O_2$ solid solution during the subsequent reactive sintering at high temperature. The homogeneity of the UO_2/PuO_2 fuels is important in relation both to uniform irradiation performance and to solubility during reprocessing.

As a matter of fact, the atomic diffusion of U and Pu under the standard sintering conditions (reducing gas at 1700°C) is limited and to obtain a final homogeneous MOX fuel requires a blending of the different powders at a micronic scale. The milling stage is then necessary to produce a micronised and correctly mixed powder to be pelletized.

The milling/blending stage of the MIMAS (MIcronization of a MASterblend) fabrication process used in the COGEMA plants has been the subject of extensive investigation and refinement over several years. The study which is presented here is a part of this R and D program and is related to the mechanisms which take place in a dry ball miller; instead of plutonium oxide, equivalent (for the powder

metallurgy characteristics) cerium oxide is used to facilitate the preliminary experiments outside confinement glove boxes. The motion of different kinds of balls is observed as a function of the rotation speed of the pot and the amount of balls and powder. The milled powders are characterized with a particular emphasis about the quality of blending with respect to the duration of the milling operation which is an important parameter for industrial capacity of production of MOX fuels.

2. MECHANISMS OF DRY BALL MILLING

2.1 CONFIGURATIONS OF THE BALLS

Previous work [1] indicates the existence of three types of configurations of the balls in a ball mill as a function of the rotation speed (Fig. 1). The rotation speed is expressed in percentage of the critical speed in order to take into account various values of the diameter of the mill. The critical speed V_c corresponds to the rotation speed for which the balls are just centrifuged if the friction forces are neglected and is expressed by the following relationship: $v_c = \sqrt{g/(R - r)}$, R being the radius of the mill and r the radius of the balls.

At low rotation speed, the balls behave like a compact body which simply oscillates with the motion of the pot (milling by sliding). Then the rotation speed is increased, the amplitude of the oscillation increases and the bulk of the balls takes the shape of an inclined plane from which some balls are ejected and roll over all the other balls (milling by rolling over). The bed of balls is then less compact and the powders to be milled can penetrate between the balls. For still higher rotation speeds (higher than 75% of V_c) the balls are thrown out at the top of the inclined plane by their kinetic energy and follow a parabolic path (milling by impact). The two first configurations (sliding and rolling over) lead to a milling by abrasion involved in the balls-powder or lining-powder friction since the impact mechanism splits up the particles under the effect of shearing or crushing stresses. The abrasion concurs to the formation of small size particles in the milled product [2].

2.2 INFLUENCE OF MILLING MECHANISMS, ON THE POWDERS

First observations of the motion of the balls have been carried out through a transparent cap of a cylindrical milling jar by video recordings with no powder inside the jar. These experiments, performed with uranium metal orthocylindrical balls (size: 15 x 15 mm; weight: 45 g), indicate that the centrifugation is never reached in a 15 cm diameter drum even for rotation speed higher that the critical speed V_c = 108 rpm. When same size but lighter (7 g) alumina balls are used, the centrifugation is normally obtained at V_c.

Observations have also been made with powder inside the mill with various filling values described by the parameter J = balls volume/mill volume; the studied criteria are then the size of the balls (15 x 15 mm, 45 g or 25 x 25 mm, 225 g), the rotation speed (up to 130 rpm) and the J parameter (between 20 % and 50 %). In this broad experimental range, the milling of the powder is mainly achieved by the sliding mechanism with the rolling over becoming significant when the rotation speed is increased; this variation is observed at lower speed when the volume of balls is raised. The impact mechanism is almost never present. The micronisation of the mixed UO_2 - CeO_2 powders is therefore carried out by abrasion with the minor mechanism being a "static" crushing of the blended powders between the milling balls.

The influence of the different kinds of mechanisms on the morphological aspect of the powders has been investigated by scanning electron microscopy: sliding and rolling over lead to the formation of very fine particles (Fig. 2a) due to the abrasion of the initial UO_2 granulates. If the impact mechanism is preponderant (which is sparsely the case), the granulates are crushed in agglomerates of various shapes and of higher compactness (Fig. 2b).

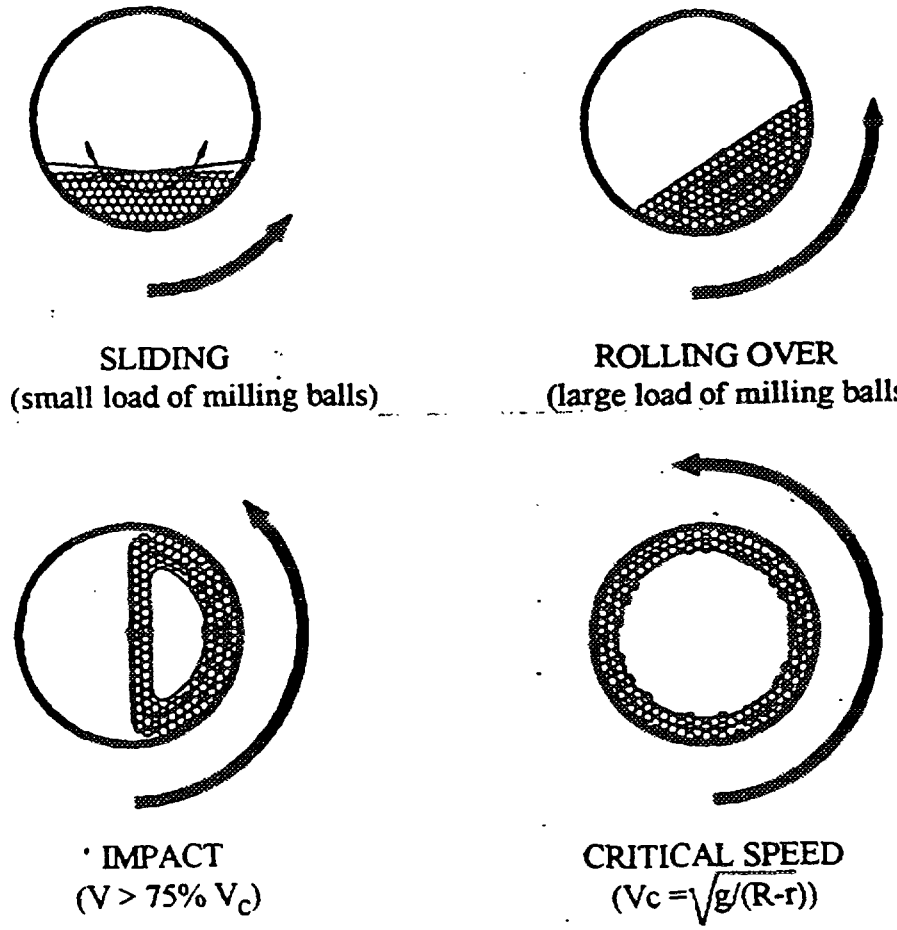


FIG. 1 Theoretical Milling Mechanisms as a Function of Balls Load and Rotation Speed

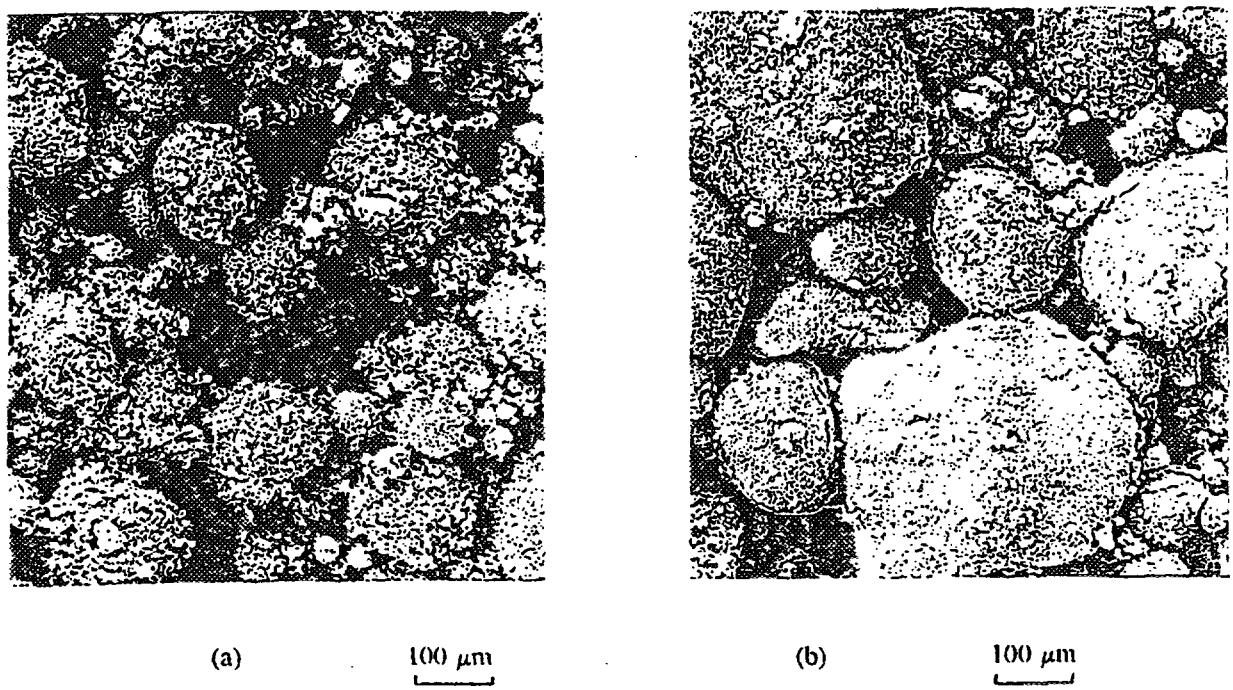


FIG. 2 Electron Images of UO_2 Powder Milled by Abrasion (a) or by Impact (b)

3. CHARACTERIZATION OF MILLED AND BLENDED POWDERS

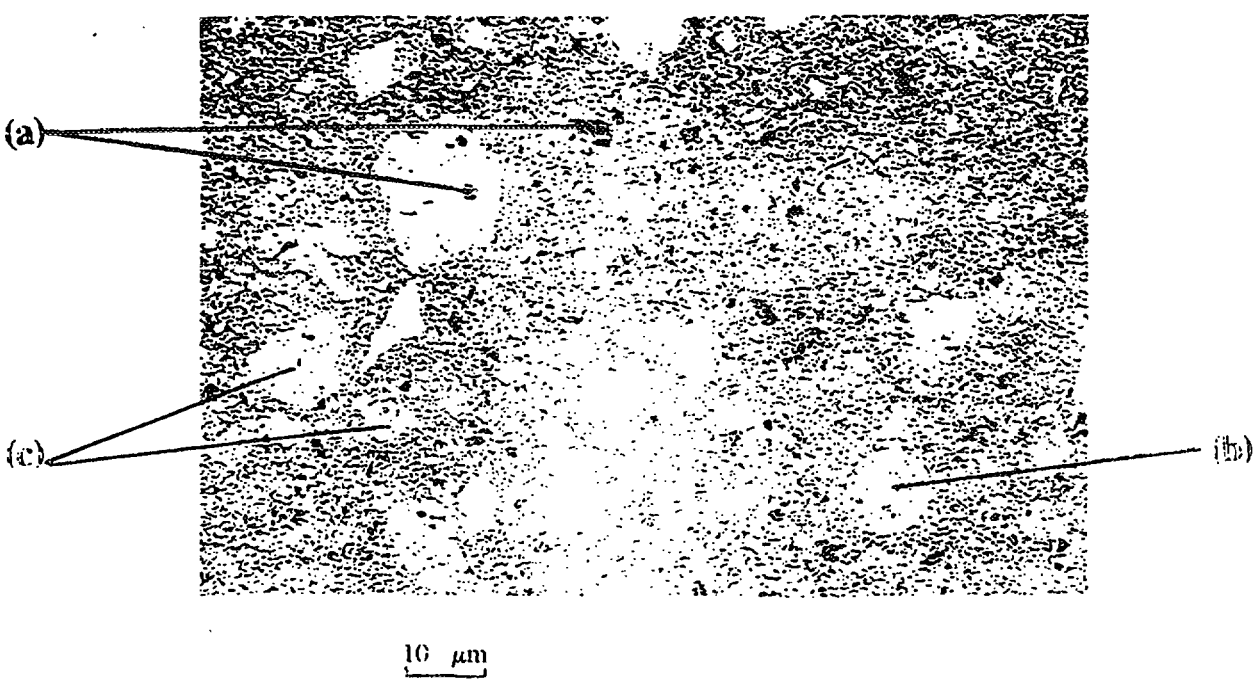
3.1 CHARACTERIZATION TECHNIQUE

The quality of the mixing of the UO_2 - CeO_2 or UO_2 - PuO_2 powders cannot be easily described by simple analytical techniques, especially at a micronic scale: chemical determinations on very small samples, X-ray diffraction measurements or optical microscopy are not sufficiently sensitive for detecting very small scale heterogeneities. For instance, optical observations are inadequate to permit the identification of the two kinds of oxides despite the strong contrast of colors between these two elements (white cerium and dark uranium) and could even lead to some misinterpretation.

To overcome this difficulty, the dispersion of the minor phase has been observed by scanning electron microscopy coupled with image analysis technique on green pellets of 60% d.th.; this analytical method allows the direct observation of the mixed powders without the modifications induced by the diffusion of the cations during sintering. The green pellets have to be smoothly polished which allows SEM observations with backscattered electrons. The large difference between the atomic numbers of cerium and uranium shows the lighter cerium in grey zone whereas the heavy uranium appears in white (Fig. 3). In this case, two magnifications (x650 and x2500) are used with respectively 30 and 90 investigated fields so that particles of size between 0.5 μm and 25 μm can be counted with a satisfactory statistics. This technique has been applied for instance to a milling of (78 vol% UO_2, 22 vol% CeO_2), which corresponds roughly to the proportions of a master blending in the industrial MOX process, with 15 x 15 mm uranium balls, J = 57 % and U = (powder apparent volume / between-balls vacuum volume) = 1. The rotation speed is kept at 48 rpm with a total duration of the milling of 8 h (samplings are also taken each 2 h of milling).

3.2 GRANULOMETRIC RESULTS ON GREEN PELLETS

Under these conditions, the granulometric spectrum shows no particles of cerium oxide larger than 25 μm after 2 h of milling; for this duration, there is only 1.3 % of particles between 13 μm and 25 μm.



(a) cerium oxide (b) no milled uranium oxide (c) dense agglomerates

FIG. 3 Backscattered Electron Image of a Green Pellet of 60% D.TH. (78 vol% UO_2, 22 vol% CeO_2)

After 4 h, this class of particles has completely disappeared. For the particles between 13 and 0,5 μm, the numbers counted in each category remain remarkably and surprisingly constant versus test time. The surface occupied by cerium oxide over the total surface of the sample has also been measured and shows a significant decrease as a function of this same parameter. This overall set of results can only be interpreted by the fact that there must be some formation of very small particles of size less than 0.5 μm which is the detection limit of our SEM analysis; the initial large particles are not directly divided in new fragments (this should lead to the unobserved large variation of the granulometric spectrum) but continuously weared during the milling. Moreover, the mean diameter of "imaginary" grains for each class has been computed from their influence zone data by image analysis and presents a strong increase with the duration of milling whatever the size category: this means that, as expected, the inter-particles distance continuously increases with almost no segregation of the minor component.

4. CONCLUSION

These experimental results confirm clearly that the abrasion is the main mechanism occurring during the milling of nuclear powders and is responsible for the reduction of the size of the initial particles and of their subsequent blending; the impact mechanism is never reached under these conditions. The detailed analysis of the quantitative data in terms of the homogeneity of the distribution of the minor powder (i.e. plutonium oxide) in the bulk of the main compound (i.e. uranium oxide) is therefore of great importance in order to determine the optimal values of the parameters of the milling stage in the fabrication of MOX fuels.

The influence of other parameters such as the utilization of a chemical agent capable of avoiding the agglomeration of powders or the introduction of scraps with the raw materials has also been investigated. This information will help to establish a model describing the mechanisms of UO_2 - PuO_2 dry ball milling under a large range of conditions and have already been successfully used in the development of the industrial fabrication process of the new MELOX plant of COGEMA.

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CATION DIFFUSION AND CRYSTAL GROWTH AT THE UO_2-PuO_2 INTERFACE

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Abstract

A better understanding of the homogenizing phenomena involved in the dry route fabrication of MOX fuel has led to the study of interdiffusion coefficients of plutonium and uranium during the sintering. Interdiffusion coefficients have been calculated from electron microprobe analysis of diffusion couples UO_2 (green compact) / PuO_2 (green compact). A series of these diffusion couples has been sintered at 1773, 1873 and 2023K during 0 to 50 hours and under controlled oxygen potential (-88, -95 and -125 kcal/mol). The results show that grain-boundary diffusion predominates, essentially at the end of the intermediate stage of the sintering. On the opposite, the lattice diffusion is very slow. The sintering and particularly the crystal growth enhance the interdiffusion coefficients which are slightly greater compared to those of polycrystalline diffusion couples.

INTRODUCTION

A significant problem associated with fabrication of MOX fuel is that it contains plutonium distribution heterogeneities which can produce high burnup zones during irradiation and create residue that cannot be dissolved during reprocessing. To improve plutonium distribution during fabrication, it seems necessary to better understand the homogenizing phenomena involved, namely the grinding of the uranium and the plutonium oxide powders and the reactive sintering of the uranium and plutonium oxides. This study considers more specifically the sintering in an attempt to understand the U and Pu cation interdiffusion mechanisms, which take place during the formation of the solid solution and the densification. This phenomenological study will help to establish models for matching the sintering parameters to the quality of the powder mixtures produced by grinding.

EXPERIMENTAL METHOD

The traditional approach to modelling the thermal homogenization of the Pu and U cations is to investigate the chemical interdiffusion for a couple of uranium oxide and mixed uranium/plutonium oxide mono or polycrystals bonded together by hot-pressing [1-6]. In this experimental work, the diffusion couple is realized from a pellet of PuO_2 powder placed inside a pellet of UO_2 powder. The contact between these two green pellets is obtained thanks to the differential shrinkage during the heat treatment, which is more important for the outer UO_2 pellet than for the inner PuO_2 pellet. Thus, the interdiffusion coefficients, measured under variable thermodynamic conditions, take into account mechanisms that occur during sintering and lead to a more realistic approach.

After the heat treatment, the diffusion couple is sliced at right angles to the UO_2/PuO_2 interface, polished and then subjected to a chemical attack in order to examine its grain structure near the diffusion interface by optical microcopy. This analysis is completed by electron microprobe examinations to obtain X-ray images of the plutonium and uranium distributions, and the concentration profiles of uranium and plutonium along the diffusion zone. After smoothing of the profiles using the least squares method, the apparent interdiffusion coefficients Da were calculated using the MATANO method.

The parameters studied in this work were temperature (1773, 1873, 2023K), sintering time (0, 4 or 50 hours) and oxygen potential of the sintering gas (-88, -95 and -125 kcal/mol).

RESULTS AND DISCUSSION

Phenomenological study

The X-ray images at the UO_2 and PuO_2 interface (see example in Figure 1) show that the plutonium migrates very quickly and in depth through the grain boundaries of the plutonium oxide, essentially at the end of the intermediate stage of densification, when all the porosity is closed and remains discontinuously on grain boundaries. This intergranular diffusion of plutonium is simultaneously completed by a lattice interdiffusion, which is much slower than the grain boundary diffusion. The intergranular interdiffusion is therefore the kinetic factor controlling the homogenization of U and Pu cations. The uranium diffusion is very similar to the plutonium, but with a more limited extensiveness.

These diffusion phenomena, which occur during the final stage of densification ($T \approx 1773K$), disturb the UO_2 and PuO_2 grain growth, as it can be seen on Figure 2. The average grain size is all the finer as the diffusion processes are important, that is to say near the UO_2/PuO_2 interface. It is probably due to the fact that the activation energy of grain growth is more important than that of solid solution formation. The latter occurs also to the detriment of the former.

Oxygen potential effect on the global interdiffusion coefficient

Figure 3 gives values of the apparent interdiffusion coefficients versus oxygen potentials at 2023K and for a sintering time of 50 hours. Influence of the oxygen potential in this range of measures is not very simple to explain, because of the probable presence of a minimum of diffusion, as it has been reported in previous works [1-2]. This one is not really determined because of the disparity and the lack of data. In any case, the interdiffusion coefficients, which are measured here, are systematically higher than those observed in previous works. This increase of the diffusion coefficient may be the direct consequence of introducing the additional diffusion factor constituted by the densification and the grain growth which occurs during the final stage of the sintering.

Temperature effect on the global interdiffusion coefficient

Figure 4 gives the interdiffusion coefficients according to an Arrhenius representation. The measured interdiffusion coefficients reported in this work are in reasonable agreement with those calculated in previous works. By comparing these results with those obtained thanks to monocrystals, an important decrease of the activation energy can be observed because of the predominance of the grain boundaries diffusion with regard to the lattice interdiffusion. But the disparity of the results, for the reasons which are described forward, does not permit to detect any influence of the densification and grain growth on the thermal activation of interdiffusion mechanisms

Sintering time effect on the global interdiffusion coefficient

Variation of the apparent interdiffusion coefficient as a function of the sintering time (Figure 4) clearly shows the evolution of the mechanisms over time. It is evident that there is practically no diffusion immediately after the temperature rise ($t=0$). After 4 hours of sintering time, the diffusion mechanisms are essentially intergranular, the lattice diffusion being too slow to participate greatly to the overall diffusion process. On the opposite, after 50 hours of sintering, the lattice interdiffusion contributes to decrease the global diffusion coefficient by nearly one order of magnitude.

Generally speaking, it can be seen that there is no sensitive influence of the plutonium concentration on the global interdiffusion coefficient, except a slight increase for the high plutonium content. This increase can be explained by the alone contribution of the grain boundary diffusion in the most depth region which is represented by the highest Pu level.

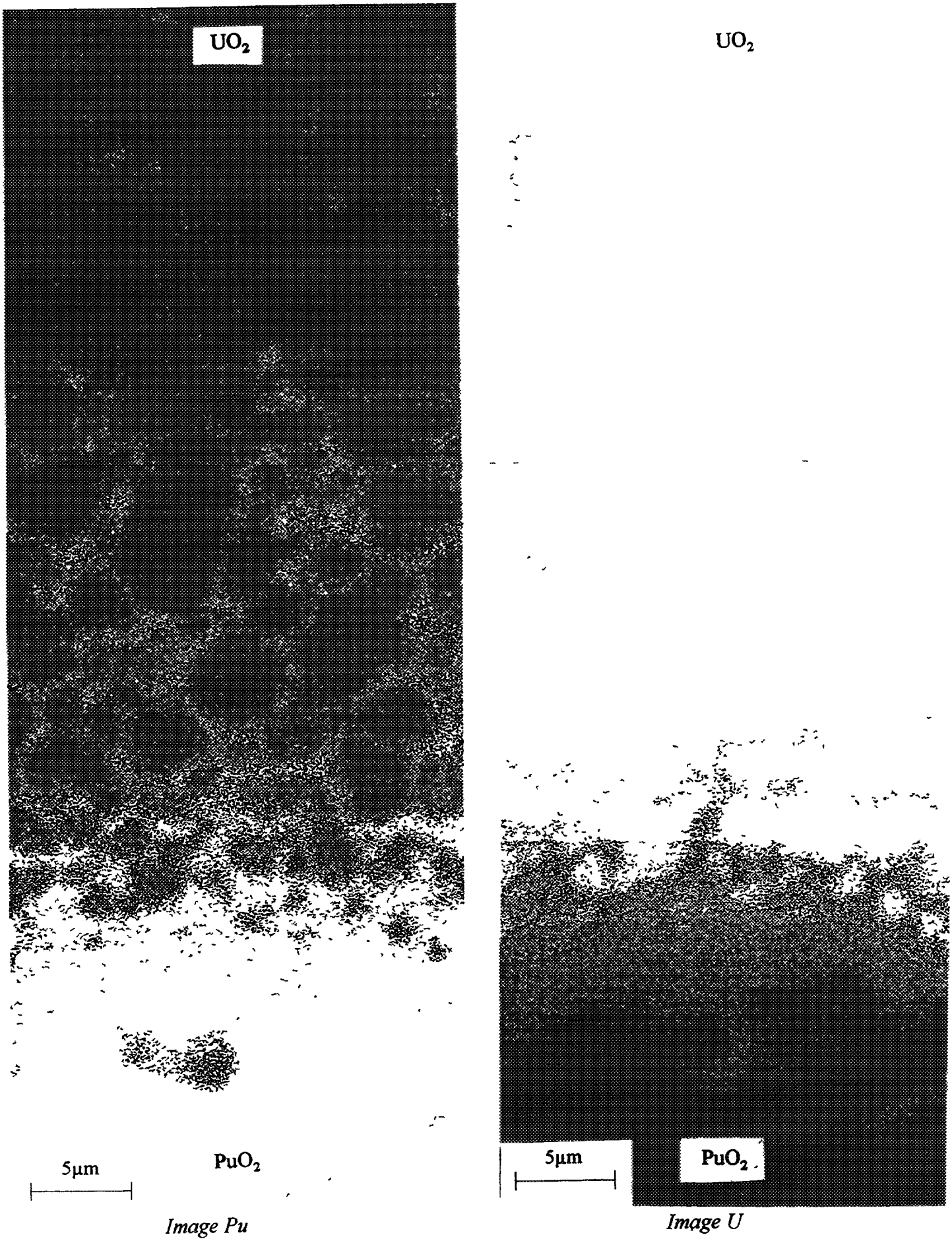

FIG. 1. *X-ray images at the interface of the UO_2-PuO_2 couple sintered at 2023K for 50 hours under N_2-5%H_2 (pH_2/PH_2O = 20).*

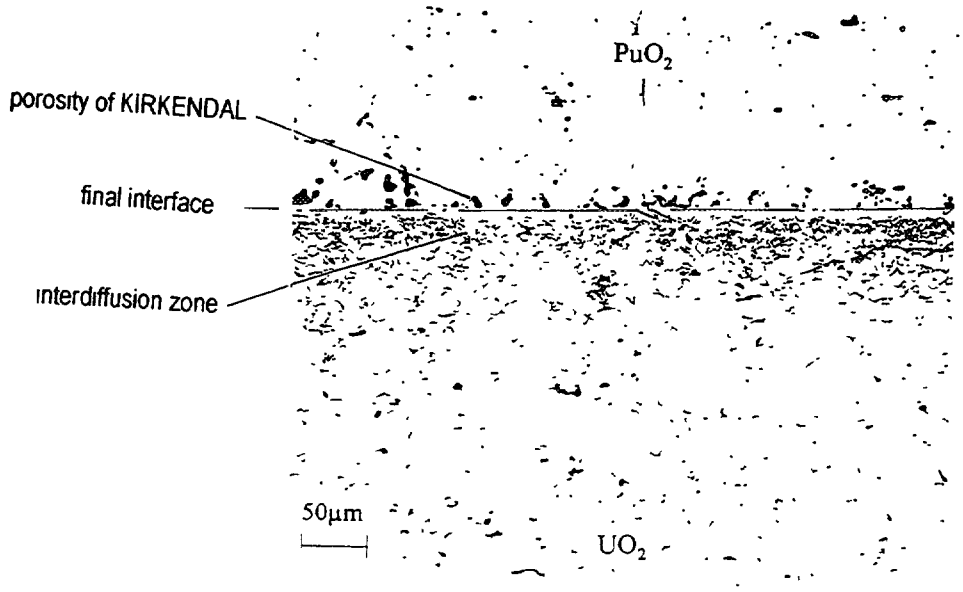

FIG. 2. Evolution of UO_2 grain sizes along the interdiffusion zone (x200).

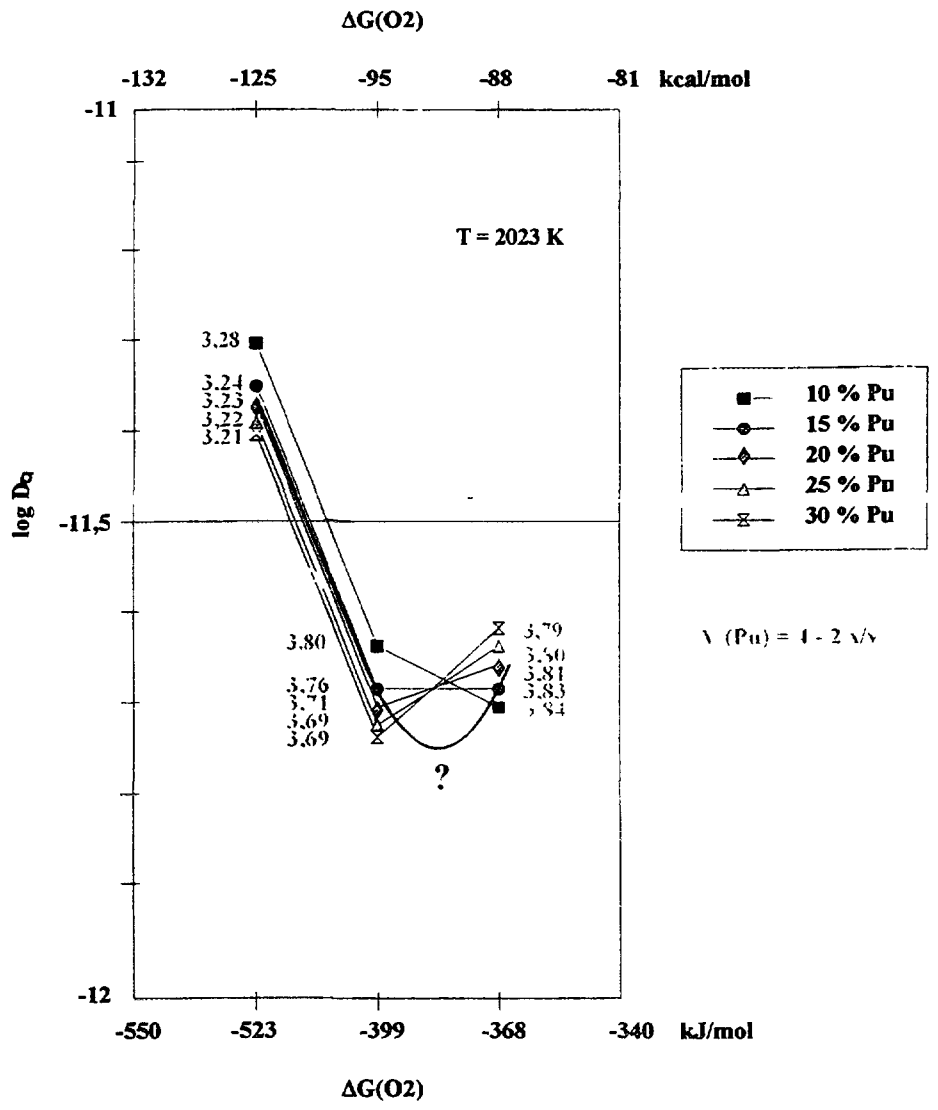

FIG. 3. Variation of the apparent interdiffusion coefficient in a UO_2/PuO_2 couple sintered at 2023K for 50 hours as a function of oxygen potential.

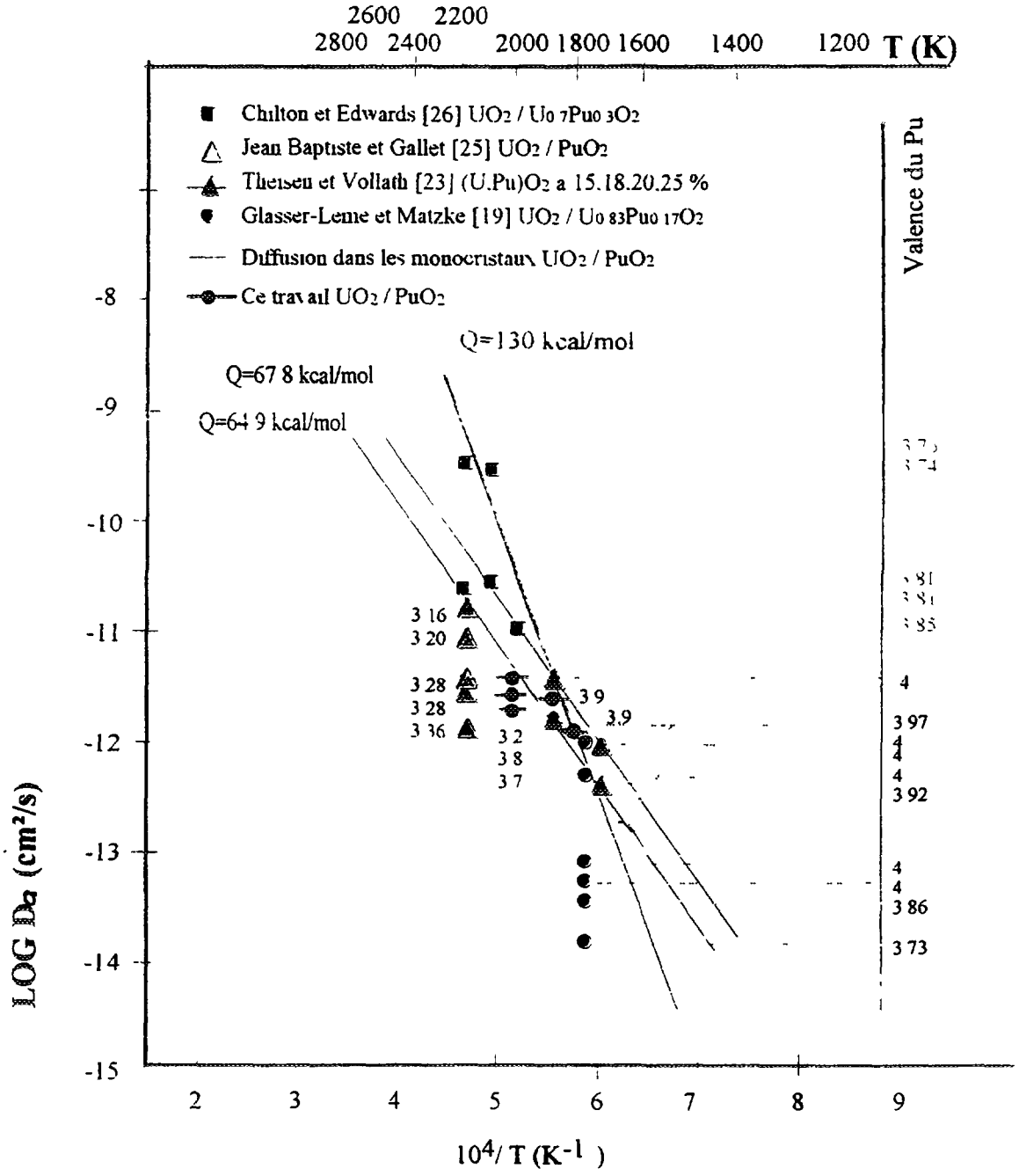

FIG. 4. *Variation of the apparent interdiffusion coefficient in a UO_2/PuO_2 couple sintered for 50 hours under N_2-5%H_2 (pH_2/PH_2O = 20) as a function of temperature (Arrhenius diagram).*

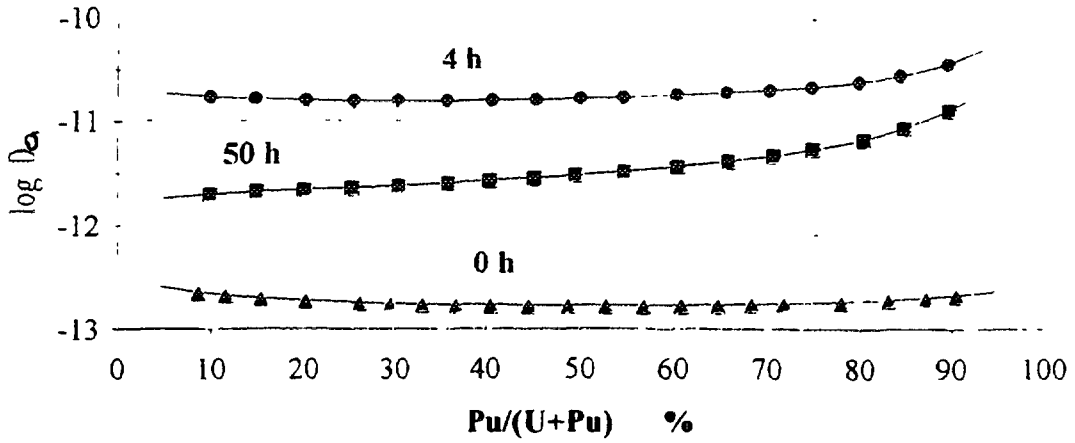

FIG. 5. *Variation of the apparent interdiffusion coefficient in a UO_2/PuO_2 couple sintered for 50 hours under N_2-5%H_2 (pH_2/PH_2O = 20) as a function of Pu content for different sintering times.*

CONCLUSION

Investigation of the U and Pu interdiffusion coefficients obtained from compacted uranium and plutonium oxide powders enables determination of the diffusion mechanisms involved, taking into account the additional diffusion effect of densification and grain growth. This work has shown that, whatever the sintering conditions, plutonium is found to migrate very quickly and in depth through the grain boundaries of uranium oxide. The intergranular diffusion starts at about 1773K, at the end of the intermediate sintering stage. Uranium diffuses through the grain boundaries of plutonium oxide, but much more slowly.

On the other hand, the lattice interdiffusion of U and Pu is a slow process which contributes very slightly to the overall diffusion during 4 hours of the usual industrial sintering time. Thus, it limits the homogenization kinetics during the sintering of MOX fuels.

At last, apparent interdiffusion coefficients, measured at 1873K for the low oxygen potentials considered in this study, are slightly greater than those measured in previous works for a sintered couple. Furthermore, plutonium content does not seem to affect the value of interdiffusion coefficient. These two behaviours differ from that observed during previous works and might be interpreted as an indication of a significant effect of simultaneous densification and grain growth in the overall homogenizing diffusion process.

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ADVANCED ANALYSIS TECHNOLOGY FOR MOX FUEL

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Abstract

PNC has developed MOX fuels for advanced thermal reactor (ATR) and fast breeder reactor (FBR). The MOX samples have been chemically analysed to characterize the MOX fuel for JOYO, MONJU, FUGEN and so on. The analysis of the MOX samples in glove box has required complicated and highly skilled operations. Therefore, for quality control analysis of the MOX fuel in a fabrication plant, simple, rapid and accurate analysis methods are necessary. To solve the above problems, we have developed instrumental analysis and techniques. This paper describes some of the recent developments in PNC. 2. Outline of recently developed analysis methods by PNC. 2.1 Determination of oxygen to metal atomic ratio (O/M) in MOX by non-dispersive infrared spectrophotometry (NDIR) after inert gas fusion. The sample and nickel metal flux were fused in a graphite crucible using an impulse furnace under a helium atmosphere. The oxygen in the MOX fuel was quantitatively reacted with the carbon in the graphite crucible and carbon monoxide was evolved, which was determined using the NDIR. The oxygen content in the MOX and the O/M were calculated from the carbon monoxide analysis results. The relative standard deviations (RSD) were less than 0.35% for the MOX pellets of 5% and 25% plutonium. The time required for one determination was about 10 minutes. The apparatus was originally developed in collaboration with PNC and Horiba Co. Ltd. 2.2. Determination of occluded gas component analysis in MOX using gas sampling unit and gas chromatography. The gases extracted from the sample at vacuum condition were injected into a gas chromatograph through an originally developed gas sampling unit. The gases were simultaneously determined using the improved gas chromatograph equipped with several separation columns and a photo-ionization detector. The RSD were 1.0, 3.4, 0.6, 1.0, 1.4, 0.8 and 0.9% for H_2, O_2, N_2, CH_4, CO_2, CO and C_2H_6, respectively. The results of gas analysis using this method agreed with the high temperature vacuum extraction method in the range of less than 300μl of extracted gas. 2.3. Determination of nitrogen in MOX by gas chromatography after inert gas fusion. This technique is based on chromatographic determination of nitrogen in released gases from fused sample. The sample and iron metal flux in a graphite crucible were heated up to 2700pC under a helium atmosphere. From the fused sample, which were gases evolved such as hydrogen, nitrogen and a large amount of carbon monoxide. The carbon monoxide produced was interfered with the measurement of nitrogen. Therefore the aim of nitrogen separation was determined using a thermal conductivity detector. The RSD was less than 5%, and the time required for one determination was about only 10 minutes. 3. Future Plans. In order to put these analysis technologies for MOX to practical use, further improvements are needed. Particularly, the development of automatic handling systems, simultaneous determination methods for impurity and more accurate methods need to be developed.

1. INTRODUCTION

PNC has developed MOX fuels for advanced thermal reactor (ATR) and fast breeder reactor (FBR). The MOX samples have been chemically analysed to characterize the MOX fuel for JOYO, MONJU, FUGEN and so on. The analysis of the MOX samples in glove box has required to complicated and high skilled operations. Therefore, for quality control analysis of the MOX fuel in a fabrication plant, simple, rapid and accurate analysis methods are essential. To solve the above problems, we have developed instrumental analysis and techniques. This paper describes some of the recent developments in PNC.

2. DETERMINATION OF OXYGEN TO METAL ATOMIC RATIO IN MOX BY NONDISPERSIVE INFRARED SPECTROPHOTOMETRY AFTER INERT GAS FUSION

The present method is based on the extraction gas analysis using nondispersive infrared spectrophotometry (NDIR) after inert gas fusion. The oxygen in the fused sample was quantitatively reacted with the carbon in a graphite crucible and the CO generated was determined using the NDIR. The relative standard deviation (RSD) was less than 0.20%, and the time required for one determination was about 10 min for all analytical operations. This method is applicable to sintered MOX fuels over a wide range of plutonium content.

2.1 EXPERIMENTAL

2.1.1 The O/M analysis apparatus

The O/M analysis apparatus consists of a gas extraction unit, a gas dilution unit, the NDIR, an operating controller, and a computerized peak-area integrator. The schematic diagram of the apparatus is shown in Fig. 1. The gas extraction unit consists of an impulse furnace, a power supply, a refrigerator and a compressor. The impulse furnace consists of the upper and lower electrodes and the electrode driving cylinder. The heating conditions of the sample were controlled using the operating controller. The gas dilution unit consisted of a dilution vessel, a pressure regulator and a needle valve. The gas extracted from the fused sample was divided into 2 and 8 by the gas dilution unit. This is the reason why the NDIR that utilized linear response range for carbon monoxide determination is designed and constructed. A computerized peak-area integrator from Shimazu Co., Ltd. (Kyoto, Japan) CR-3A was used.

2.1.2 Measurement Procedure

The sample (ca. 20 - 5Omg) was packed into a nickel capsule and weighed to an accuracy of 0.1mg. The inlet of the capsule was sealed off using a manual type press with a pair of dies. The sealed capsule was placed into a degassed graphite crucible and then placed in the impulse furnace. It was

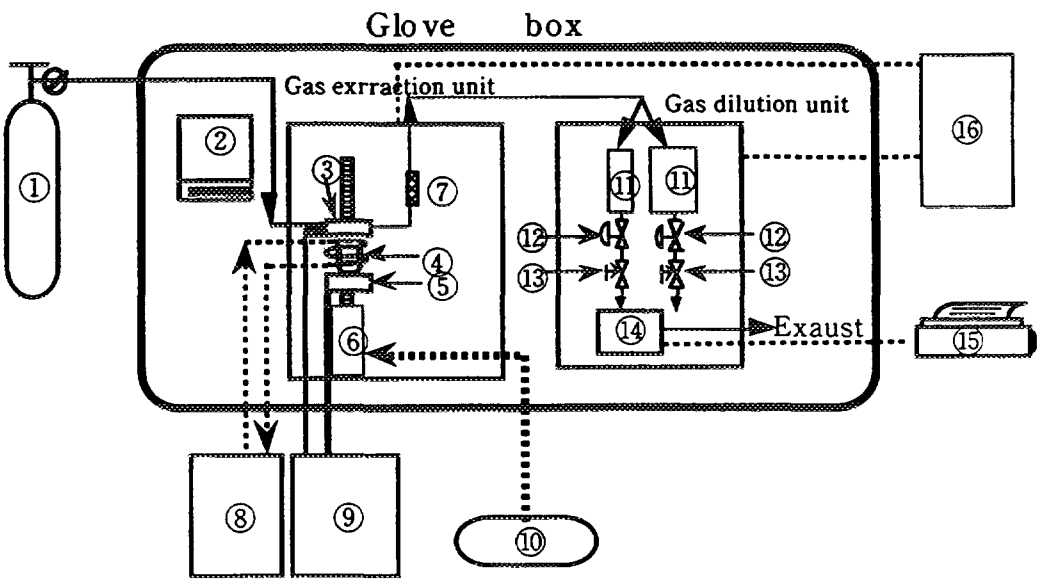

Fig. 1. Schematic diagram of O/M analysis apparatus. ① Helium bomb; ② Balance; ③ Upper electrode; ④ Graphite crucible; ⑤ Lower electrode; ⑥ Electrode driving cylinder; ⑦ Dust filter; ⑧ Refrigerator; ⑨ Power supply, ⑩ Compressor; ⑪ Gas dilution vessel; ⑫ Pressure regurator; ⑬ Needle valve; ⑭ Nondispersive infrared spectrophotometric detector; ⑮ Integrator; ⑯ Operating controller.

continually heated at about 2000°C for 30 s and 2500°C for 30 s in a current of helium gas. The O_2 in the sample was quantitatively reacted with carbon in the graphite crucible, and the CO was evolved. These gases were transferred to the buffer vessel and diluted by helium carrier gas. The CO diluted was determined using the NDIR, and the peak-area was integrated using a computerized integrator. The concentration of O_2 in the nickel capsule and the crucible was analysed beforehand and this blank value was deducted from the sample analysis value. The O/M of the sample was calculated using the following equations:

$$Co = Mo/(Mc + Mo) \times Cco \quad \ldots\ldots\ldots\ldots \quad (1)$$

$$Co = Mo\,X/(M + Mo{\cdot}X) \times 100 \quad \ldots\ldots\ldots\ldots \quad (2)$$

$$X = Co{\cdot}\,M/(100 - Co) \times 1/Mo \quad \ldots\ldots\ldots\ldots \quad (3)$$

where Co is the concentration of O_2 in the sample (wt%), Cco is the analysis result of CO using the present method (wt%), Mo and Mc are the atomic weights of O_2 and carbon, M is the mean atomic weight of metal in the sample, and X is the O/M ratio. Equation (3) was derived from equation (2).

2.2 RESULTS

2.2.1 Calibration curve

The calibration curve for the determination of O_2 was obtained using a O/M = 2.00 controlled MOX fuel by varying the weight over the range of 10 to 60 mg. The calibration curve obtained is shown in Fig. 2. The calibration curve gave good linearity in the range of 1.2 to 7.2 mg for the theoretical value of oxygen content.

2.2.2 Comparison with gravimetric method and precision

The O/M ratio measurement for the sintered MOX fuel pellet containing less than 10% plutonium has been determined using the oxidation weighing method [1]. On the other hand, for sintered MOX fuel containing more than 10% plutonium the O/M ratio has been determined using the oxidation-reduction method [2,3]. These methods are based on gravimetry.

In order to compare the gravimetric method and presented method, sintered MOX fuels containing about 3% plutonium was used. Moreover, accuracy of this method was measured using 25% plutonium MOX. The measurement results of the O/M ratio are shown in Table I and Table 2. The result of the present method was in good agreement with the gravimetric methods. The RSD was less than 0.20%, and the time required for one determination was about 10min for all analytical operations

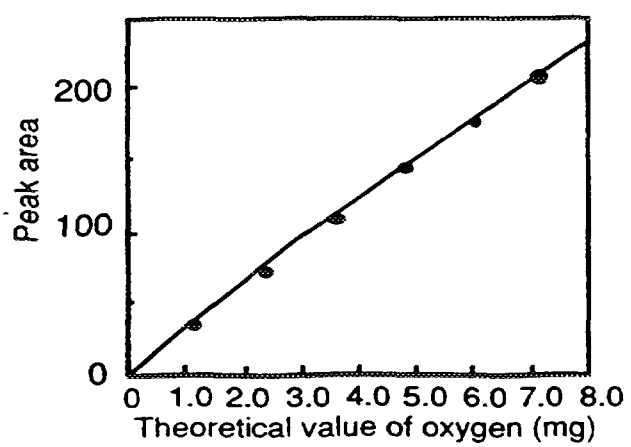

Fig. 2. Calibration curve for oxygen.

TABLE 1 **Comparison of the gravimetric method and the present method for O/M analysis in 3% plutonium MOX**

Method	O/M ratio[a]
Gravimetric method	$2.003_9 \pm 0.003_4$
Present method	$2.004_1 \pm 0.003_7$

a Mean ± standard deviation (n = 8)

TABLE 2 **Determination of O/M in 25% plutonium MOX by the present method**

Expt. No.	O/M ratio
1.	1.975
2.	1.985
3.	1.977
4.	1.980
5.	1.984
6.	1.978
7.	1.977
8.	1.981
Mean Value	1.979_6
SD	0.003_5
RSD (%)	0.179

SD Standard deviation

RSD Relative standard deviation

3. DETERMINATION OF OCCLUDED GAS COMPONENT ANALYSIS IN MOX USING GAS SAMPLING UNIT AND GAS CHROMATOGRAPHY

The gases extracted from the sample at vacuum condition were injected into a gas chromatograph through an originally developed gas sampling unit (GSU). The gases were simultaneously determined using the improved gas chromatograph equipped with several separation columns and a photo-ionization detector (PID). The relative standard deviations (n=10) were 1.0, 3.4, 0.6, 1.0, 1.4, 0.8 and 0.9% for H_2, O_2, N_2, CH_4, CO_2, CO and C_2H_6, respectively. The gases H_2, O_2, N_2, CH_4, CO_2 and CO from the MOX fuel pellets were simultaneously determined in this study. The results of gas analysis using this method were in agreement with that of the high temperature vacuum extraction method in the range less than 300μ l of extracted gas [4].

3.1 EXPERIMENTAL

3.1.1 The gas components analysis apparatus

The gas components analysis apparatus consists of a gas extraction unit, a gas collection and a volumetric measurement unit, the GSU, the gas chromatograph and a computerized integrator. Except for the GSU and the gas chromatograph, this apparatus has been widely used for the volumetric analysis of total gas in nuclear fuel pellets by the high temperature vacuum extraction method [5]. The schematic diagram of the apparatus is shown in Fig.3. The GSU consists of special three-way and four-way valves, a buffer vessel for pressure change and a reference gas injection port, as shown in Fig. 4. Both valves were connected to glass capillary tube of i.d.2mm. The GSU could be connected with the gas collection and volumetric measurement units at vacuum condition (10^{-2}Pa) and the gas chromatograph at carrier gas pressure (2 ~ 5Kg/cm^2) without pressure change of the system and chromatogram fluctuation. The gas chromatograph (Hitachi, GC-3000, Tokyo, Japan) and a computerized integrator (Hitachi, D-2500) were used in our experiments. For gas separation, 4.0m × 3mm i.d. stainless steel columns packed with Porapak Q (50 ~ 80 mesh), 2.0m × 3mm i.d. stainless steel columns packed with Molecular sieve 5A(60 ~ 80 mesh) and 2.0m × 3mm i.d. stainless steel columns packed with Chromosorb W(60 ~ 80 mesh) were used. In order to regulate the retention time, an empty stainless steel column of 2.0m × 3mm i.d. was used.

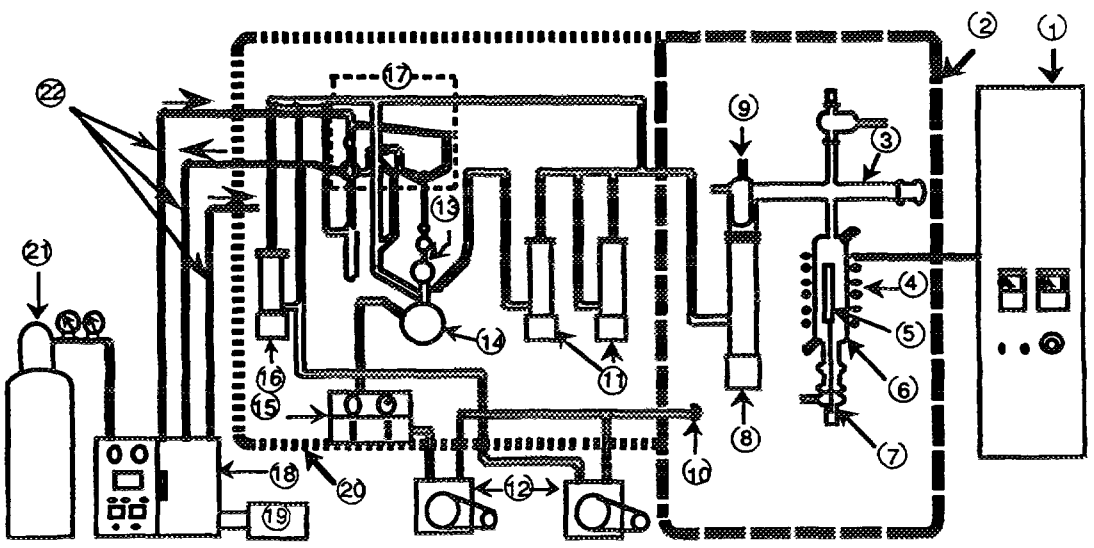

Fig.3. Schematic diagram of gas extraction and analysis apparatus.

①,High frequency induction equipment; ②,Glove box; ③,Pellets loading part; ④,Induction coil; ⑤,Molybdenum crucible; ⑥,Quartz furnace tube; ⑦,Pellets unloading part; ⑧,Mercury diffusion pump ; ⑨,Mercury trap; ⑩,Dust filter; ⑪,Mercury diffusion pump ; ⑫,Mechanical pump; ⑬,McLeod gauge; ⑭,Toepler pump; ⑮,Toepler pump controller; ⑯,Oil diffusion pump; ⑰,GSU; ⑱,Gas chromatograph; ⑲,Integrator; ⑳,Open port box; ㉑,Helium cylinder; ㉒,Helium carrier gas.

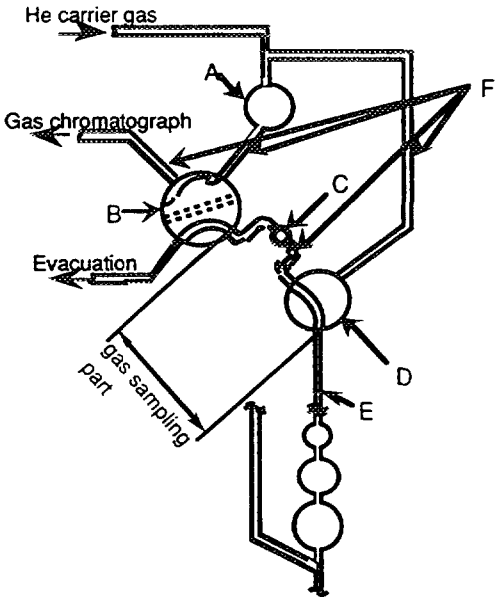

Fig.4. Gas sampling unit.

A, Spherical buffer vessel; B, Improved four-way valve;C, Reference gas injection port; D,Improved three-way valve;E, MacLeod gauge; F, Glass capillary tube of 2mm i.d.

3.1.2 Procedure of gas analysis by the gas chromatograph

The gases extracted could be simultaneously analyzed by the combination of several stainless steel columns of Porapak Q, Molecular Sieve 5A and Chromosorb W. When the objective gases were injected into the column packed with Porapak Q using the GSU, these gases were separated into two groups. The gases of both groups were injected into twin separation columns in a computer controlled flow through a changing valve. The gases of the first group (H_2, O_2, N_2 and CO) were separated by the column packed with Molecular Sieve 5A, and those of the other group (CH_4, CO_2 and C_2H_6) by the column packed with Chromosorb W. Each of the separated gases was detected by the PID. A schematic diagram of the gas chromatography is shown in Fig. 5.

3.2.3 Measurement procedure

The weighed sample (about 1g) was transferred into the pellets loading part of the apparatus, which was evacuated to approximately 10^{-2} Pa using the mercury diffusion pump and the mechanical pump. The molybdenum crucible without the sample was outgassed beforehand until less than 10μ 1 at the S.T.P. condition. Then the sample was dropped into the molybdenum crucible and heated at 1700°C for 30 min. The released gas from the sample was corrected to the McLeod gauge using the mercury diffusion pump and the toepler pump, where the temperature and pressure were measured. The total gas volume of the sample was calculated from the temperature, pressure and volume of the collected gas at the S.T.P. condition. Volumetrically measured gas by the McLeod gauge was transferred into the GSU. The extracted gas was injected into the gas chromatograph at the same pressure as the carrier gas, and analysed by the PID. The total volume of the gas occluded per unit mass in the sample at the S.T.P. was obtained as the sum of all components of the gas analysed.

3.2 RESULTS

3.2.1 Determination of occluded gases in MOX fuel pellets

Occluded gases in the MOX fuel pellets were quantitatively and qualitatively analysed. As a results of the standard gas analysis and verification of the retention time, the peaks were qualitatively

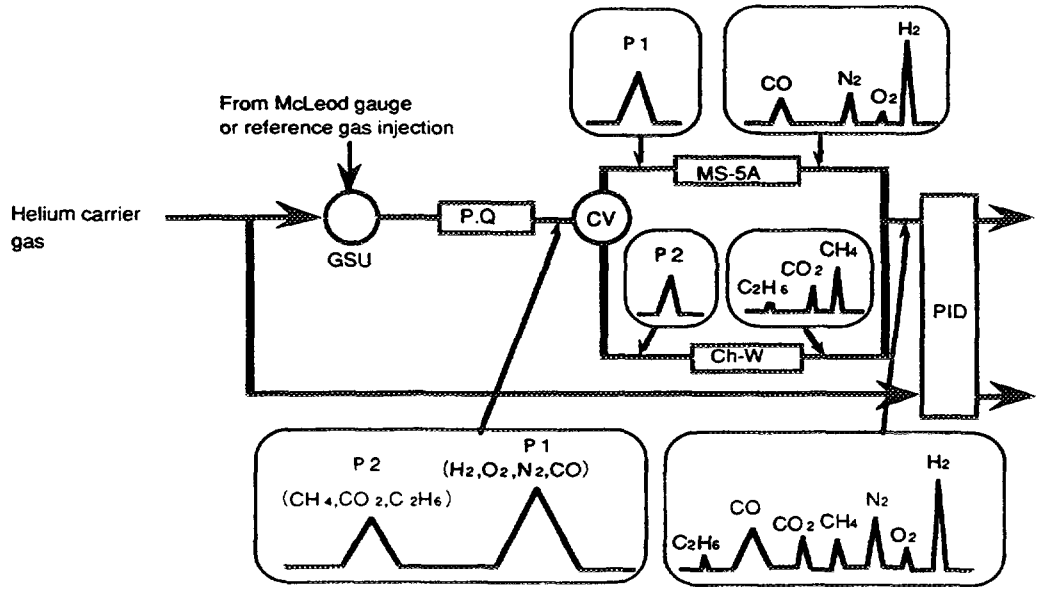

Fig.5. Principle of gas separation using gas chromatograph.

GSU,gas sampling unit; MS-5A, Separation column with Molecular Sieve 5A
CV, Flow through changing valve; Ch-W, Separation column with Chromosorb W,
P.Q, Separation column with Porapak Q; PID, Photoionization detector.

analysed as H_2, O_2, N_2, CH_4, CO_2 and CO. Further, concentrations of the objective gases in the MOX fuel pellets were quantitatively determined. The obtained results are shown in Table 3. From these experiments, as to the sintered pellets in hydrogen-nitrogen (5% - 95%) atmosphere, H_2, N_2 and CO were found to be main component gases. On the other hand, for the sintered pellets in hydrogen-argon (5% - 95%) atmosphere, H_2 and CO were the main component gases. C_2H_6 was not detected . A typical gas chromatogram of occluded gases in the MOX fuel pellets is shown in Fig.6.

TABLE 3 Analytical results of occluded gases in MOX fuel pellets

Lot No.	Sintering Atmosphere	Found [a] (μl/g)						
		H_2	O_2	N_2	CH_4	CO_2	CO	C_2H_6
1.	H_2 - N_2 (5% - 95%)	46	--[b]	20	--[b]	--[b]	9.4	ND[c]
2.	H_2 - Ar (5% - 95%)	14	--[b]	--[b]	--[b]	--[b]	1.9	ND[c]
3.	H_2 - Ar (5% - 95%)	14	--[b]	--[b]	--[b]	--[b]	12	ND[c]
4.	H_2 - N_2 (5% - 95%)	93	0.5	38	--[b]	--[b]	15	ND[c]
5.	H_2 - N_2 (5% - 95%)	21	1.3	30	--[b]	--[b]	11	ND[c]

a At S.T.P. condition
b Less than the minimum limit of determination
c Not detected

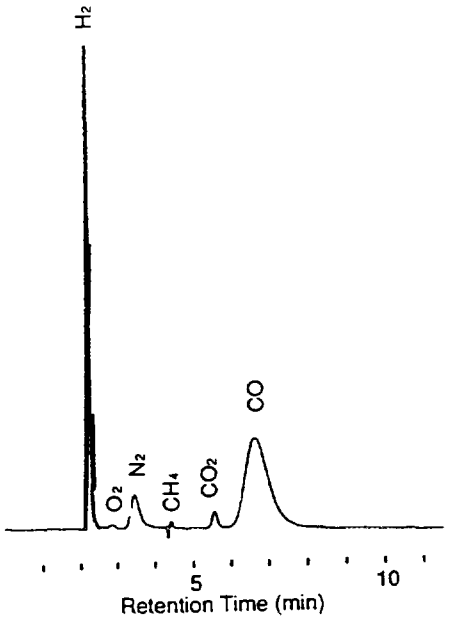

Fig.6. Typical gas chromatogram of occluded gases in MOX fuel pellets.

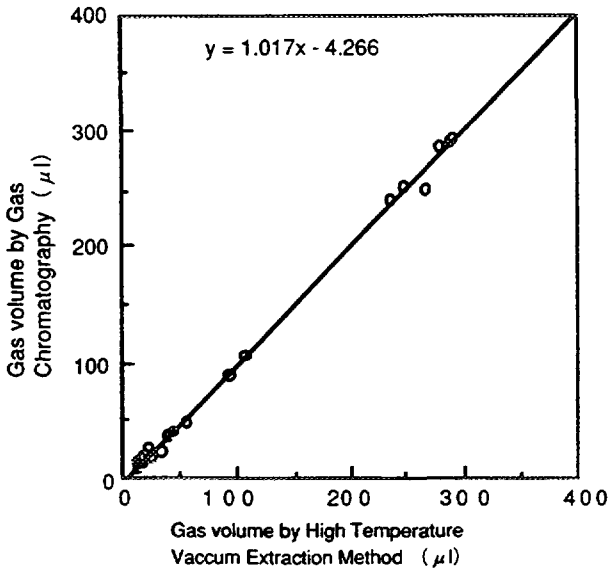

Fig.7. Correlation diagram of high temperature vaccum extraction method and extracted gas analysis by gas chromatography.

3.2.2 Comparison of the high temperature vacuum extraction method and the gas chromatography

The total volume of the occluded gases was determined for a large number of samples by the high temperature vacuum extraction method and by the present extracted gas analysis using the gas chromatography. The pellets used in this study were those fabricated for the purpose of production test of the MOX fuels. The correlation diagram of both methods is shown in Fig. 7. The correlation coefficient value was 0.998 (n = 36). The results of the present analysis of extracted gas using the gas chromatography were in good agreement with those of the high temperature vacuum extraction method in the range of less than 300μl. Therefore, this method is applicable not only to MOX pellets but also to UO_2 pellets for the analysis of occluded gas below 300μl.

4. DETERMINATION OF NITROGEN IN MOX BY GAS CHROMATOGRAPHY AFTER INERT GAS FUSION

This technique is based on chromatographic determination of nitrogen in released gases from fused sample. The sample and iron metal flux in a graphite crucible were heated to 2700°C by an impulse furnace under a helium atmosphere. From the fused sample, which was gases evolved such as H_2, N_2 and a large amount of CO . The CO produced was interfered with the measurement of N_2. Consequently, for N_2 separation from other gases and CO removed, a gas chromatograph equipped with a pre-cut system was originally developed. Nitrogen separated was determined using a thermal conductivity detector (TCD). The RSD was less than 5%, and the time required for one determination was about 10 min.

4.1 EXPERIMENTAL

4.1.1 Apparatus

The schematic diagram of the apparatus is shown in Fig. 8. An impulse furnace and a gas chromatograph equipped with the pre-cut system and the TCD were enclosed in the glove-box. The gas chromatograph was installed in the thermostat at 65.5°C. The pre-cut system consisted of 500mm × 4mm i.d. stainless steel columns packed with Porapak N of 60 ~ 80 mesh (C-l) and 600mm × 4mm i.d.

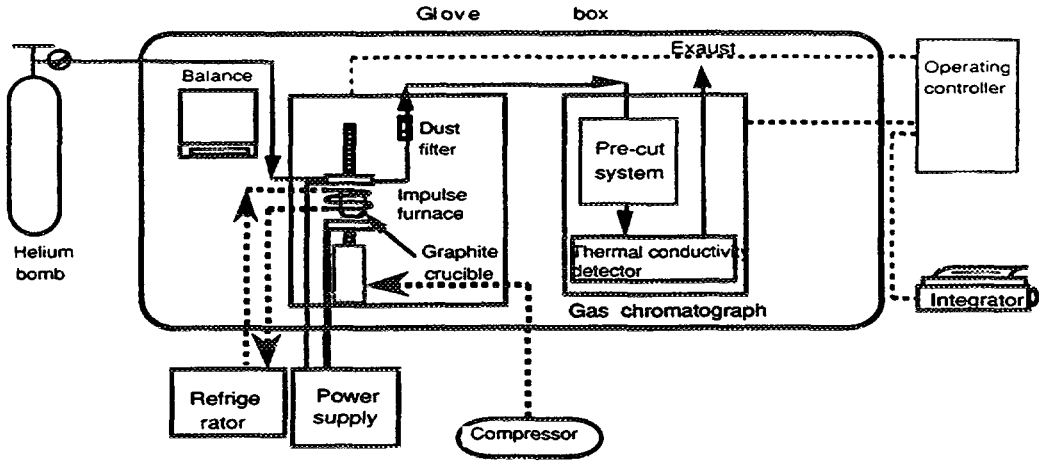

Fig.8. Schematic diagram of nitrogen analysis apparatus.

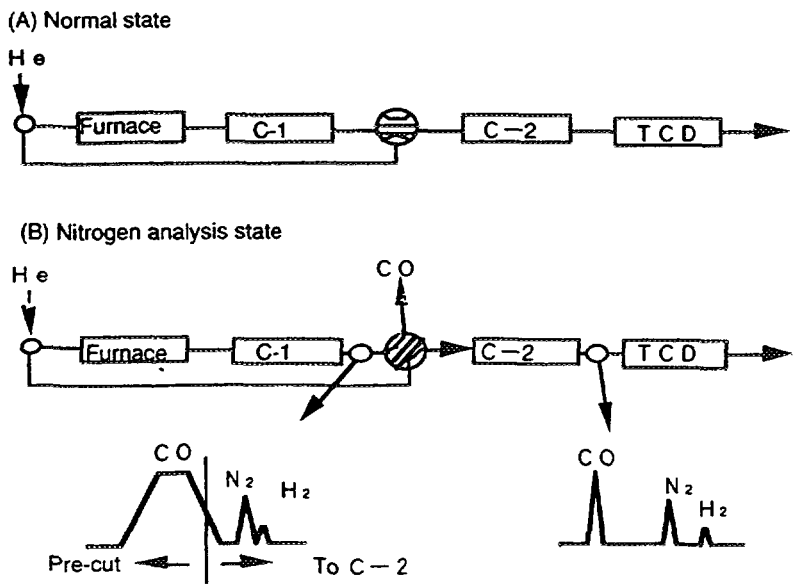

Fig.9. Schematic principle diagram of pre-cut system.
C-1, Porapak N column; C-2, Silica Gel column;
TCD,Thermal conductivity detector.

stainless steel columns packed with Silica gel of 60 ~ 80 mesh (C-2) and a carbon monoxide purge valve. The schematic diagram of the precut system is shown in Fig. 9. A computerized peak-area integrator from Shimazu Co., Ltd. CR-3A (Kyoto, Japan) was used.

4.1.2 Procedure

The sample was placed into a degassed graphite crucible, weighed to an accuracy of 0.1mg and the metal flux was added. The crucible was placed in the impulse furnace, and it was heated up to 2700°C for 30 sec under a helium atmosphere. Gases released from the sample were carried to the pre-cut system by the helium carrier gas. N_2 was determined by the TCD, and the chromatogram peak-area of N_2 was integrated by the computerized peak area integrator. The concentration of N_2 in the iron powder was analysed beforehand, and this blank value was subtracted from the sample analysis value. The calibration curve for N_2 was made using the N_2 standard gas or the standard materials.

4.2 RESULTS AND DISCUSSION

4.2.1 Calibration curve

The Japanese Standards of Iron and Steel (JSS) were used as the standard materials, which were purchased from the Iron and Steel Institute of Japan. The certified nitrogen analysis values of JSS 366-6, JSS 023-3, JSS 368-6 and JSS 153-7 were 9ppm, 39ppm, 129ppm and 165ppm, respectively.

The calibration curve for the determination of nitrogen was prepared using the standard materials. The calibration curve obtained was linear in the range of 9 to 165ppm.

4.2.2 Comparison of the Kjeldahl method with the present method

In order to compare the Kjeldahl method with the present method, the two sintered pellets used in this study were fabricated for the purpose of production testing of the MOX fuels. The N_2 contents in these pellets were adjusted to be higher than that generally used in the MOX fuels. The plutonium contents of two pellets were 2% for the sample A and 30% for the sample B. The analytical results of N_2 values for sample A and sample B are summarized in Table 4. In both methods, the mean values and the standard deviation were 65.3 ± 9.2ppm and 77.1 ± 3.1 ppm for sample A and 152.5 ± 18.7ppm and 156.6 ± 7.4ppm for sample B, respectively. The results obtained by the Kjeldahl method were lower than those of this method. The RSD of this method were less than 5%.

In the Kjeldahl method, which is based on complete conversion to ammonia ions of the N_2 contained in the sample, the dissolution process for the sample is very important. For uranium metal, uranium dioxide and uranium nitrides, Lathouse et al. have shown that loss of N_2 can occur in the dissolution process for the Kjeldahl method [6]. For uranium nitride composed of the UN phase, Takahashi has shown that the results obtained by the Kjeldahl method is lower by about 5% than those obtained by the Dumas method [7]. Compared to the uranium dioxide pellets, the MOX fuel is slightly soluble in acid solutions because of the coexistence of plutonium, and loss of N_2 can occur in the dissolution process. Therefore, the analytical results of N_2 by the Kjeldahl method can show a low value compared to the present method.

TABLE 4 Comparison of the Kjeldahl method with the present method

Method	Sample No.	Nitrogen found, ppm[a]
Kjeldahl method	A	65.3 ± 9.2
	B	152.5 ± 18.7
Present method	A	77.1 ± 3.1
	B	156.6 ± 7.4

a Mean ± standard deviation, 10 results

4. CONCLUSION

(1) The O/M analysis method for MOX fuel was developed using nondispersive infrared spectrophotometry (NDIR) after inert gas fusion. The result of the present method was in good agreement with the gravimetric methods. The RSD was less than 0.20%. and the time required

for one determination was about 10min for all analytical operations. This method is applicable to sintered MOX fuels over a wide range of plutonium content.

(2) The gases were simultaneously determined using an originally developed GSU, the improved gas chromatograph equipped with several separation columns and PID. The RSD were 1.0, 3.4, 0.6, 1.0, 1.4, 0.8 and 0.9% for H_2, O_2, N_2, CH_4, CO_2, CO and C_2H_6, respectively. The gases H_2, O_2, N_2, CH_4, CO_2 and CO from the MOX fuel pellets were simultaneously determined. The results of gas analysis using this method were in agreement with that of the high temperature vacuum extraction method in the range less than 300μl of extracted gas .

(3) The nitrogen analysis method for MOX fuel was developed using a gas chromatograph equipped with a pre-cut system and TCD. The RSD was less than 5%, and the time required for one determination was about 10 min.

5. FUTURE PLANS

In order to put these analysis technologies for MOX to practical use, further improvements are necessary. Particularly, the developments of automatic handling system, simultaneous determination methods for impurity and more accurate methods need to be proceed.

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SESSION 3
MOX FUEL DESIGN

MOX FUEL DESIGN AND DEVELOPMENT CONSIDERATION

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Abstract

Pu thermal utilization in Japan will be realized in several plants in late 1990's, and will be expanded gradually. For this target, adequacy of methods for MOX fuel design, nuclear design, and safety analysis has been evaluated by the committee of competent authorities organized by government in advance of the licensing application. There is no big difference of physical properties and irradiation behaviors between MOX fuel and UO_2 fuel, because Pu content of MOX fuel for Pu thermal utilization is low. The fuel design code for UO_2 fuel will be applied with some modifications, taking into account of characteristic of MOX fuel. For nuclear design, new code system is to be applied to treat the heterogeneity in MOX fuel assembly and the neutron spectrum interaction with UO_2 fuel more accurately. For 1/3 MOX fueled core in three loop plant, it was confirmed that the fuel rod mechanical design could meet the design criteria, with slight reduction of initial back-filling pressure, and with appropriate fuel loading patterns in the core to match power with UO_2 fuel. With the increase of MOX fuel fraction in the core, control rod worth and boron worth decrease. Compensating the decrease by adding control rod and utilizing enriched B-10 in safety injection system, 100% MOX fueled core could be possible. Up to 1/3 MOX fueled core in three loop plant, no such modification of the plant is necessary. The fraction of MOX fuel in PWR is designed to be less than 1/3 in the present program. In order to improve Pu thermal utilization in future, various R & D program on fuel design and nuclear design are being performed, such as the irradiation program of MOX fuel manufactured through new process to the extent of high burnup.

1. INTRODUCTION

Japanese government announced a long-term plan conceming nuclear utilization in June, 1994, where the prospect for Pu thermal utilization is also described; i.e., Pu thermal utilization will be realized in several plants in late 1990's, and will be expanded to approximately ten plants around 2000. It is foreseen more than ten plants of Pu thermal utilization in the next period from 2000 to 2010.

For this target, adequacy of methods for MOX fuel mechanical design, core nuclear design, and safety analysis has been evaluated by the committee of competent authorities organized by government in advance of the licensing application.

There is no big difference of physical properties and irradiation behaviors between MOX fuel and UO_2 fuel, because Pu content of MOX fuel for Pu thermal utilization is low compared to MOX fuel for fast reactor. Taking into account of characteristic of MOX fuel, the fuel mechanical design code for UO_2 fuel will be applied with some modification. For nuclear design, the new code system will be used to treat the heterogeneity in MOX fuel assembly and the neutron spectrum interaction with UO_2 fuel more accurately.

This paper describes the status and future plan of fuel design, core design, and research and development for MOX fuel.

2. OUTLINE OF PWR MOX FUEL ASSEMBLY

All fuel rods within a PWR MOX fuel assembly are made of MOX, with the distribution of Pu content of fuel rod to reduce local power peaking at the periphery of the assembly. An example of 17 x 17 type fuel is shown in Fig.1.

The average Pu fissile content of MOX fuel assembly is designed so that the reactivity of MOX fuel is equivalent to that of current standard UO_2 fuel for Japanese PWR's whose enrichment is 4.1wt%. Typical plutonium fissile and total content as a function of fissile isotope fraction is shown in Fig.2. The Pu fissile content is evaluated approximately 6wt% for the Pu isotopic composition described in Fig.2, which is considered typical of Pu recovered from PWR spent fuel. Maximum assembly average burnup of UO_2 fuel is 48GWd/t, while that of MOX fuel assembly is designed lower to be 45GWd/t. It is due to large peak to average burnup ratio in MOX fuel assembly.

The other structure of MOX fuel assembly is almost same as that of UO_2 fuel assembly to be compatible with UO_2 fuel assemblies in the core. And also, from a view point of keeping reliability as

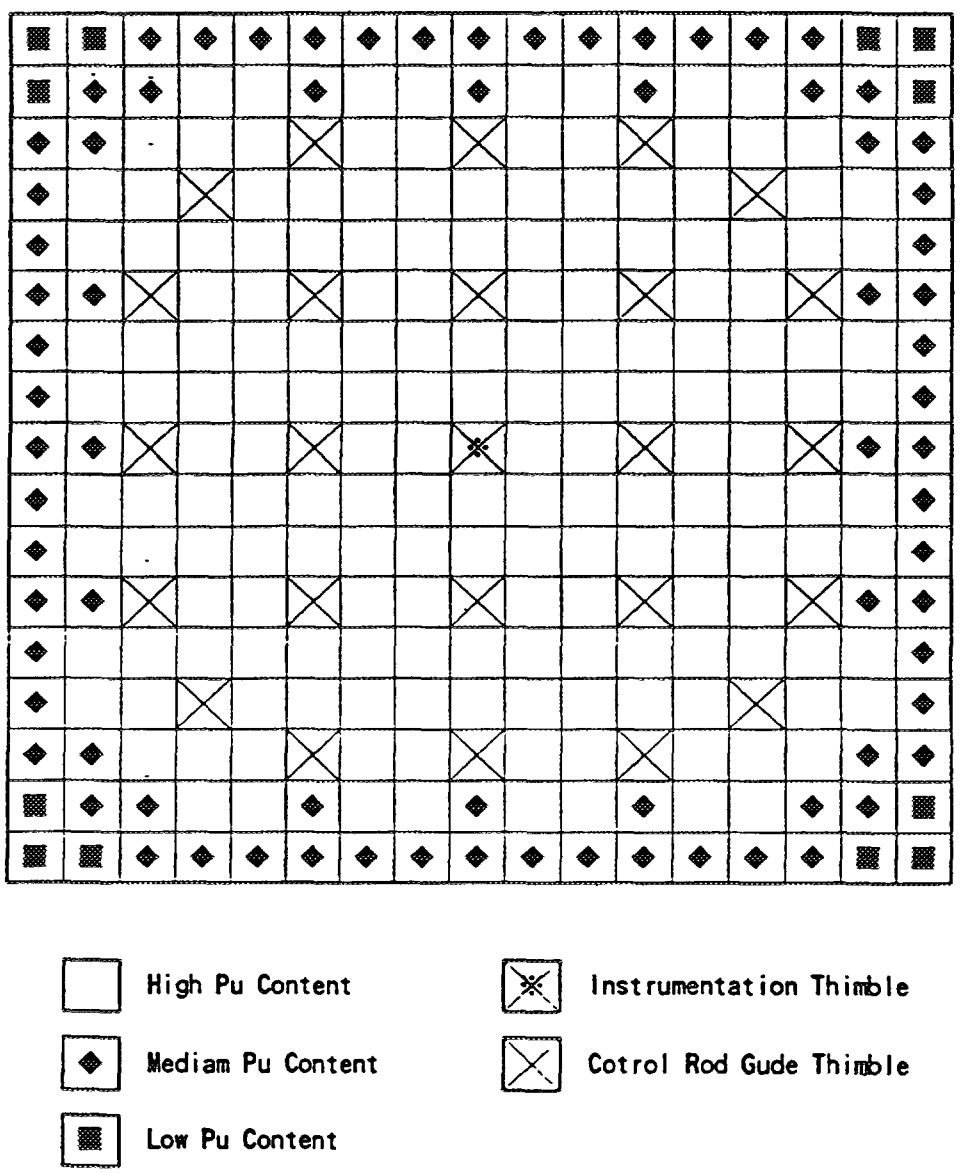

FIG. 1. Pu Content Distribution in PWR MOX Fuel Assembly

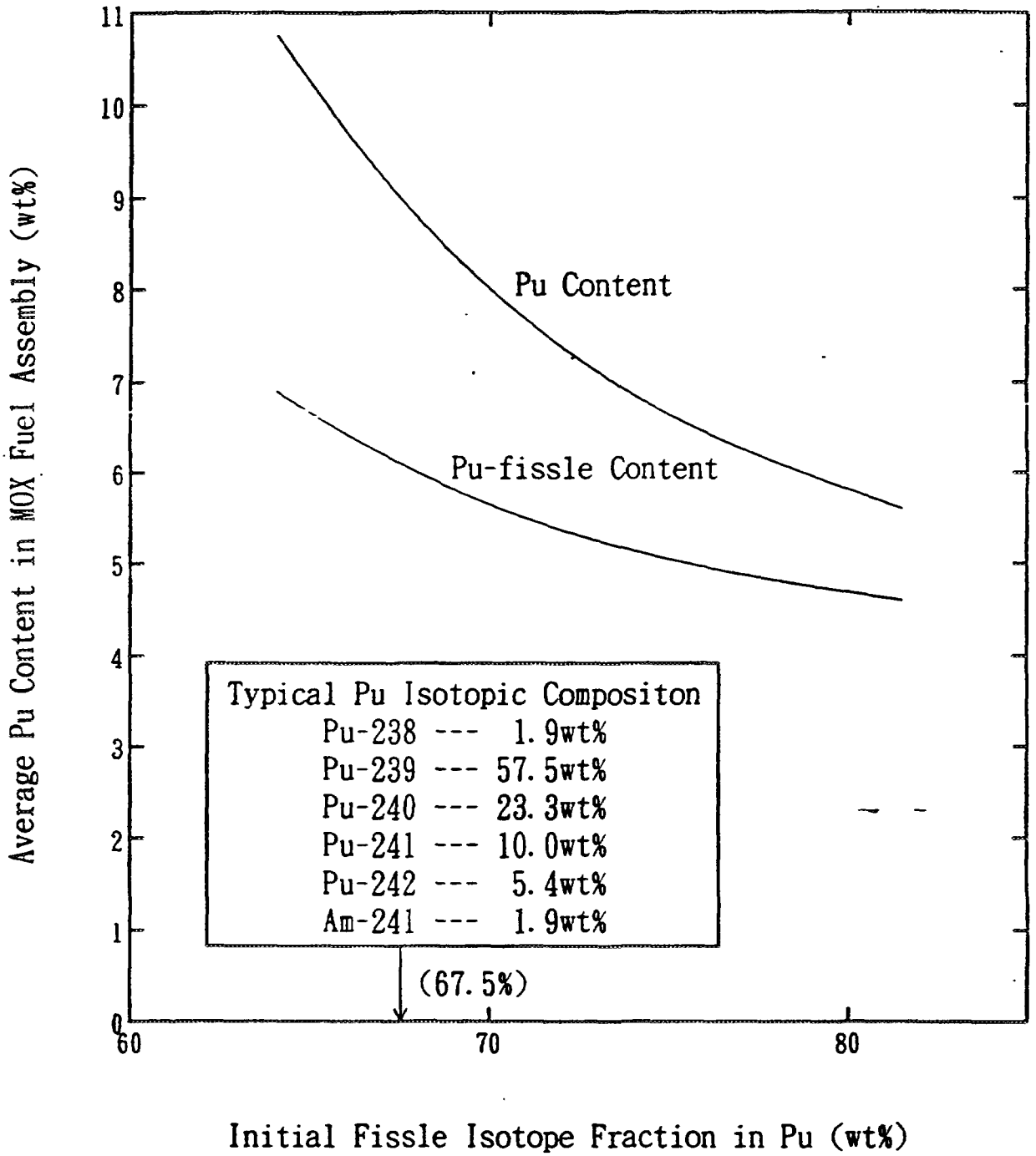

FIG. 2. Typical Pu Content vs Fissile Isotope Fraction

high as at present, it is planned that all components except for MOX pellets will be supplied by domestic vendors to overseas manufactures, who will fabricate MOX fuels for certain time period. The basic description of PWR MOX fuel is shown in TABLE 1.

3. FUEL MECHANICAL DESIGN OF MOX FUEL

3.1 MOX CHARACTERISTICS AND DESIGN CONSIDERATIONS

PuO_2 and UO_2 form solid solution, and material properties of MOX are very similar to those of UO_2 to approximately 13wt% of Pu content. Two of the most important properties, melting point and

thermal conductivity are reduced by only several percent [1]. Thermal conductivity is illustrated in Fig. 3 with UO_2 design model, and small reduction appears in MOX fuel. Thermal expansion of MOX fuel is very close to that of UO_2.

TABLE I **Fuel Description of PWR**
(Typical Specification for 17 x 17 Fuel Assemblies)

		UO_2 Fuel	MOX Fuel
1. Fuel Assemblies			
Rod Array		17 x 17	17 x 17
Active Fuel Height,	m	~3.6	~3.6
Rods per Assemblies		264	264
U-235 Enrichment,	wt%	4.1 (2.6 in Gd fuel)	~0.2
Average Pu-fissile Content,	wt%	--	~6.1
Maximum burnup,	GWd/t	48	45
2. Fuel Rods			
Fuel Pellets Diameter,	mm	8.19	8.19
Fuel pellets Material		UO_2 UO_2 - Gd_2O_3	UO_2 - PuO_2
Clad Outside Diameter,	mm	9.50	9.50
Clad Thickness,	mm	0.57	0.57
Clad Material		Zircaloy-4	Zircaloy-4
Pellet--Clad Diameter Gap,	mm	0.17	0.17

Fission gas release data are illustrated in Fig.4 [2,3,4,5]. It is said that fission gas release of MOX fuel is larger than that of UO_2 fuel due to inhomogeneity and possibly microstructure differences. Particularly, MOX pellet which were manufactured through old process have considerable amount of Pu enriched spots. These spots burn faster than surrounding MOX matrix, and the temperature of these area is also higher and therefore more fission gas is released. However fission gas release of current MOX fuel which were manufactured through advanced process reduced to similar level of UO_2 fuel. In fuel rod design, it is conservatively assumed that fission gas release from MOX fuel is higher than that of UO_2 in order to bound old data also. These data were analyzed by fuel design code FINE [6], which is one of Japanese codes developed by Mitsubishi. Comparison of predictions with measurement are illustrated in Fig. 5 and shows good agreement.

Fuel stack length changes are illustrated in Fig. 6 [2,4] which shows that MOX fuel data stay within variation of UO_2 fuel data. From these data, it can be said that addition of PuO_2 hardly affects on pellet densification and swelling, and the behavior is rather dependent on UO_2 characteristics.

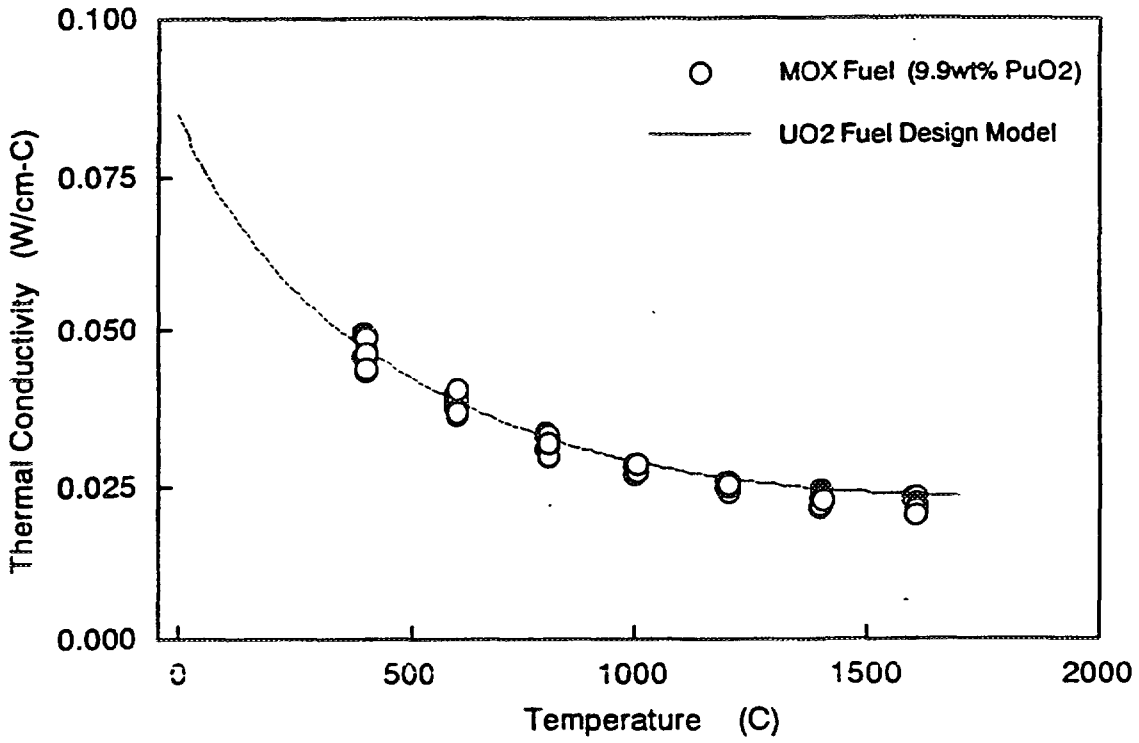

FIG. 3. Thermal Conductivity of MOX Pellet

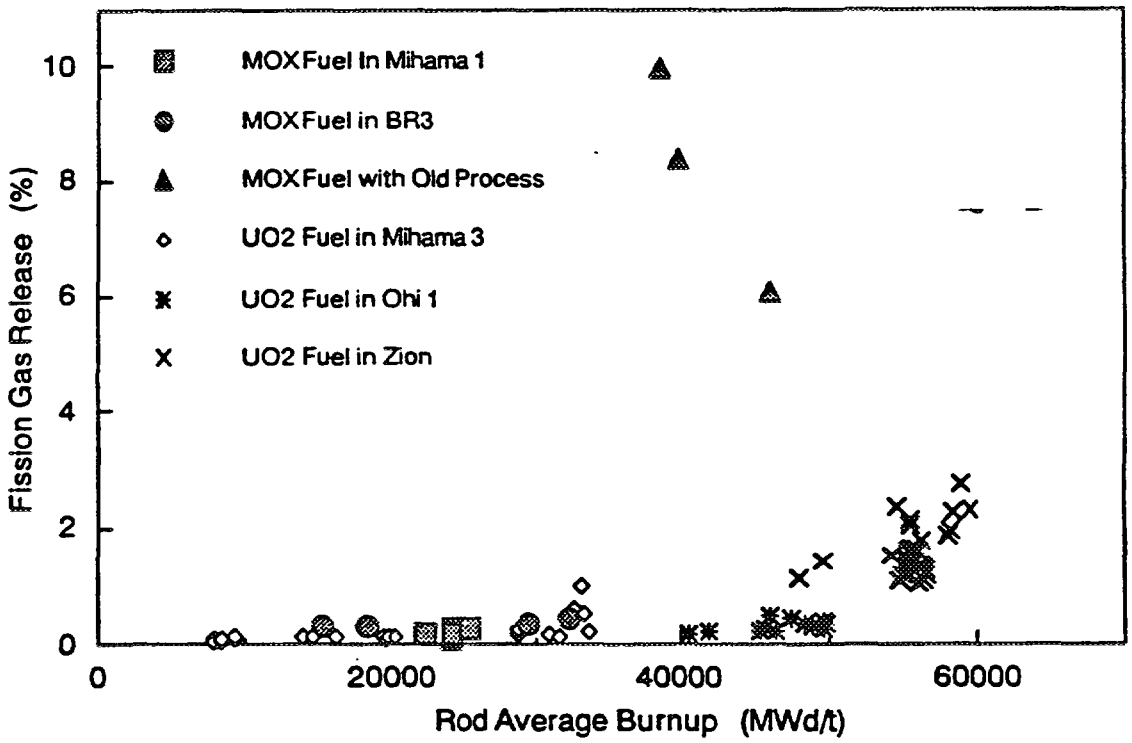

FIG. 4. Fission Gas Release

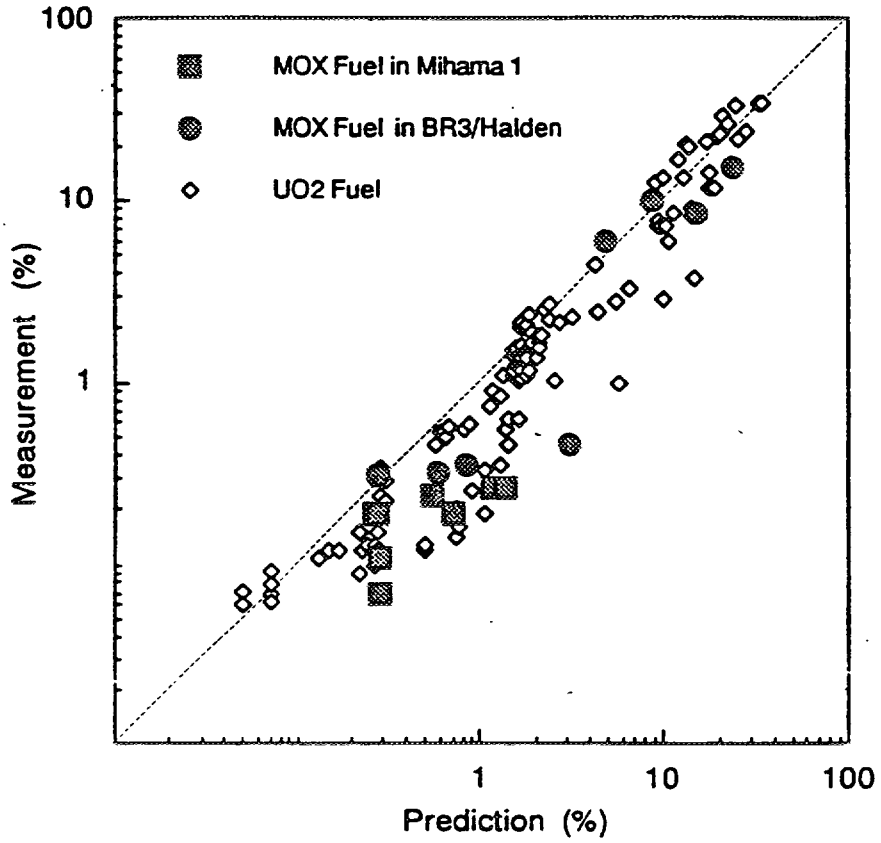

FIG. 5. Comparison of Fission Gas Release

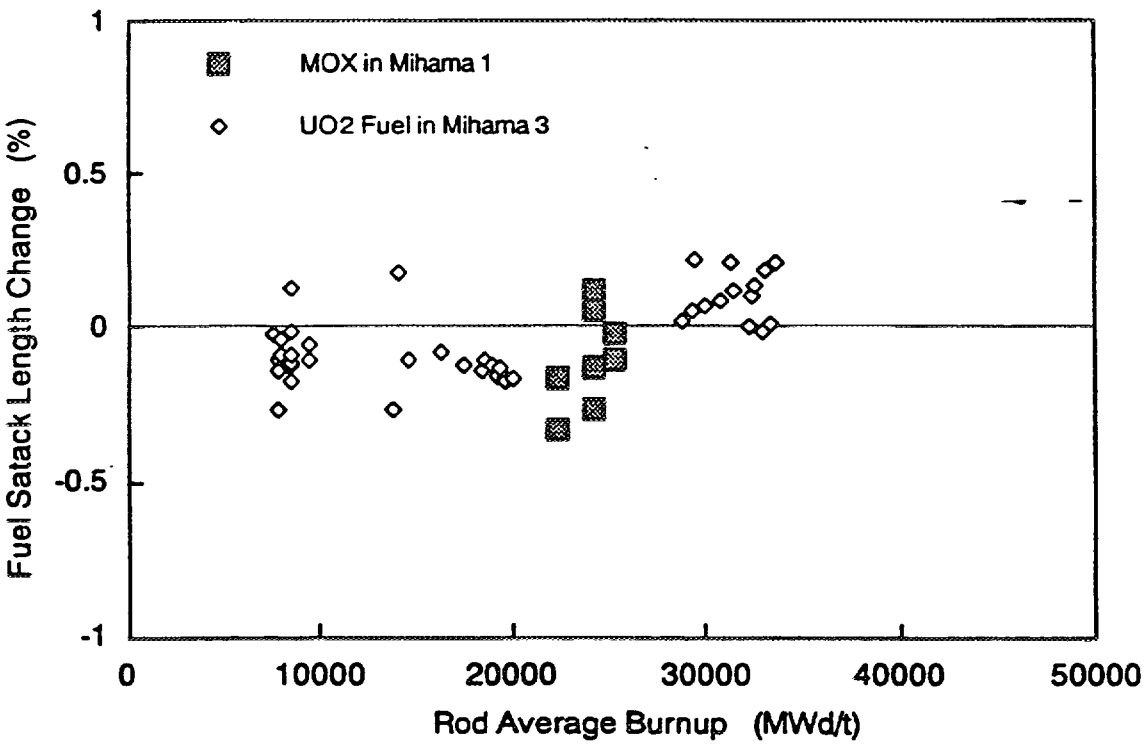

FIG. 6. Fuel Stack Length of Changes

More Helium is produced in MOX fuel through α-decay. This Helium production and release from pellet increase fuel rod internal pressure. This is modelled in the fuel design code.

Based on these considerations, the fuel design code can fairly predict fuel rod internal pressure and fuel temperature as illustrated in Fig. 7 and Fig. 8, and both of them show good agreement.

3.2 FUEL DESIGN FEATURES

Nuclear characteristics of MOX fuel is slightly different from that of UO_2. Reactivity of MOX fuel decreases slower than that of UO_2, and then power of MOX fuel remains higher at high burnup. This also causes higher internal pressure in MOX fuel. Considering also the characteristics of MOX fuel as described in the last section, fuel rod internal pressure is important. As the results of the analysis for 1/3 MOX in three loop plant, taking into account the fuel and nuclear characteristics of MOX, it was confirmed that fuel rod intemal pressure could meet the design criteria with slight reduction of initial back-filling pressure, and with appropriate fuel assembly loading pattems in the core to match power with UO_2 fuel.

4. NUCLEAR DESIGN OF MOX FUEL

4.1 METHOD OF NUCLEAR DESIGN

For nuclear design of MOX fueled core, the new code system has been used. PHOENIX--P/ANC [7, 8] is one of the Japanese code systems applied by Mitsubishi. The accuracy of the new code system has been verified by comparing the calculation results with the measurement of critical experiments and core operating data.

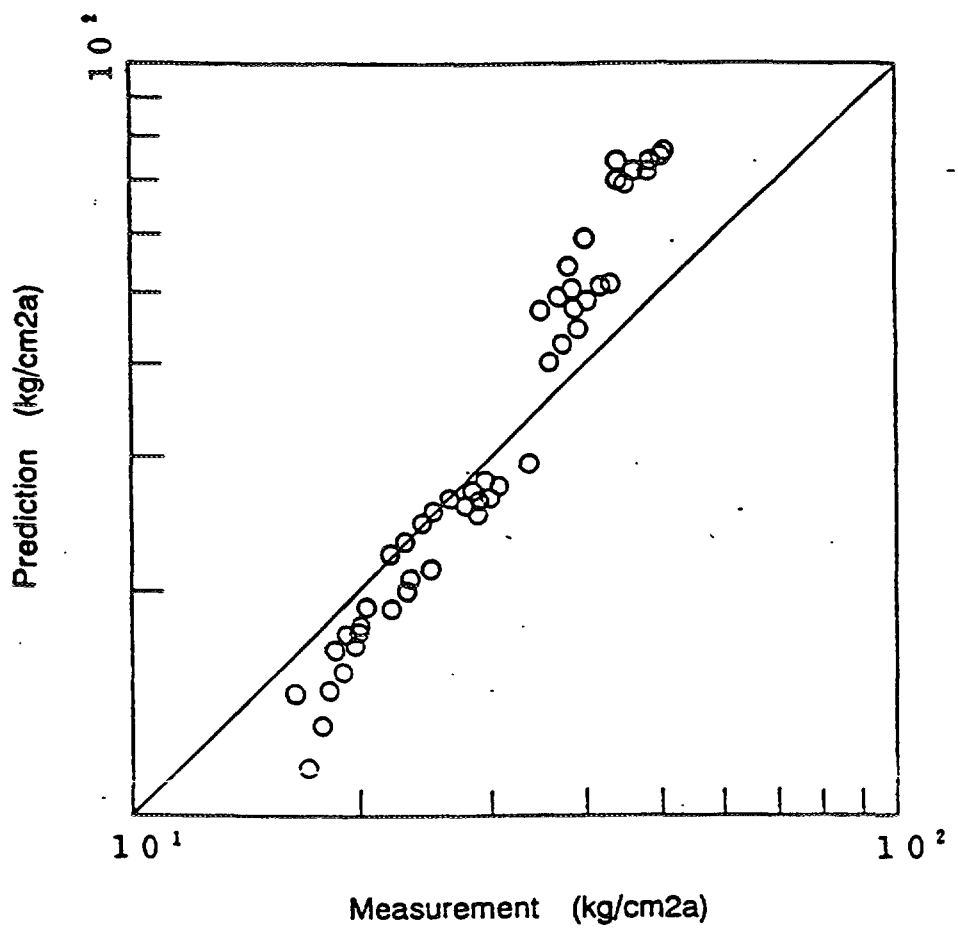

FIG. 7. Comparison of Rod Internal Pressure

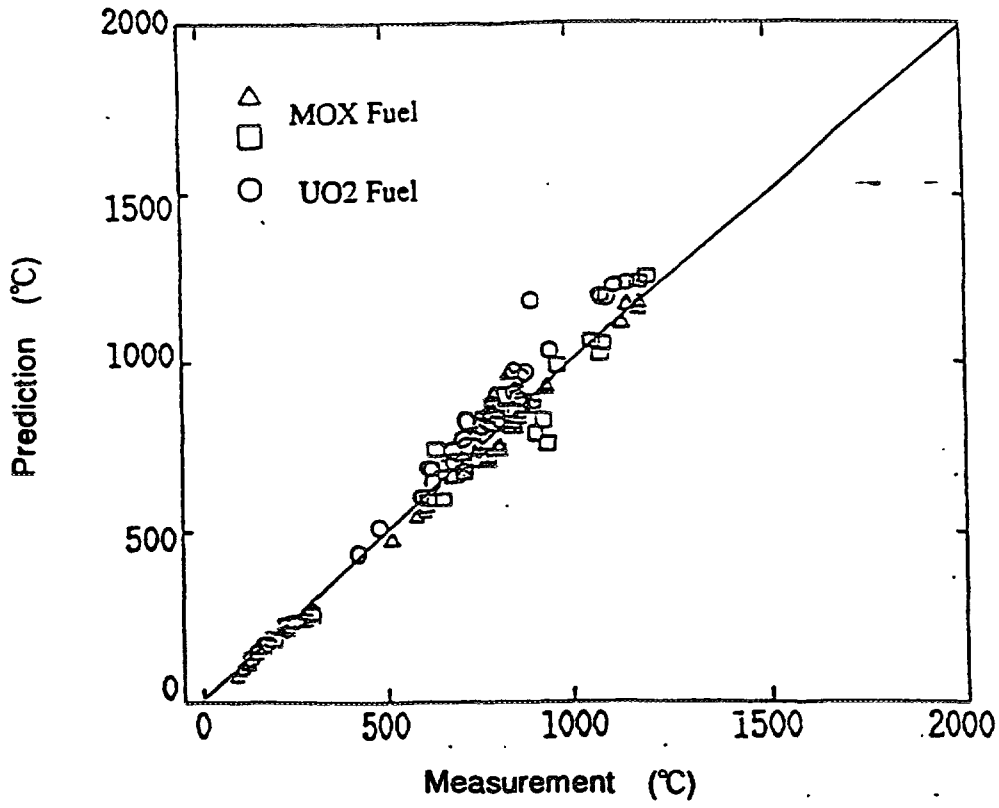

FIG. 8. Comparison of Fuel Centre Temperature

4.2 CHARACTERISTICS OF MOX FUELED CORES

With the increase of MOX fuel fraction, some core physics parameters change from UO_2 fueled core. Typical variation of these parameters are shown in Fig. 9. The reactivity effect of control rod and boron in the coolant decrease with MOX fuel fraction. To 1/3 MOX fuel fraction, degradation of control rod worth can be mitigated by loading MOX fuel avoiding the control rod position. To increase MOX fuel fraction more than 1/3, it will be necessary to increase the number of control rods or change the rod absorber material from Ag-In-Cd. Decrease of boron worth can be compensated by increasing the boron concentration in safety injection system to 1/3 MOX. To increase MOX fuel fraction more than 1/3, it will be necessary to utilize enriched B-10 in safety injection system. Moderator temperature coefficient becomes more negative, causing larger reactivity insertion during cool down event, but that can be overcome by strengthening the ability of control rod and safety injection system as described above. Delayed neutron fraction becomes smaller with the increase of MOX fuel fraction. But the decrease of control rod worth and power peaking factor will mitigate the results of reactivity insertion event.

Comparison of typical core design parameters of UO_2 fueled core, 1/3MOX, and 100% MOX fueled core for three loop PWR plant is shown in Table II. For 1/3MOX fuel core, fuel assemblies with three plutonium fissile contents as shown in Fig.1 are used to match with adjacent UO_2 fuel and reduce power peaking factor in the assembly. On the other hand, single plutonium fissile content in the fuel assembly can be used for 100% MOX core. Many burnable poison rods are necessary to adjust power distribution especially for fuel rod internal pressure in 1/3MOX fueled core, while no burnable poison is needed in 100% MOX fueled core. Addition of control rods and utilization of enriched B-10 in refueling water storage tank and boron injection tank, will make 100% MOX fueled core possible.

Up to 1/3MOX fueled core in three loop plant, no such modification of the plant is necessary. Consequently, the fraction of MOX fuel in PWR is designed to be less than 113 in the present program.

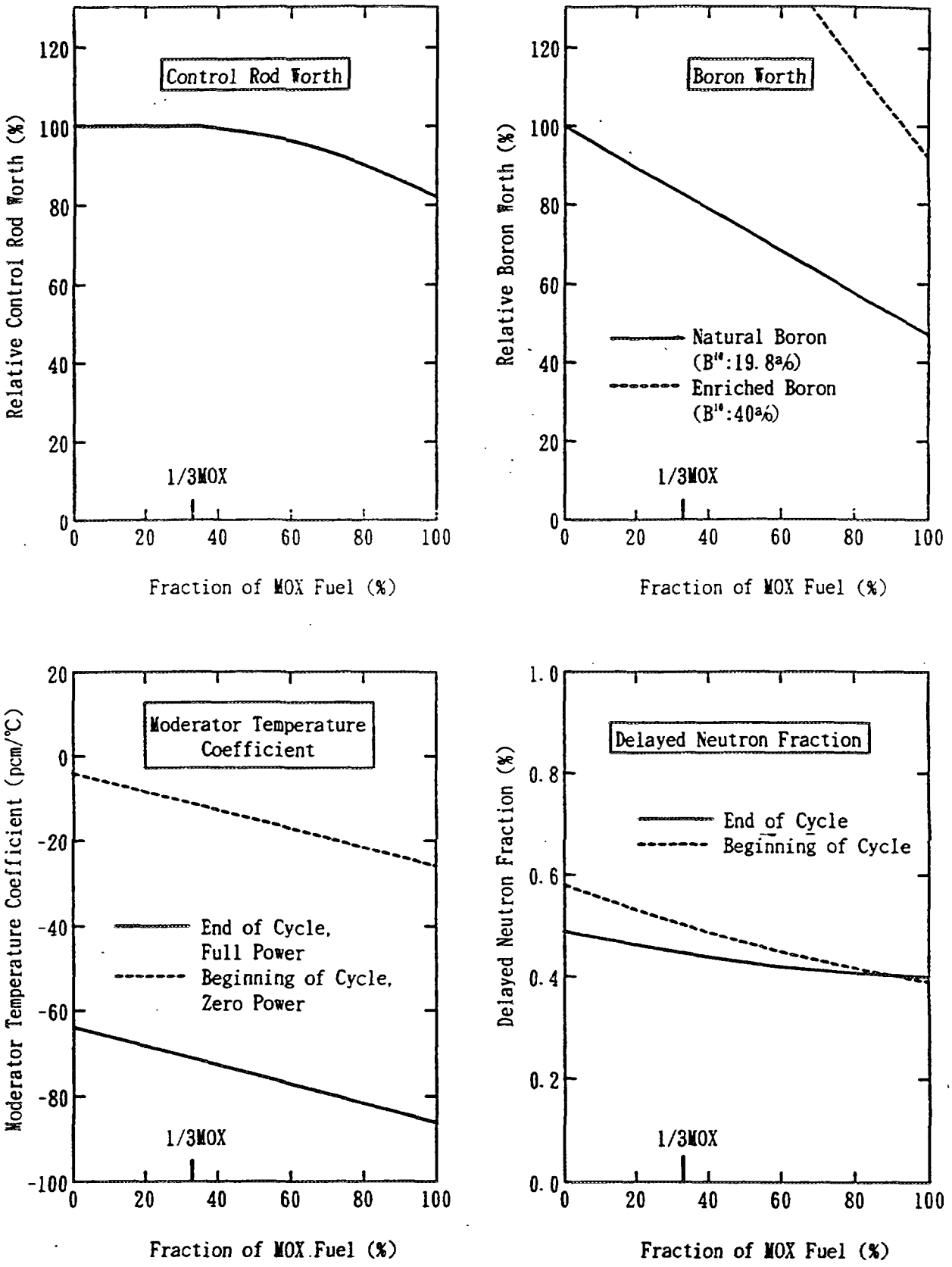

FIG. 9. Dependence of Nuclear Design Parameters on Fraction of MOX Fuel in the Core

TABLE II Characteristics of MOX Fueled Core and UO_2 Fueled Core

	UO_2Core	1/3 MOX Core	1/1 MOX Core
Number of total fuel assemblies	157	157	157
Number of fresh fuel assemblies			
UO_2 Fuel	24	28	0
UO_2 Fuel with gadolinia	36	16	0
MOX Fuel	0	16	60
Total	60	60	60
Number of burnable poison assemblies	0	52	0
Number of Pu fissile content in MOX fuel assemblies	--	3	1
Cycle burnup [GWd/t]	15.2	15.2	15.2
Maximum burnup of fuel assemblies			
UO_2 fuel [Gwd/t]	48	48	--
MOX fuel [Gwd/t]	--	45	48
Number of control rod clusters			
In the case of Ag-In-Cd rod	48	48	~68
In the case of B4C rod with 90% B10	--	--	~60
Boron concentration in refueling water storage tank			
In the case of natural boron [ppm]	2200	~3000	~6000
In the case of 60% B10 [ppm]	--	--	~2000
Boron concentration in boron injection tank			
In the case of natural boron [ppm]	20000	-21000	~60000
In the case of 60% B10 [ppm]	--	--	~20000

5. FUTURE DESIGN CONSIDERATION OF PWR MOX FUEL

It is considered very important to make further efforts on MOX fuel R & D in order to enhance Pu thermal utilization in more flexible and economical design in future. The following design considerations are now being investigated and planed.

5.1 REVISION OF FISSION GAS RELEASE (FGR) MODEL

The present data base of MOX fuels includes the data of MOX fuels manufactured through the old mechanical blending processes whose homogeneity of Pu and U is poor and FGR is large.

This fact causes high FGR on the design of MOX fuel. However, the MOX fuels to be used in Japanese PWR's will be manufactured through new processes, which gives the improved homogeneity of Pu and U and are expected to have lower FGR. The irradiation data of MOX fuels manufactured through the new processes are now being accumulated to revise FGR model.

It is hereby important to obtain the data at high bumup and high power, considering the design conditions for the further MOX fuel utilization. The data for the peak pellet burnup of 60 to 70 GWd/t and the data of the irradiation power level of 300W/cm will be accumulated. For example, two program of MOX fuel irradiation is now in progress in Halden reactor. One is the irradiation program of small diameter of MOX fuel to achieve high burnup data in near future. And the another is the irradiation program of Japanese specification MOX fuel to demonstrate the integrity of Japanese MOX fuel.

5.2 FURTHER REDUCTION OF INITIAL He PRESSURE

Further reduction of initial He pressure will be investigated from a view point of the influence on fuel rod elongation and bowing.

5.3 INCREASE OF PLENUM VOLUME

The increase of plenum volume is an another countermeasure to decrease the fuel rod internal pressure. The additionnal length of plenum will be determined taking into account of local power peaking at the end of fuel stack due to thermal neutron current from surrounding UO_2 region.

5.4 ACCUMULATION OF MOX RAMPING DATA

Although PCI resistance of MOX fuel is expected to be better than that of UO_2, considering available ramping test results and its characteristics of high creep rate, it is still necessary to enhance the ramping test data of MOX fuel, especially at high burnup region.

5.5 LARGE GRAIN MOX FUEL PELLET

In the bumup extension of UO_2 fuels, large grain pellet to reduce fission gas release has been developed. The investigation of large grain MOX pellet will be considered in future,

5.6 GADOLINIA MOX FUEL ASSEMBLY

If gadolinia fuel rods are implemented in MOX fuel assemblies, the flexibility of MOX core design will be increased. There are two designs of gadolinia MOX fuel assembly. One uses UO_2 - Gd_2O_3 fuel rods in a MOX fuel assembly, while the other uses MOX - Gd_2O_3 fuel rods. The nuclear characteristics of these design and core design as well as the possibility of the manufacturing of MOX - Gd_2O_3 pellet will be studied.

5.7 REDUCTION OF PU CONTENT VARIETIES IN ASSEMBLY

It is very effective to decrease the variety of Pu content to reduce the fabrication cost of MOX fuel. The feasibility will be considered in future within the limit of the present fuel and core design.

6. CONCLUSION

The adequacy of the methods for MOX fuel mechanical design, nuclear design, and safety evaluation was authorized recently for the practical Pu thermal utilization in PWR's. Up to 1/3 MOX fueled core, modification of the plant is not necessary. Consequently, the fraction of MOX fuel in PWR is designed to be less than 1/3 in the present program. For further improvement of MOX fuel in PWR, Japanese PWR utilities and vendors are now continuing R & D works of MOX fuel irradiation programs

at experimental reactors to study the MOX fuel performance such as the fission gas release behavior and PCI resistance.

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AN ADVANCED BWR MOX FUEL DESIGN CONCEPT FOR JAPANESE LWR PLUTONIUM RECYCLING

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Abstract

This paper presents a concept of the advanced BWR MOX fuel which allows both a simplified fabrication process and higher burnup design, as well as common use of the plutonium source material for PWR and BWR MOX fuels, thereby reducing the MOX fuel cycle costs. The advanced BWR MOX design uses the 9 × 9 fuel assembly with a large square water-channel which enhances the neutron moderation in the fuel assembly. This design merit is fully utilized to minimize the number of Pu enrichment levels to only 2 levels except for the four corner rods and the gadolinium bearing rods which are designed to use uranium, thus allowing a simple process in the MOX fuel fabrication. The lower linear heat generating rate of the 9 × 9 fuel assembly with respect to the 8 × 8 fuel assembly is a key factor to the high burnup design. A core analysis has shown that the advanced BWR MOX fuel has a large operating margin of the linear heat generating rate over the fuel lifetime of peak pellet burnup of 65 GWd/t. Also discussed is a possibility to make common use of Pu enrichment for the PWR and BWR MOX fuels, thereby providing economic benefit in the fabrication process.

1. INTRODUCTION

The Long-Term Program for Research, Development and Utilization of Nuclear Energy by the Japanese Atomic Energy Commission was revised last year (1994) [1]. As discussed in this LongTerm Program, the recycling of nuclear fuel including the use of MOX fuel in the LWRs (Light Water Reactors) is a Japanese policy for the future energy security, economy and resource saving. LWR MOX fuel has been already used extensively in Europe, and Japanese LWR MOX fuel demonstration was started in 1986 at Tsuruga Unit-1 for BWR and in 1988 at Mihama Unit-1 for PWR [2] .

The MOX fuel economy is strongly dependent on the fabrication cost and fuel burnup when it is compared with the uranium fuel [7]. The first objective of this paper is to present a BWR MOX fuel which allows both a simplified fabrication process and higher burnup design. The second objective is to discuss a common use of the Pu source material for PWR and BWR MOX fuels.

2. ADVANCED BWR MOX FUEL DESIGN

2.1 GENERAL DESIGN FEATURE OF NFI 9 × 9 BWR FUEL

The advanced NFI BWR fuel design uses the 9 × 9 FA (Fuel Assembly) with an optimized size LSWC (Large Square Water-Channel) in the BWR fuel assembly [3]. Fig. 1 illustrates the FA structure with its design objectives and key design features.

The lower LHGR (Linear Heat Generating Rate) of the 9 × 9 FA with respect to the 8 × 8 FA is a key to the high burnup design. This is particularly true for the MOX fuel in which the FGR (Fission Gas Release) tends to increase with higher LHGR than the UO_2 fuel at high burnup. While the high burnup design requires a high enrichment fuel, the LSWC enhances the neutron moderation for the high enrichment fuel or the MOX fuel, thus improving the reactivity characteristics related to the void coefficient, hot to cold reactivity swing and shutdown margin. The LSWC also provides for a more uniform moderator distribution in the fuel bundle. This design merit is fully utilized for a simple enrichment design to minimize the number of Pu enrichment levels as discussed later.

Design Objectives

- Batch Average Discharge Burnup : 45 GWd/t
- Maximum Discharge Burnup: 55 GWd/t
- High Reactivity Performance :Large Shutdown Margin for High Enrichment Design
- Large Margin for LHGR, MCPR and Stability for Extended Flow Window Operation
- Optimized Design for Extension to MOX Fuel Application

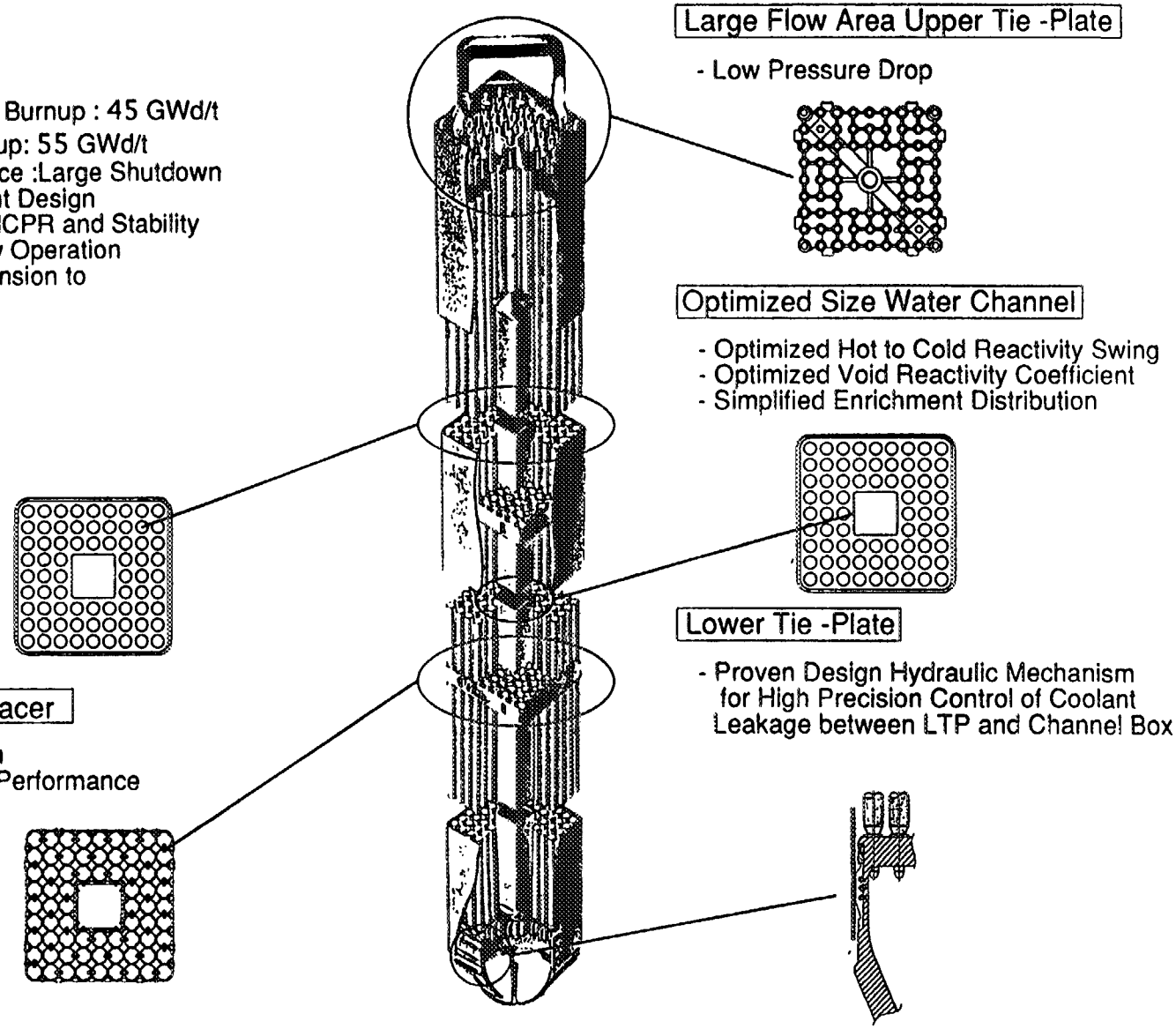
Design Features

9×9 Lattice

- Reduced Average LHGR

Optimized Ring-Type Spacer

- Increased Thermal Margin due to Enhanced Dryout Performance
- Low Pressure Drop



Large Flow Area Upper Tie -Plate

- Low Pressure Drop

Optimized Size Water Channel

- Optimized Hot to Cold Reactivity Swing
- Optimized Void Reactivity Coefficient
- Simplified Enrichment Distribution

Lower Tie -Plate

- Proven Design Hydraulic Mechanism for High Precision Control of Coolant Leakage between LTP and Channel Box

FIG. 1 NFI 9 × 9 High Burnup BWR Fuel

The spacer, upper tie-plate (UTP) and lower tie-plate (LTP) play important roles for the thermal-hydraulic performance. The ring-type spacer provides a high critical power which has been demonstrated by testing [4]. The low pressure drop spacer and UTP compensate for the relatively high frictional pressure drop of the 9 × 9 FA. With respect to the pressure drop, the advanced 9 × 9 FA has been proven by thermal-hydraulic testing including the channel stability test [5]. The LTP design has a unique feature of the hydraulic mechanism for sealing the gap between the LTP casting and the channel box. The flexible LFCD (Leakage Flow Control Device) made of inconel sheet effectively seals the leakage flow which otherwise increases with burnup due to the channel-box bulge, thereby providing high precision control of coolant leakage to the core bypass. The performance of the LFCD has been proven by hydraulic testing and NFI's operating fuel experience has shown its effectiveness.

Further development of the spacer design (ULTRAFLOW) has been made and provides 8% higher critical power and 2% lower pressure drop than that of the ring type spacer, when it is used for the 9 × 9 FA [4]. Either operational margin to the minimum critical power ratio (MCPR) or stability of the core decreases with the more negative void coefficient of the MOX fuel assemblies. The higher critical power provided by the new spacer accommodates the larger margin of MCPR. The lower pressure drop of the spacer increases the channel stability, which in turn increases the core stability.

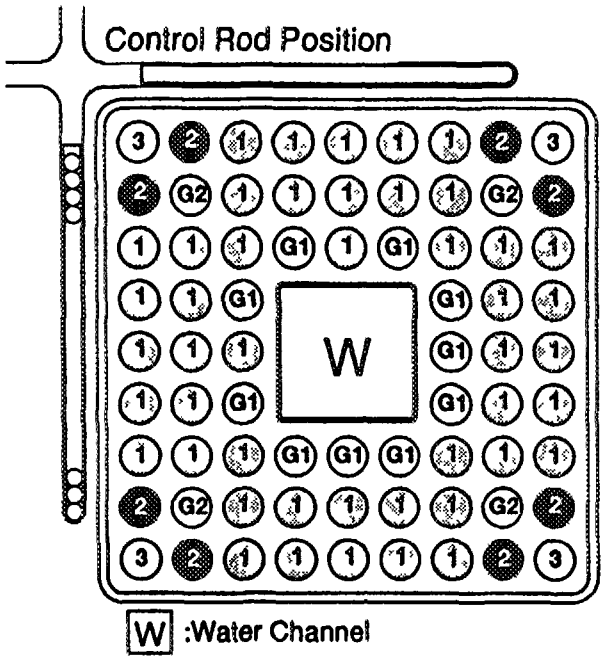
2.2 NUCLEAR DESIGN OF 9 × 9 BWR MOX FUEL

All nuclear designs to be discussed assume a Pu material from the reprocessing of the BWR spent fuel with burnup of 39.5 GWd/t. Total fissile Pu content becomes 63.4 wt% with decay of Pu241 into Am-241 for 8 years after the discharge. The depleted uranium (0.2wt% U-235) is used for the MOX carrying material. A standard fuel active length of the UO_2 FA is about 3.7 m, but for the MOX fuel in this study a short active length of 3.55 m with extended rod plenum was used to assure sufficient margin against potentially higher FGR from the MOX fuel. The burnable absorber (gadolinia) was used in the UO_2 fuel rods, but not in the MOX fuel rods.

The enrichment designs that will be discussed below are shown in Fig. 2 and Fig. 3 for the average discharge burnup of 39.5 GWd/t and 45 GWd/t, respectively. Both designs use only 2 enrichment Pu levels. The four corner rods and the gadolinia bearing rods contain uranium but not Pu. The enrichment of the Pu or uranium as well as the gadolinia content are axially uniform over the active length. The current MOX fuel design practice for both PWR and BWR is to use 3 to 6 Pu enrichment levels in order to decrease the higher local peaking factor [6]. Such design may be best utilized to increase the MOX inventory, but the fabrication becomes complex. The total cost of the MOX utilization in the LWR is dependent on the capital and running costs of the MOX fuel fabrication plant. Simplified BWR MOX design as proposed in this study is a compromise of these factors, but the MOX inventory loss (corresponding to 4 corner rods) is only 6 %. Another important design merit pertaining to the LSWC is relatively lower maximum Pu enrichment. The maximum fissile content (Puf) is only 3.9 wt% or 4.5 wt% for the high burnup design of average discharge burnup of 39.5GWd/t and 45GWd/t, respectively. This is in sharp contrast with the other type BWR FA, e.g. 9 × 9FA with one small water rod [6]. In the MOX FA fabrication plant design, the lower the maximum enrichment, the larger the batch size in Pu handling which may be limited by the criticality control and the shielding design. When the batch size becomes large, availability of the equipment is increased, thus the MOX fabrication cost is reduced.

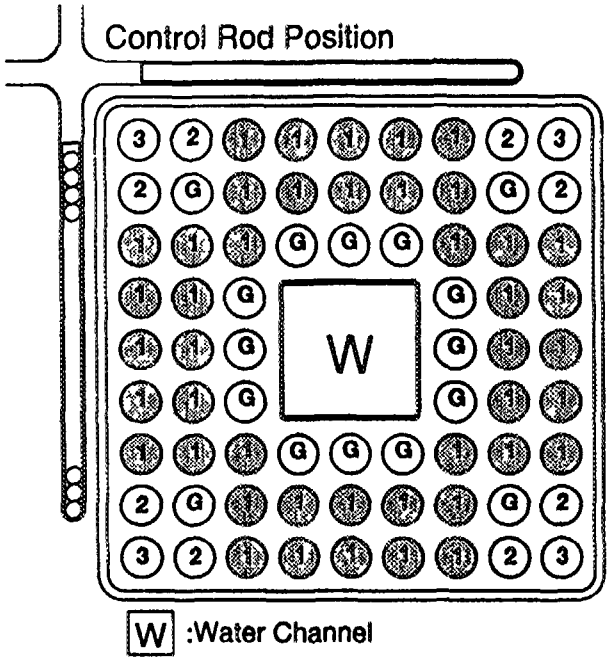
2.3 CORE PERFORMANCE OF THE ADVANCED 9 × 9 BWR MOX FUEL

A reference plant in this study is a 1100 MWe class BWR with 764 fuel assemblies in the core. An equilibrium core of the cycle length of 13 effective-full-power-months (EFPM) was investigated for the core performance of the advanced 9 × 9 BWR MOX fuel. Table I shows the results of the core performance calculations. In this Table, Case U45 represents a reference core of the UO_2 fuel assembly, Case M39 and Case M45 for the uniform core of the MOX fuel assemblies, and Case M39/U45 and Case M45/U45 for the mixed core. The figures "45" and "39" after the letter "U" or "M" mean the average discharge burnup of 45 GWd/t and 39.5 GWd/t, respectively. It is obvious from these calculation results related to MLHGR, MCPR and Shutdown Margin that the steady-state core performance of the MOX FA



Rod Type	wt% Puf	wt% U235	wt%Gd_2O_3	# of Rod
1	3.9	0.2	—	46
2	2.8	0.2	—	8
3	—	3.1	—	4
G1	—	4.9	2.0	10
G2	—	4.9	1.5	4
Average	2.8	1.3	#Total	72

FIG. 2 Lattice Enrichment Design, M39 (Discharge Burnup 39.5GWd/t)



Rod Type	wt% Puf	wt% U235	wt%Gd_2O_3	# of Rod
1	4.5	0.2	—	44
2	3.2	0.2	—	8
3	—	3.3	—	4
G	—	4.9	2.0	16
Average	3.1	1.4	#Total	72

FIG. 3 Lattice Enrichment Design, M45 (Discharge Burnup 45.0GWd/t)

Table I **Calculated Results of Equilibrium Core Performance**

Core Design Parameter	Uniform Core			Mixed Core	
	U45	M39	M45	U45/M39	U45/M45
# of Reload Bundles					
UO_2	164	--	--	120	112
MOX	--	196	172	56	56
Average.Discharge.Burnup (GWd/t)					
UO_2	45	--	--	45	45
MOX	--	40	45	39	45
MLHGR (kW/m)	35	40	39	39	39
MCPR	1.6	1.6	1.5	1.6	1.6
Shutdown Margin (%Δk)	2.5	1.6	1.6	1.9	1.8

Table II **Examples of 17 × 17 PWR MOX Fuel Enrichment Design for Pu Enrichment Common Use with BWR MOX Fuel**

Rod Type	# of Rod	Common Use Pu Enrichment (cf. Fig. 2, Fig. 3)	
		Case 1	**Case 2**
H	176	7.8	7.5
M	76	3.9	4.5
L	12	2.8	3.2

Remarks:

(1) **Case 1 and 2** use the common Pu enrichments with the BWR design case **M39** and **M45**, respectively. (cf. Fig.2, Fig.3)

(2) Both cases are designed for the maximum discharge burnup of 48 GWd/t with average Pu enrichment of 6.4 wt% Puf, and depleted uranium (0.2 wt% U235) for the MOX carrying material.

compares well with the UO_2 FA regardless of the full and partial insertions of the MOX FAs. Fig. 4 shows a comparison of the MLHGR envelope curves of the uniform core cases (U45, M39, M45). Each curve is an envelope of MLHGR vs. fuel pellet burnup. It is noted that the MOX FA in this study exhibits about 5 kW/m higher MLHGR than the UO_2 FA, but there is no remarkable difference between the MOX and UO_2 FAs on the power history as a function of burnup and the MOX fuel has a large operating margin of MLHGR over the fuel life time of peak pellet burnup of 65 GWd/t. Fig. 5 shows the envelope curves for the UO_2 and MOX FAs in a mixed core case (U45/M45). Notwithstanding relatively higher reactivity of the MOX fuel than the UO_2 fuel at high burnup, the mixed core cases were designed so that the MOX fuel gives a lower MLHGR in comparison with the UO_2 fuel at higher burnup (> 50 GWd/t).

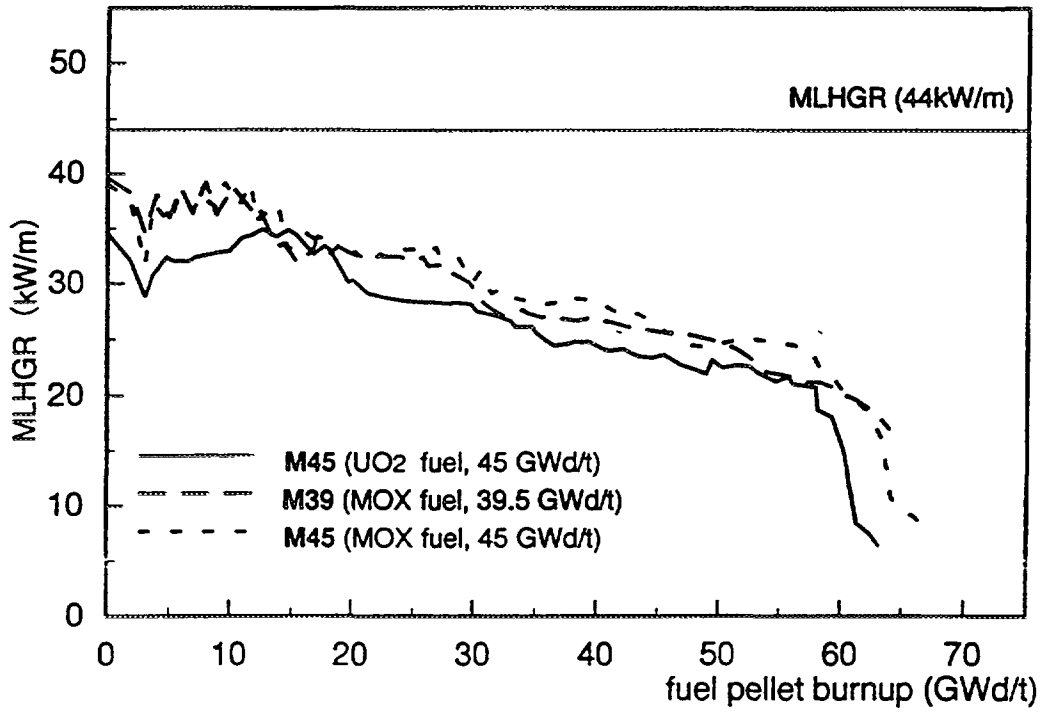


FIG. 4 *MLHGR Envelope Curves of Uniform Core Cases*

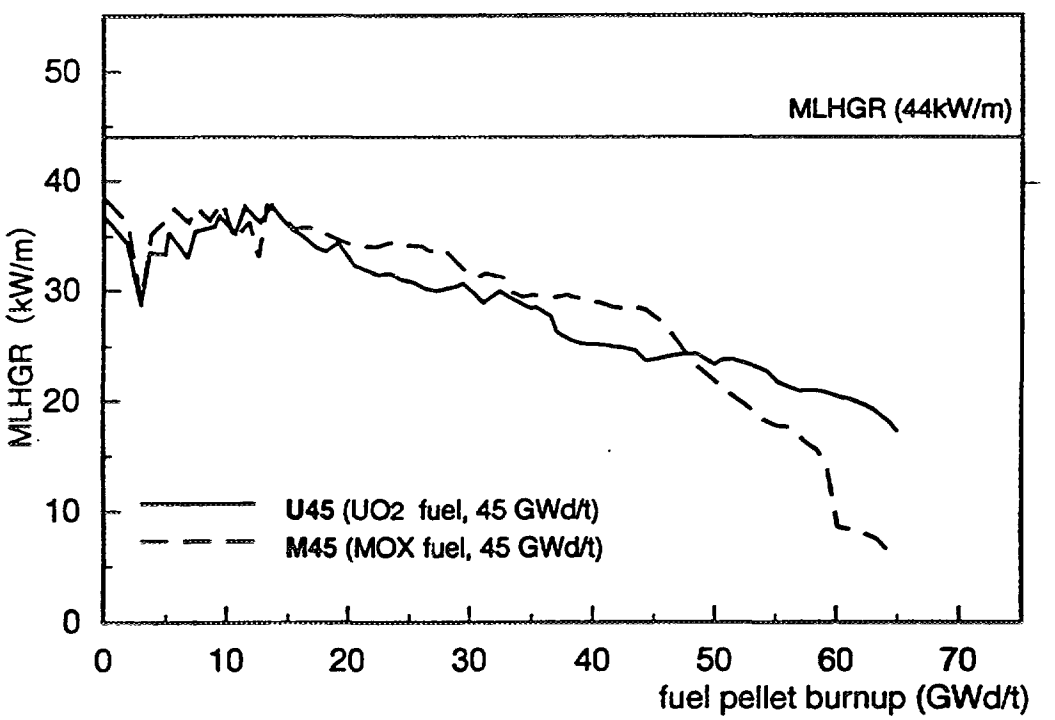


FIG. 5 *MLHGR Envelope Curves of Mixed Core Case (U45/M45)*

It has been demonstrated from the core performance calculation results that the advanced 9 × 9 MOX FA is feasible with the high burnup design with the full MOX core as well as the mixed core with the UO_2 fuel.

2.4 SAFETY CHARACTERISTICS OF ADVANCED 9 × 9 BWR MOX FUEL

Pu-240 in the MOX fuel makes the Doppler coefficient more negative than the UO_2 fuel. This is an augmented safety characteristic of the MOX fuel in the reactor accidents and abnormal transients, in particular the reactivity initiated accident (RIA). On the other hand, approximately 30% more negative void coefficient of the MOX fuel than the UO_2 fuel has a reverse effect in the case of core pressurization transient and core stability. However, the advanced 9 × 9 BWR FA gives about 20% less negative void coefficient than the standard 8 × 8 FA for the same enrichment. This feature partly offsets the more negative void coefficient of the MOX fuel. The mixed core of MOX and UO_2 fuels takes some intermediate value of the void coefficient depending on the fraction of the MOX FAs in the core. The control rod worth tends to become smaller with the increased neutron absorption of the MOX fuel, but the scram characteristic is also a function of the axial power distribution. More negative void coefficient skews the axial power distribution into a bottom peaked shape. The net effect of the MOX fuel on the scram characteristic is not much different from that of the UO_2 fuel.

Although somewhat more quantitative analysis is required when more specific boundary conditions including the plant design are taken into account, it may be concluded from the above discussions that there is no special technological issue for the use of MOX fuel related to the core performance and the safety characteristics.

3. FUEL CYCLE COSTS OF MOX FUEL

3.1 GENERAL TRENDS IN THE MOX FUEL CYCLE COST

It is well known that the fuel cycle costs of MOX fuel strongly depend on the fabrication cost, because only this cost component comprises most of the front-end costs for MOX fuel. Moreover, in the current status of LWR fuel burnup, i.e. the average discharge burnup is less than 60 GWdlt, the fuel cycle costs decrease substantially with higher burnup, in particular for the MOX fuel [7]. This is because all cost components of the MOX fuel decrease directly relative to the generated electricity, i.e. the cost decreases simply with the fuel burnup, whereas the uranium purchase and enrichment costs for UO_2 fuel remain almost constant with burnup.

Fuel cycle costs were calculated for the fuel design cases discussed above in section 2.3. The same economic input data as given in Table 9.1 of reference [7] was used, but no credit for Pu value was taken. Fig. 6 (a) and Fig. 6 (b) show the cost comparison of MOX cases (M39 and M45) against the UO_2 case (U45). As shown in Fig. 6 (a), if the MOX fabrication cost is below 4.0 times that of UO_2, the MOX fuel cycle with the same discharge burnup of 45 GWd/t becomes competitive to the UO_2 fuel cycle. If the MOX discharge burnup is lower than 40 GWd/t, the corresponding MOX fabrication cost is lower than 3.3 times that of UO_2. Based on the proposed BWR MOX fuel design, the importance of the MOX fuel discharge burnup as well as the fabrication cost was confirmed.

3.2 MOX FUEL FABRICATION COST

Taking it for granted that the MOX fuel equivalent to UO_2 fuel in fuel cycle costs is a measure of the MOX fuel economy, the key factor is the MOX fabrication cost. The fabrication cost depends on the throughput and the construction cost of the fabrication plant. If a simple enrichment design as presented in this study is used for the BWR MOX fuel, it seems obvious that both the throughput and number of the process lines will be optimized, particularly for the new MOX fabrication plant in Japan.

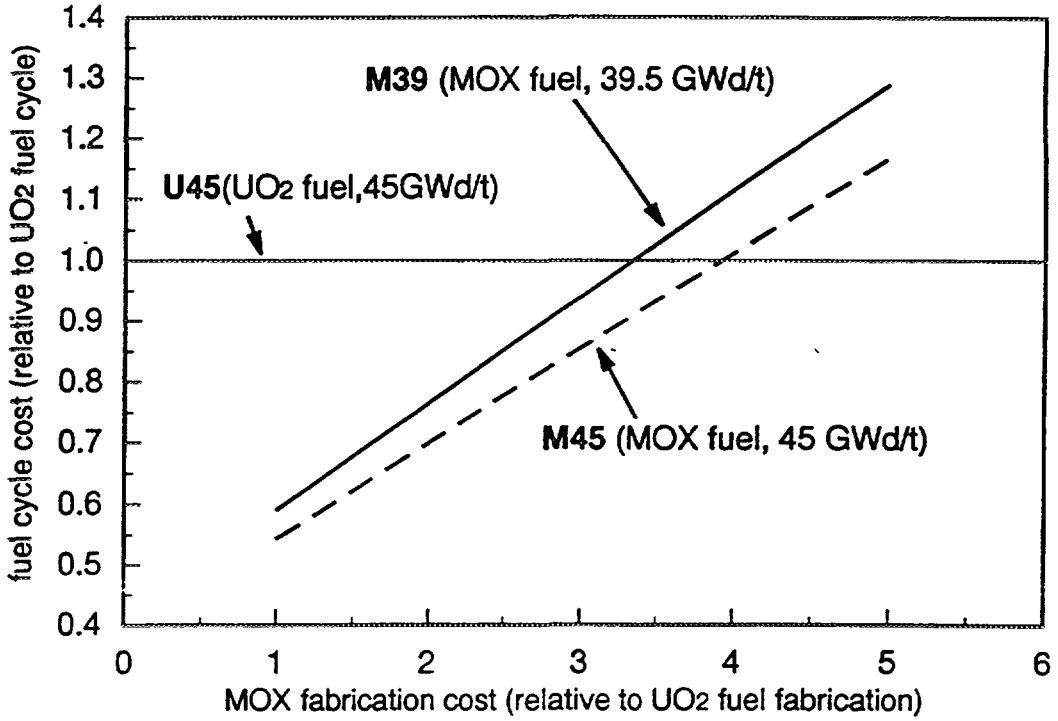


FIG. 6a Fuel Cycle Costs of MOX Fuel

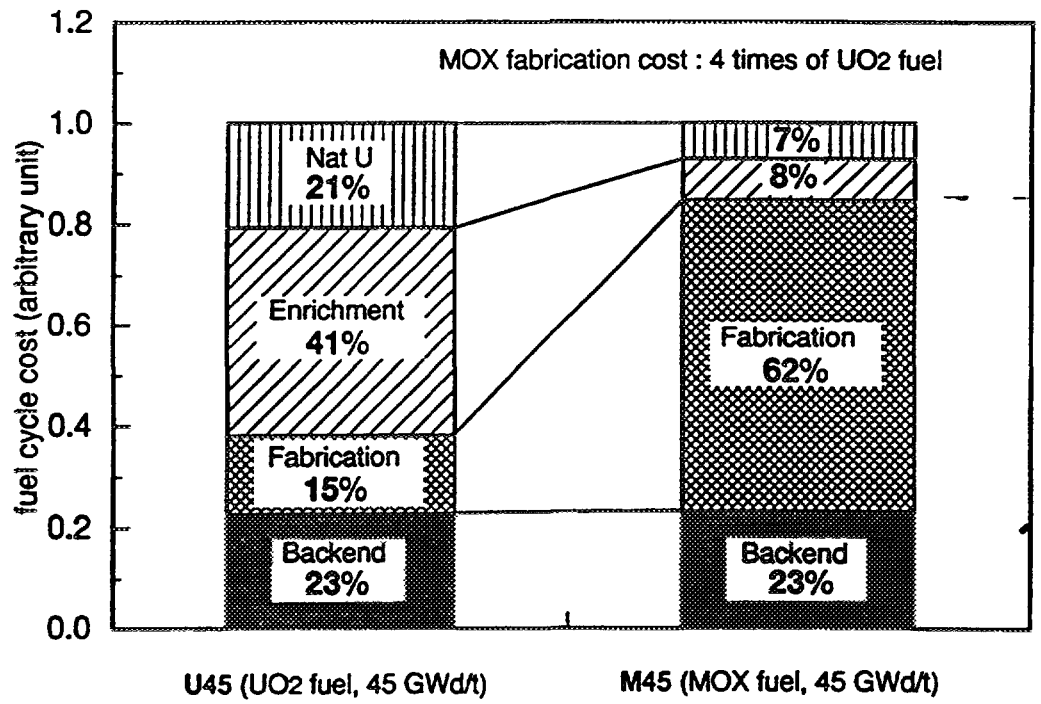


FIG. 6b Fuel Cycle Cost Components

4. COMMON USE OF PU ENRICHMENT FOR PWR AND BWR MOX FUELS

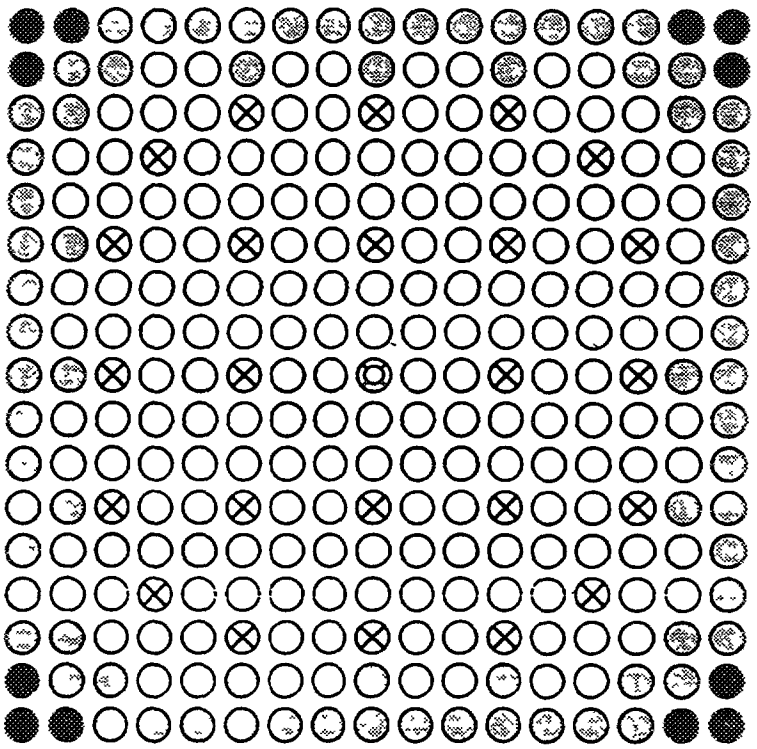
4.1 COMMON USE OF PU MATERIAL

It is of further benefit for the MOX fuel plant design that a common use of the Pu enrichments is made for the PWR and BWR MOX fuels. Since the PWR and BWR fuels use different dimensions and

other specifications of the fuel pellet, only Pu enrichment can be commonly used. However, the common use saves the time required for the cleaning of the process line from the powder preparation to the pelletizing. Thus the process utilization is increased. Another merit of such common use is to make the process batch size more uniform so that the powder preparation process is utilized efficiently, since a small batch of the common enrichment for the PWR and BWR MOX rods can be combined to form a larger batch size. Such examples are presented below.

4.2 EXAMPLE OF COMMON USE OF PU ENRICHMENT

Table II and Fig. 7 show the PWR MOX fuel enrichment designs for maximum burnup of 48 GWd/t. In these designs, two enrichments of the BWR MOX fuel in Fig. 2 or Fig. 3 are commonly used, i.e. 3.9 wt% and 2.8wt% Puf (M39) or 4.5 wt% and 3.2wt% Puf (M45) are used in the PWR MOX bundle. Preliminary analysis has shown that both enrichment designs are compatible with the UO_2 fuel of the same discharge burnup.



⊗ :Instrumentation guide tube

⊗ :RCC guide tube

Rod Type	wt% Puf	wt% U235	# of Rod
○	7.8	0.2	176
○	3.9	0.2	76
●	2.8	0.2	12
Average	6.4	0.2	# Total 264

(3.9wt%Puf and 2.8wt% Puf are commonly used with the BWR MOX fuel)

FIG. 7 PWR MOX Fuel Enrichment Design

5. CONCLUSION

The advanced 9 × 9 BWR MOX fuel design concept that uses 2 Pu enrichment levels and allows a simple process in the MOX fuel fabrication was proposed. A core analysis has shown that the advanced BWR MOX fuel has a large operating margin of LHGR over the fuel lifetime. Based on the proposed BWR MOX fuel design, the effects of the MOX fabrication costs and the high burnup on the MOX fuel cycle costs were calculated. Also discussed was a possibility to make common use of Pu enrichment for the BWR and PWR MOX fuels. As a summary, it can be concluded that the simplified enrichment design of the BWR MOX fuel as well as the common use of Pu enrichment for the PWR and BWR MOX fuels would contribute to the MOX fuel economy.

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TECHNOLOGY DEVELOPMENTS FOR JAPANESE BWR MOX FUEL UTILIZATION

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Abstract

The Long-Term Program for Research, Development and Utilization of Nuclear Energy established by the Atomic Energy Commission of Japan asserts that Japan will promote systematic utilization of MOX fuel in LWRs. Based on this Japanese nuclear energy policy, we have been pushing development of MOX fuel technology aimed at future full scale utilization of this fuel in BWRs. In this paper, the main R & D topics are described from three subject areas, MOX core and fuel design, MOX fuel irradiation behavior, and MOX fuel fabrication technology. For the first area, we explain the compatibility of MOX fuel with UO_2 core, the feasibility of the full MOX core, and the adaptability of MOX design methods based on a mock-up criticality experiment. In the second, we outline the Tsuruga MOX irradiation program and the DOMO program, and suggest that MOX fuel behavior is comparable to ordinary BWR UO_2 fuel behavior. In the third, we examine the development of a fully automated MOX bundle assembling apparatus and its features.

1. INTRODUCTION

The Long-Term Program for Research, Development and Utilization of Nuclear Energy revised by the Atomic Energy Commission of Japan June, 1994 asserts that Japan will promote systematic utilization of MOX fuel in light water reactors (LWRs). This utilization is important from the viewpoint of establishment of the technology and systems needed for nuclear fuel recycling on a practical scale as a prerequisite for future practical use of fast breeder reactors (FBRs). Furthermore in advancing the utilization of MOX fuel in LWRs, the program suggests that for the time up to commercial commissioning of FBRs, it is necessary to undertake use of MOX fuel in LWRs on an appropriate scale which considers the scale of nuclear fuel recycling. Specifically, the program indicates it is appropriate to start using MOX fuel in a few LWRs in the second half of the nineties and to increase the number of such reactors in a planned manner, but with some flexibility, to about ten by the year 2000 and over ten by 2010.

In line with this Japanese nuclear energy policy, we have been pushing development of MOX fuel technology aimed at future full scale utilization of MOX fuel in boiling water reactors (BWRs). The technology developments consist of a wide range of technical areas, such as MOX core and fuel design, MOX fuel irradiation behavior, MOX fuel fabrication technology, design and performance codes for MOX fuel, MOX fuel bundle handling and shipping, and MOX bundle inspection technique, etc. Some of these research and development (R & D) programs have been performed with support from electrical utilities or the Japanese government.

In this paper we select three main areas, the MOX core and fuel design, the MOX fuel irradiation behavior, and the MOX fuel fabrication technology, and describe the main topics of the R & D programs in these areas.

2. DESIGN STUDY OF BWR-MOX CORE AND FUEL

2.1 BASIC FEATURES OF MOX FUEL IN BWR

For the design of MOX fuel and MOX loaded core, good understanding of the basic characteristics of MOX and adequate design considerations are required because of the differences between UO_2 and MOX. The MOX behavior in the LWRs is quite similar to that for UO_2 and just a few design considerations are enough to ensure proper utilization of MOX fuels in BWRs without significant modification in reactor plant designs. Compared to UO_2, the major features of MOX are summarized as follows.

(1) Nuclear characteristics

a. More negative reactivity coefficient in void, Doppler and moderator temperature.
b. Smaller reactivity control worth in the control rod and for burnable poison.
c. Possible higher power peaking in the MOX fuel rod which is adjacent to the soft neutron spectrum area.
d. Shorter prompt neutron lifetime and smaller fraction of delayed neutron.
e. Less reactivity reduction during burnup.
f. Slight differences in He generation and fission yield.

(2) Material properties and fuel irradiation behavior

a. Lower pellet melting point and pellet thermal conductivity as a function of Pu content.
b. Higher pellet creep rate as a function of Pu content.
c. Slightly higher fission product release rate.

2.2 DESIGN CONSIDERATIONS IN MOX FUEL AND CORE

There are several ways to eliminate any effects from the difference between UO_2 and MOX fuels in practical MOX design. For the present use of MOX fuels in Japanese BWRs, MOX fuels are planned as part of reload fuels. Therefore, fuel assembly configurations for MOX fuels are designed to maintain compatibility and exchangeability with those of UO_2 assemblies. That is, fuel rod arrangement, rod dimensions, materials and thicknesses for fuel cladding, and fuel spacers and tie-plates designs are identical to those of UO_2 fuel assemblies. Maximum bundle exposure is designed to stay less than that of UO_2 fuels.

From a nuclear characteristics viewpoint, it is important to sustain the proper void reactivity coefficient to avoid complexity in reactor operation and control. For BWRs, void reactivity coefficient is a major concern. A more negative void reactivity coefficient may result in a smaller thermal margin during a pressure increasing transient. Hence, it is desirable for the MOX fuel to have the same magnitude of reactivity coefficient and the same thermal hydraulic behavior as the UO_2 core, when MOX fuel assemblies make up a substantial part of reload assemblies. In this regard, MOX fuel assemblies for BWRs are identical to the UO_2 fuel assemblies for high burnup, which have a large water rod in place of four fuel rods as shown in Fig. 1. A fuel assembly with a large volume of water rod helps neutron moderation and improves core characteristics of void reactivity and shutdown margin. Until now, most MOX fuel assemblies used for BWRs have been the island type, in which only the central rods in the assembly are MOX fuel rods and others are UO_2 rods. For the coming commercial use of MOX fuel in Japan, we have selected the all-MOX type assemblies rather than the traditional island type assemblies. The all-MOX type assembly can use several types of MOX rods with different Pu enrichments and can contain a large quantity of Pu. Such a design is preferable one using Pu concentrated within a small number of fuel assemblies.

For simplification of MOX fuel fabrication, it is also important to reduce the number of MOX rod types with different Pu enrichments, resulting in a smaller number of MOX assembly production steps. We optimized the number of MOX fuel rod types and quantity of Pu per assembly and developed the

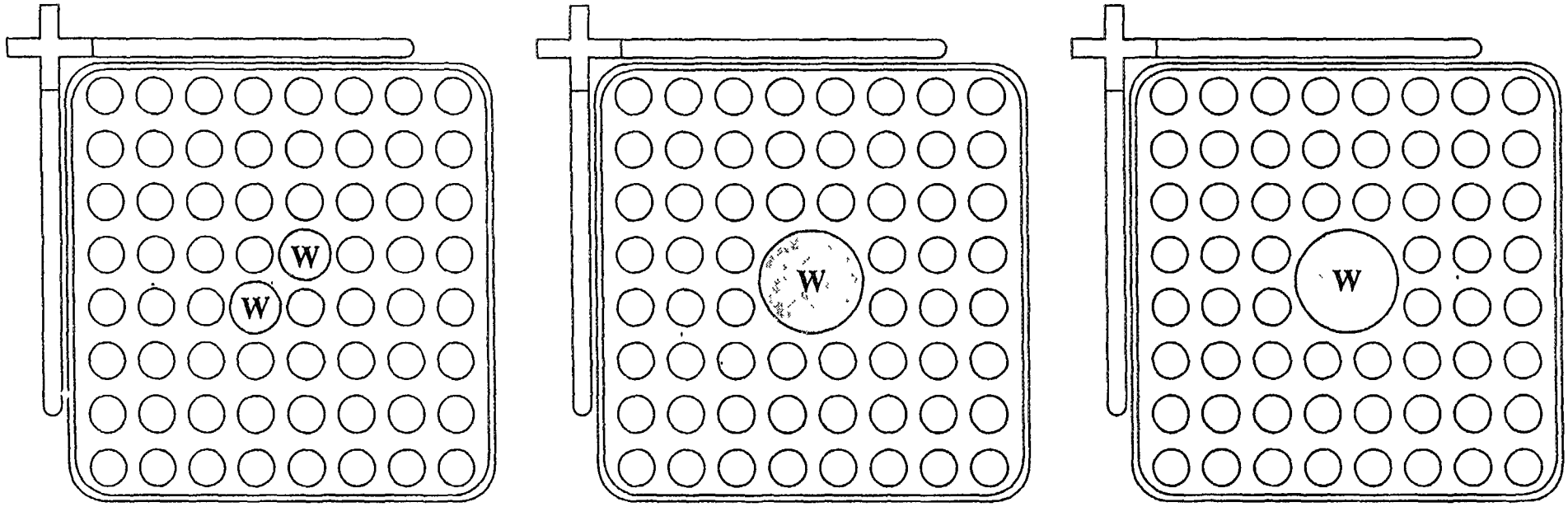


Fig.1 Comparison of fuel assembly configuration for uranium fuel and MOX fuel

all-MOX type assembly with four types of MOX rods of different Pu enrichments and small number of uranium rods at the location of high power peaking. Based on the BWR-MOX design and irradiation experience at Tsuruga Unit - 1 [1], a detailed Pu and Gd distribution design for the commercial BWR-MOX assembly is now in progress. The design is expected to be suitable for the MOX utilization program in BWRs with the UO_2 assemblies for high burnup.

2.3 FEASIBILITY STUDY OF FULL-MOX BWR CORE

It is desirable for BWR to have some flexibility in the quantity of Pu utilization by adding Pu inventory per reactor. It is helpful to adjust Pu surplus from the imbalance of demand and supply. For this reason, Hitachi, Ltd. (Hitachi) studied an Advanced BWR (ABWR) core loaded fully with MOX fuel assemblies. Table I describes the specifications of this MOX core. Preliminary core characteristics evaluation showed satisfactory results in shutdown margin, thermal margin and reactivity coefficient against core design criteria as shown in Figs. 2 and 3. This study also showed that greater moderation of neutrons such as by addition of water rods is a key factor for further improvement of burnup and core characteristics.

2.4 MOX CORE DESIGN METHOD AND ITS VERIFICATION

The BWR core design method for the UO_2 core is based on the three dimensional neutron diffusion analysis of the whole core with a few energy-group nuclear constants from fuel lattice cell neutron transport. This method includes heterogeneity in the BWR core which comes from the void distribution in core, the enrichment distribution and the local gadolinia location in the lattice cell. We planned to use this method for design of the MOX fuel core.

One verification of this method was confirmed by the power profile trace of Tsuruga MOX fuel. The difference in the power profile between TIP (Traversing In-core Probe) measurements and core

Table 1 Specifications of full-MOX BWR core

Reactor core	
Electric power	1356 MWe
Thermal power	3926 MWt
No. of fuel bundles	872
Power density	50.6kW/l
Fuel bundle	
Configuration	8 x 8
Discharge exposure	~33 GWd/t
Puf enrichment (*1)	~2.9 wt%
U enrichment	~1.0 wt%
Core characteristics	
Annual Puf feed	~1.1 ton-year
Shutdown margin (*2)	$\geq$ 2.4 % Δk
MLHGR (*3)	$\leq$ 12.4 kW/ft
Core stability decay ratio	$\leq$ 0.7

Notes: *1 Puf = fissile Pu
*2 design limit is 1% Δk.
*3 Maximum Linear Heat Generation Rate, operation limit is 13.4 kW/ft.

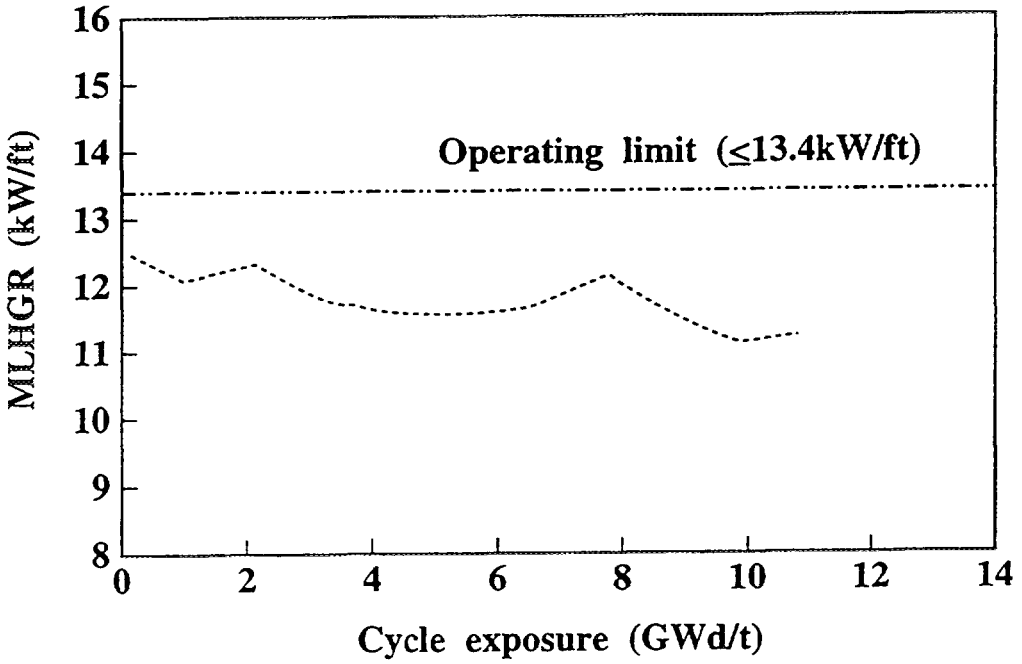


Fig.2 Maximum Linear Heat Generation Rate (MLHGR) for reload full-MOX core design

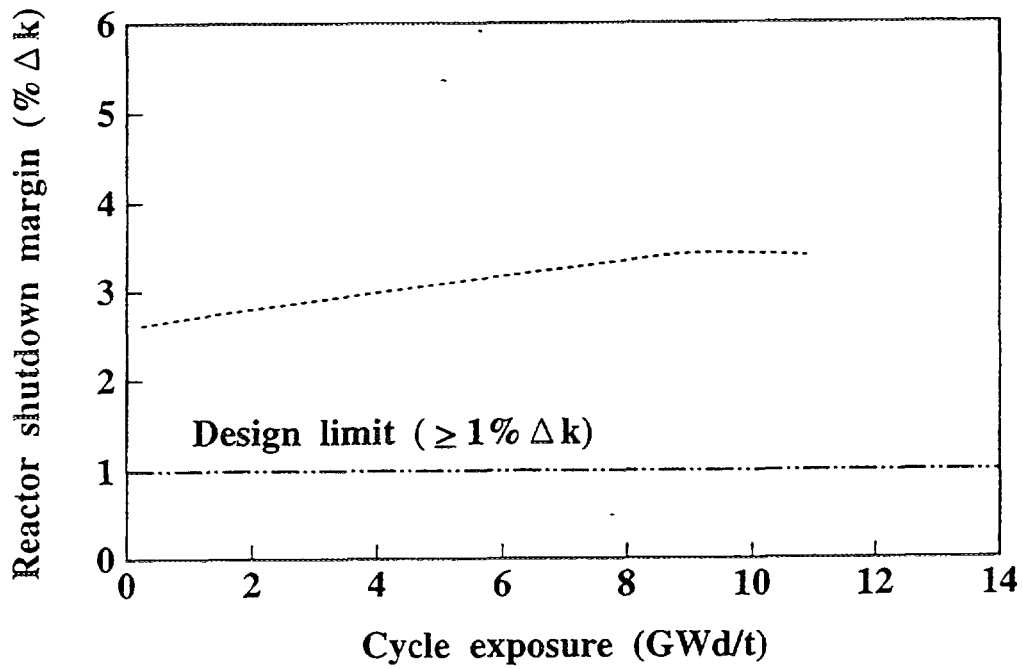


Fig.3 Reactor shutdown margin for reload full-MOX core design

analysis for the MOX fuel assembly was not larger than corresponding results for a UO_2 fuel assembly [1]. Another verification was based on the MOX core mock-up criticality experiment, in which experimental data were compared with the results from a BWR analysis method. Figure 4 compares calculation and measurement for the MOX mock-up experiment. Both eigenvalue and root mean square error in rod power for the MOX mock-up have the same magnitude as for a UO_2 mock-up [2].

An advanced lattice cell analysis method, VMONT, based on Monte Carlo neutron transport and with an isotope depletion capability, has been developed by Hitachi for future sophisticated design work of the core and fuel [3].

3. MOX FUEL IRRADIATION STUDIES

We have been carrying out various irradiation studies on BWR-MOX fuel in order to obtain information on its behavior. In this paper, two irradiation programs are outlined. One is the Tsuruga MOX irradiation program, which was the first MOX fuel irradiation in commercial LWR in Japan, and the other is the DOMO program, which was featured as a high burnup irradiation experiment for BWR-MOX fuels.

3.1 TSURUGA MOX FUEL IRRADIATION PROGRAM

3.1.1 Outline of the program

The program was carried out to demonstrate plutonium usage in BWRs, in which two BWR MOX island type fuel assemblies were irradiated in Tsuruga Unit-1 (BWR) of Japan Atomic Power Company (JAPCO) [1]. Six BWR-operating utilities, Power Reactor and Nuclear Fuel Development Corporation (PNC), and reactor and fuel manufacturers participated in the program. We joined in the fuel design and fabrication, and in the data evaluation. The fuel design work started in 1980, the licensing in 1984, and the fuel fabrication in 1985. Irradiation was begun in July, 1986. This was the first time MOX fuel was irradiated in an LWR in Japan [4, 5]. The fuel rod inspection work included visual observations and fuel rod length measurements which were carried out on the MOX assemblies during the scheduled plant outage after each reactor operating cycle. After completion of three irradiation cycles, two MOX assemblies were shipped to the hot laboratory of Nippon Nuclear Fuel Company (NFD). Pre-scheduled MOX fuel rods and UO_2 fuel rods were retrieved from the assemblies and post irradiation experiments (PIEs) were carried out at PNC on the former and at NFD on the latter.

Average and local peak burnups of the MOX discharged assemblies were about 26 GWd/t and about 37 GWd/t, respectively. The maximum linear heat rate was about 330 W/cm.

3.1.2 Fuel design and fuel rod specifications

The main specifications of the MOX assemblies, which were the identical with the BWR 8 x 8 type UO_2 fuel assembly (STEP-2 assembly) are shown in Table II [1]. The fuel rod arrangement in the MOX assembly was the island type in which 24 MOX fuel rods were placed in the center region and 38 low enrichment UO_2 fuel rods were in the periphery of the assembly. Figure 5 shows the arrangement of the fuel rods [6]. There were three types of MOX fuel rods which differed in plutonium content, low, medium, high, ranging from approximately 2 to 5 %, and four kinds of UO_2 fuel rods having different ^{235}U enrichment in the range of about 2 to 3 %. Fuel pellets for the UO_2 fuel rods had the ordinary solid shape, while the MOX fuel rods adopted hollow pellets in order to secure the needed nuclear-thermal margin by increasing the water to uranium ratio, and to reduce the fuel center temperature [1].

3.1.3 MOX fuel behavior when irradiated at Tsuruga Unit-1

The fuel rod inspections at the reactor site revealed that no abnormalities occurred in the MOX rods during irradiation [1]. As shown in Fig. 6, MOX fuel rod elongation was almost the same as for UO_2

Table 2 **Specifications of Tsuruga MOX fuel assembly**

Assembly	
Fuel rod array	8 × 8
Average Pu content	~ 3.2 wt%
Average ^{235}U enrichment	~ 2.4 wt%
Number of MOX fuel rods	24
Number of Pu contents	3
Number of UO_2 fuel rods	38
Number of $Gd_2 O_3$ fuel rods	7
Weight of heavy metal	~163 Kg
Weight of Puf	~2 Kg
Fuel Rod	
Active fuel length	366 cm
Filling gas	helium
MOX fuel pellet	
Material	$PuO_2 + UO_2$
Initial enrichment	~ 2.9 wt% (*)
External diameter	~ 1.0 cm
Internal diameter	~ 0.4 cm
Theoretical density	~ 95 %TD

(*); $(^{235}U + ^{239}Pu + ^{241}Pu)/(U + Pu)$

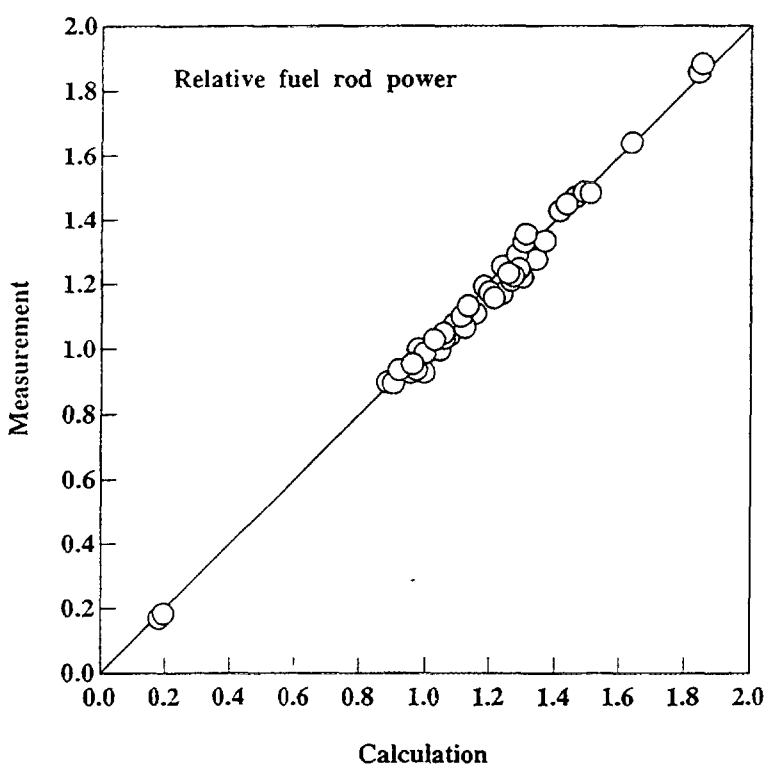
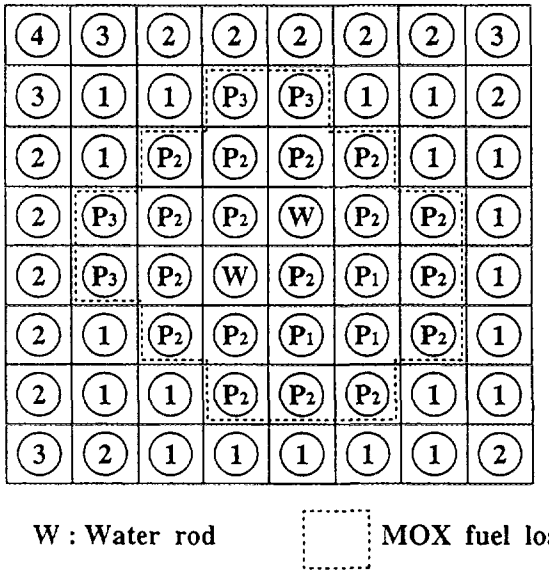


Fig.4 Comparison of calculation and measurement of relative fuel rod power (fission reaction rate) for MOX mock-up criticality experiment



UO_2 fuel rod		MOX fuel rod	
1	high enrichment rod	P_1	high Pu content rod
2	medium-high enrichment rod	P_2	medium Pu content rod
3	medium-low enrichment rod	P_3	low Pu content rod
4	low enrichment rod		

Fig.5 Fuel rod arrangement in Tsuruga MOX fuel assembly

fuel rods [1]. The non-destructive examinations on MOX fuel rods and UO_2 fuel rods were completed and the following marked results were obtained.

(a) Visual inspections, eddy current testing, γ scanning and dimensional measurements confirmed that the MOX fuel rods and the components of the MOX fuel assembly, such as the upper and lower tie-plates, bundle spacers, extension springs, finger springs etc. remained intact during the irradiation [7].

(b) Outer surface oxide thicknesses of the MOX fuel rods were comparable to those of UO_2 fuel rods with Zircaloy-2 (Zry-2) fuel claddings irradiated in various BWRs, [7].

(c) FP gas release rates of the MOX fuel rods were lower than those of UO_2 fuel rods as shown in Fig.7. This was considered due to lower temperature in the hollow MOX fuel pellet [8].

Detailed examinations on MOX fuel pellets and claddings, such as radial distribution of FP elements, melting temperature, thermal diffusivity, oxygen-metal ratio (O/M ratio), and Pu spots were made at PNC and a comprehensive evaluation of them is under way by comparing these data with those of UO_2 fuel pellets and cladding obtained at NFD. A few results from measurements of MOX pellets has been published [9,10]. The results indicated a significant burnup increase in the periphery region of the MOX pellets, and a decrease in pellet melting temperature due to addition of plutonium. However these phenomena are considered too small to affect fuel performance. It can therefore be safely concluded from the data available so far that there is no significant difference in irradiation behavior between MOX fuel and UO_2 fuel.

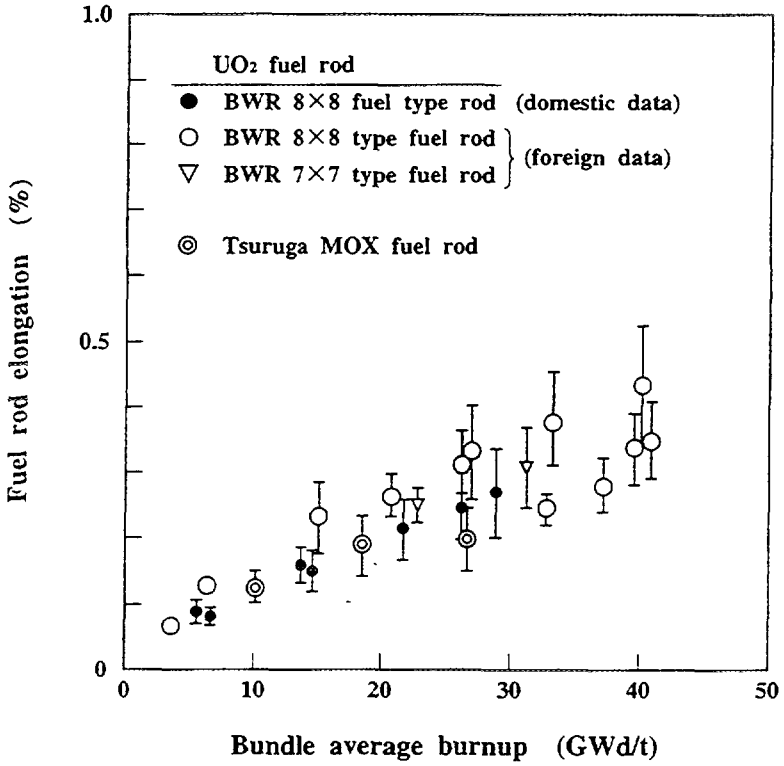


Fig.6 Fuel rod elongation of Tsuruga MOX fuel rods

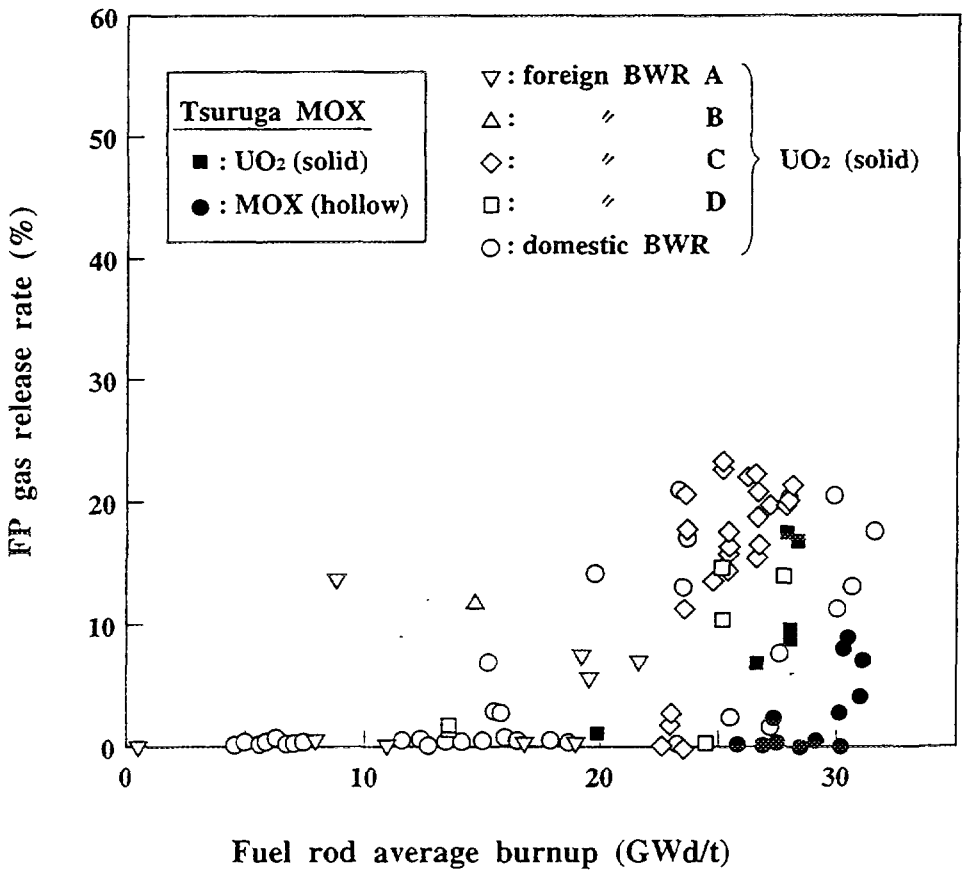


Fig.7 Fission gas release as a function of fuel rod average burnup

3.2 HIGH BURNUP MOX FUEL IRRADIATION BEHAVIOR-DOMO PROGRAM

3.2.1 Outline of the program

The high burnup program for UO_2 fuel has been progressing towards one of its goals of improved economy of BWRs [6]. In order to obtain high burnup data of BWR-MOX fuel the international research program, DOMO, was started in 1986 [11,12]. It has been carried out under the sponsorship of utilities, fuel manufacturers, reactor manufacturers and research organizations in Japan (six BWR-operating utilities, Hitachi, Toshiba, PNC, Japan Nuclear Fuel Co., Ltd (JNF) and NFD), the Netherlands (GKN and KEMA) and Switzerland (PSI) [12]. The director of the program has been Belgonucléaire (BN) of Belgium. In the program, five assemblies containing BWR-MOX fuel rods are irradiated in the Dodewaard BWR in the Netherlands. The MOX fuel rods are individually retrieved after achieving target burnups of 20, 40 and 60 GWd/t and subjected to PIEs. A couple of rods are selected out of the retrieved rods at the respective burnups and ramp tested in the BR2 up to the maximum power of 600 W/cm [12].

3.2.2 Test parameters and fuel rod specifications

The test parameters in the program were mainly related to the MOX fuel pellet fabrication process and cladding material. Some of the MOX fuel pellets were provided by BN and used BN's MIMAS (Micronized Master Blending) powder, and others were provided by PNC and used PNC's MH (Microwave Heating Process) powder. The cladding materials were Zry-2 cladding tube and Zry-2 cladding tube with zirconium-liner (Zr-liner cladding). UO_2 fuel pellets which served as a comparison were fabricated by NFD. Short length fuel rods (segments X,B,J) were fabricated by combining the respective MOX pellets and claddings as described in Table III. Short length rods (segment U) had UO_2 pellets with Zr-liner cladding, and were manufactured at JNF. Figure 8 shows a schematic view of the segment. Four segments of the same combination were mounted into a full length rod. Sets of two MOX full length rods and one UO_2 full length rod were inserted into each of five assemblies.

Table 3 Main specifications of fuel segments and rods

Segment Type			X	B	J	U
Fuel pellet						
$^{235}U/U$	(wt%)		0.239	0.239	0.70	4.95
$^{235}U + Puf/U + Pu + {}^{241}Am$	(wt%)		4.8	4.8	4.81	4.95
Theoretical density		(%TD)	95	95	95	95
Diameter		(mm)	10.35	10.35	10.35	10.35
Height	(mm)		11.5	11.5	10.3	10.3
Manufacturer			BN	BN	PNC	JNF
Zry Cladding						
Zr-liner			yes	no	yes	yes
Inner diameter	(mm)		10.55	10.55	10.55	10.55
Outer diameter	(mm)		12.27	12.27	12.27	12.27
Segment						
Fuel stack length		(mm)	281	281	281	281
Overall length	(mm)		447	447	447	447
He absolute pressure	(atm)		5	5	5	5
Fuel rod						
Overall length	(mm)		2011	2011	2011	2011

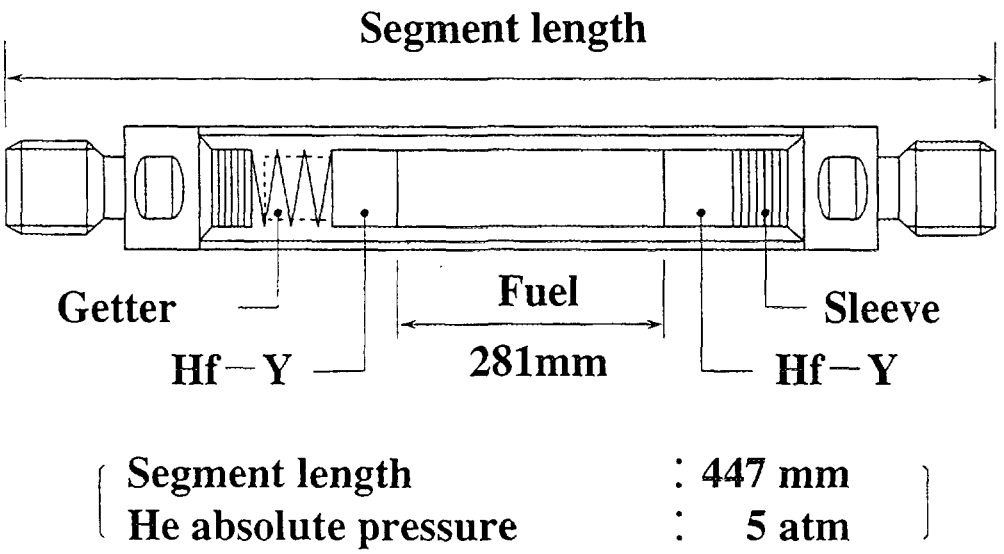


Fig.8 Schematic of segment

3.2.3 Irradiation in Dodewaard and power ramp testing at BR2

Irradiation in the Dodewaard BWR began in February 1988 and continued until January 1993. One MOX assembly was discharged in January 1990 after a two-cycle irradiation. Its segment peak burnup was about 28 GWd/t. Pre-selected segments were removed from the assembly and subjected to PIE. The two more assemblies were discharged in January 1992, after a four-cycle irradiation. Their segment peak burnup was about 50 Gwd/t. Non-destructive examinations have been completed and destructive examinations are almost completed. The remaining two MOX assemblies were retrieved from the reactor after a five-cycle irradiation. Their pellet peak burnup was estimated as about 60 GWd/t. Site inspections were done on the assemblies.

Stepwise power ramp tests were performed in the BR2 by using a pressurized water capsule [13]. The segments were preconditioned for about one day at a power of 300 W/cm. The power was then increased by 50 W/cm during 1-2 minutes, followed by a one-hour hold. The maximum power levels were about 600 W/cm, followed by about a 24-hour hold.

3.2.4 Results

a) **Steady state irradiation behavior.** For all segments irradiated for two, four and five reactor cycles in the Dodewaard, deposits of crud were apparent and nodular corrosion was observed on the outer surfaces of the fuel regions and on the plenum regions, respectively. However no other abnormal features were found during site inspections and gamma scans [12]. Eddy current tests confirmed that all segments irradiated for two and four reactor cycles were intact. No systematic difference in length change was observed between the MOX and UO_2 segments, and between Zry-2 and Zr-liner claddings. There was no apparent correlation between the increase of segment length and the maximum linear power experienced during the irradiations. Therefore it seems that the segment length increase was mainly due to cladding irradiation growth. There was no significant difference in averaged thickness of the cladding outer surface oxide between the MOX segments and UO_2 segments.

The thickness of the inner surface oxide was apt to increase with segment burnup as shown in Fig.9 [12]. However, even when the burnup reached about 55 GWd/t, the inner oxide thickness was thinner than 20 μ m. No distinct difference was observed between Zry-2 claddings with and

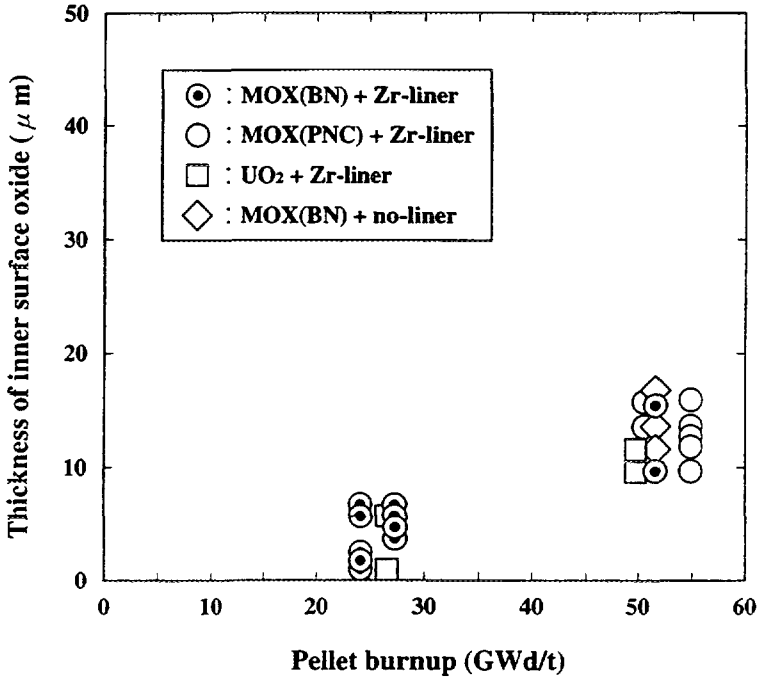


Fig.9 Thickness of inner surface oxide as a function of pellet burnup

without Zr-liner. A notable point was the fact that the maximum inner oxide thicknesses of MOX segments were comparable to those of UO_2 segments [12,13]. The chain yield for ^{235}U and ^{239}Pu shows that in plutonium fission the low-mass number peak is shifted to higher mass numbers compared to uranium fission. This means more metallic elements are produced in the plutonium fission. Therefore, on a simple thermodynamical consideration, the oxygen potential in MOX fuel seems to increase more rapidly at an early stage of burnup compared to UO_2 fuel. Accordingly, the inner surface oxidation of the MOX segments should have been more prominent than those of the UO_2 segments. The oxygen potential of fuel is affected by the chemical state of fission products, i.e. depending on the affinity of the fission products for oxygen. The SIMS (Secondary Ion Mass Spectrometry) analysis on the MOX fuel pellets, and ceramographic observation as well, revealed no evidence for significant oxidation of the metallic elements, such as Mo to MoO_3, but there was clear evidence of metallic precipitates being present in the middle-to-central region of the pellets [12]. These data suggested that free oxygen released by fission may have been consumed mainly in the cladding inner surface oxidation. Since the present thickness data obtained from each cross section were limited to very local information on the oxidation, further information, such as about the extent of the oxidation area along the cladding, is necessary for better understanding of the inner surface oxidation of the MOX fuel.

Fractional fission gas release was calculated using released fission gas quantity obtained by measuring ^{85}Kr non-destructively and by puncturing fuel segments. The fractional fission gas release rose at an average burnup of around 20 GWd/t [12]. At burnup up to about 25 GWd/t the fractional fission gas release of the MOX segments was comparable to that of UO_2 segments, but as burnup drew near 50 GWd/t the fractional fission gas release of the MOX segments seemed to be comparable to, or slightly higher than, the UO_2 segments. Figure 10 shows pellet microstructure of the BN-MOX pellets and the PNC-MOX pellets irradiated for four reactor cycles (pellet burnup: about 50 - 55 GWd/t) [12]. The microstructure in the peripheral regions where temperatures were low enough to prevent grain growth during irradiation retained their as-fabricated microstructure. In spite of a significant difference in the microstructure between the BN-MOX and PNC-MOX fuel pellets, there was no distinct difference in the fractional fission gas release between them [13]. In a comparison at the same maximum power, the fractional fission

gas release of the MOX segments was almost the same as the UO_2 segments with two reactor cycle irradiations, at four reactor cycles, the fractional fission gas release of the MOX segments seemed to be a little higher than for the UO_2 segments [12]. The above results should be confirmed by data from segments irradiated for five cycles (peak burnup: about 60 GWd/t).

b) **Transient behavior.** Five segments, four MOX segments and one UO_2 segment selected from the segments irradiated for two and four cycles, were ramp tested in the BR2. All the ramp tested segments were confirmed by a failure detection system to remain intact during the ramp testing and by non-destructive inspections after ramp testing [12]. For the segments of the two cycle irradiation, no changes in length and diameter were observed. However, additional fission gas release took place. The ceramographic examination revealed that a very large grain growth occurred in the middle-to-central regions of the pellets. For the segments of the four cycle irradiation, only non-destructive examination has been completed on each MOX segment and UO_2 segment. Neutron radiographs suggested the formation of central voids along the fuel columns for the MOX and UO_2 segments.

3.3 CONCLUSION

Results of the BWR-MOX fuel demonstration program in Tsuruga Unit - I and the high burnup MOX fuel irradiation experiments in DOMO (which includes power ramp testing at burnup up to about 50 GWd/t), showed that the BWR-MOX fuel has very high performance and integrity comparable to that of BWR-UO_2 fuel under steady and transient conditions, even though the MOX fuel had a slightly higher fission gas release rate. Further information from the PIEs on the Tsuruga MOX fuels and DOMO fuels is expected to confirm this conclusion.

4. DEVELOPMENT OF MOX FUEL FABRICATION TECHNOLOGY

4.1 EQUIPMENT FOR FUEL FABRICATION

Hitachi has a capability of designing and supplying manufacturing equipment and a process control system for FBR fuel which includes powder handling process, pellet fabricating process, pin (fuel rod) fabricating process, and assembling process.

Hitachi has much experience and a wealth of knowledge on the design and manufacturing of UO_2 fuel fabrication equipment for BWRs. In supplying the fuel manufacturing equipment, we conducted R & D on key components of the apparatus to meet particular design requirements and appropriate to handling of fissile material.

4.2 DEVELOPMENT OF A FULLY AUTOMATED MOX BUNDLE ASSEMBLING APPARATUS

In the late 1980's Hitachi launched development programs on MOX fuel fabrication technology aimed at a future domestic MOX fuel fabrication plant. Major achievements included development of a fully automated MOX bundle assembling apparatus [14] and a conceptual design study for the MOX fuel fabrication plant. These were performed as contract R & D commissioned by MITI (Ministry of International Trade and Industry). In this paper, we include only a simple explanation of the former. In designing a MOX fuel fabrication plant, various factors, such as criticality, confinement, radiation and heat generation of fissile material, should be considered in a much more rigorous manner as compared to designing a UO_2 fuel fabrication plant. Particularly important is radiation level taken into account to avoid excessive radiation exposure for plant operators. For example, if we compare the equivalent dose rates during assembling of a MOX fuel bundle having fissile plutonium contents of 3 - 5 % with those during assembling of a typical BWR UO_2 fuel bundle, the following numbers are obtained:

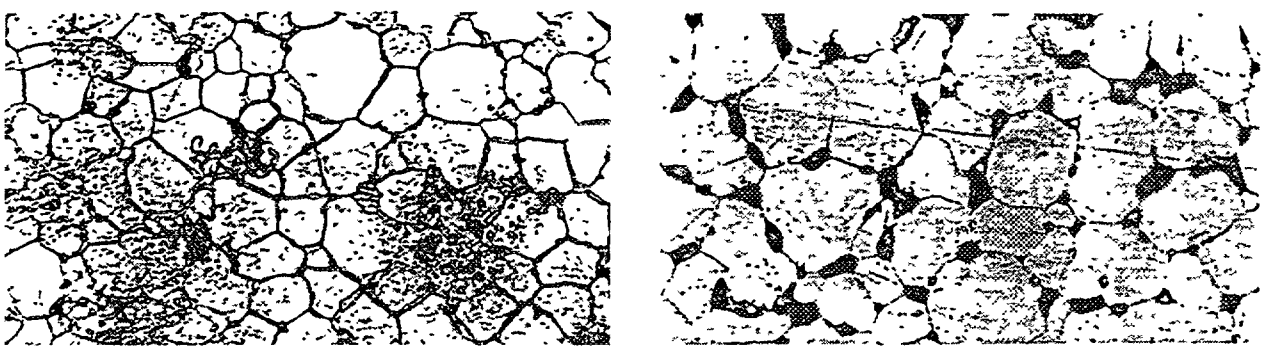
	MOX fuel bundle	UO_2 fuel bundle
assembly surface(mSv/h)	3 - 5	$\sim 2 \times 10^{-2}$
1 m from surface(mSv/h)	2×10^{-1} - 3×10^{-1}	$\sim 7 \times 10^{-4}$

The equivalent dose rates for a MOX fuel bundle are more than 200 times higher than those of the UO_2 fuel bundle. In the MOX bundle assembling process, therefore, it becomes important to automate as many features as possible for the bundle assembling apparatus and to use remote controlled steps. From this viewpoint we set out to develop the fully automated MOX bundle assembling apparatus.

The development, which took about four years, led to the apparatus shown in Fig. 11 [14]. It is composed of a fuel rod transfer system, fuel rod handling system, fuel bundle assembling station, fuel rod insertion/extraction system, and upper tie-plate installation/dismantling system. Fuel rods are assembled into a bundle by fully automated control in the fabrication process sequence; fuel rod transfer → rod number reading → spring insertion → fuel rod insertion → screwing connector rod on lower tie-plate → raising bundle assembling station → upper-tie plate installation → fixing upper-tie plate and perpendicular adjustment by screwing nuts on connector rods → fixing nuts by locking tab washers. The apparatus carries out tasks which require special skills for human workers, such as spring installation onto the upper plug, adjustment of the key thread on the plug after screwing the connector rod down to the lower tie-plate, etc., automatically and reliably through computer control. The main functions of the apparatus are summarized below.

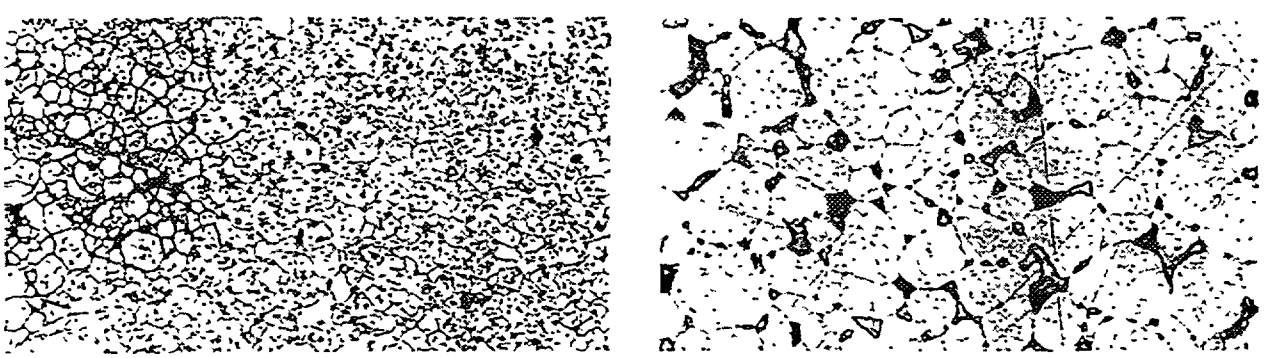
a) Fully automation of the bundle assembling process.
b) High adaptability and flexibility for various types of BWR fuel assembly designs.
c) Capability for disassembling a bundle and retrieving fuel rods to move them to a fuel rod transfer pallet automatically.

If something goes wrong during the assembling process, operators must approach the apparatus to fix it. They would be exposed to high radiation unless the fissile material is removed from the apparatus



Peripheral region ×500 Central region

(a) BN-MOX



Peripheral region ×500 Central region

(b) PNC-MOX

Fig.10 Typical microstructures of the BN-MOX and PNC-MOX pellets irradiated to about 50-55 GWd/t

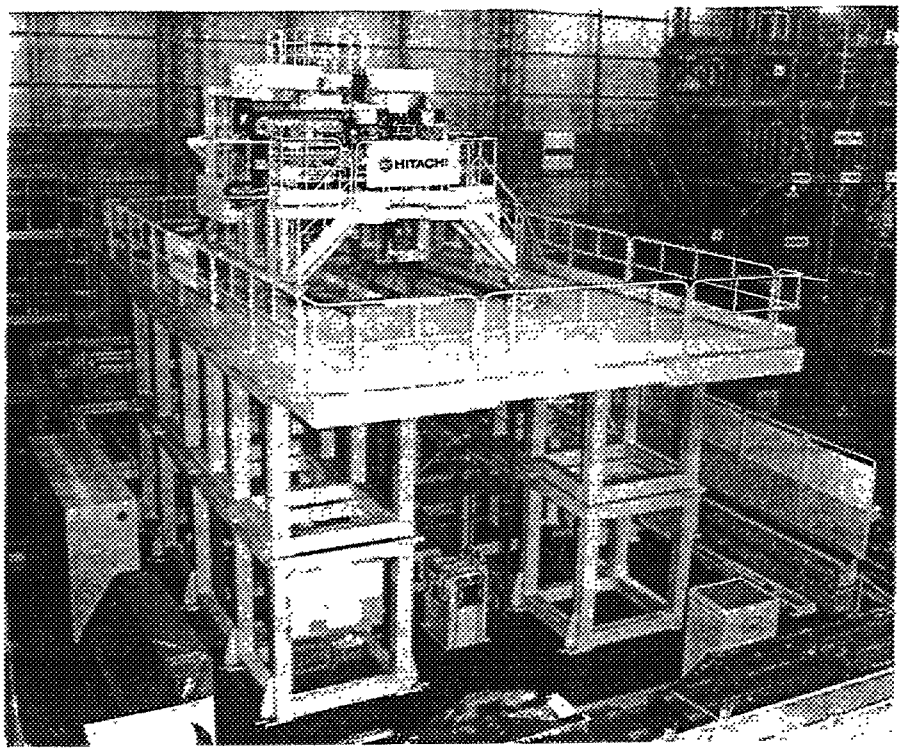


Fig.11 Fully automated MOX bundle assembling apparatus

before the repair can begin. Therefore item c) is essential to the automated apparatus. In fabrication tests using dummy MOX fuel rods, the apparatus demonstrated satisfactory performance [14].

5. CONCLUSION

In its nuclear energy policy the Japanese government is promoting systematic utilization of MOX fuel in LWRs, we have joined in the development of MOX fuel technology to allow future full scale utilization.

In this paper, we selected three subject areas, MOX core and fuel design, MOX fuel irradiation behavior, and MOX fuel fabrication technology, and described the main topics of R & D in them. In the MOX core and fuel design studies, we examined the compatibility of MOX fuel with the UO_2 core, the feasibility of a full MOX core, and the adaptability of MOX design methods based on a mock-up criticality experiment. Regarding MOX fuel irradiation behavior, we outlined the Tsuruga MOX irradiation program and the DOMO program, and showed that MOX fuel behavior was comparable to ordinary BWR UO_2 fuel behavior. In the MOX fuel fabrication technology, we dealt with the development of a fully automated MOX bundle assembling apparatus and explained its features.

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A REVIEW OF MULTIPLE RECYCLE OF PLUTONIUM IN LWRS

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Abstract

This paper presents a summary of the current status of the nuclear physics of multiple recycle in Light Water Reactors (LWRs). The paper draws attention to the principal nuclear physics issues of relevance and highlights where present knowledge is inadequate. The use of MOX fuel in PWRs is now a mature technology and one which is being applied on a commercial scale in many countries. LWR MOX was originally seen as an interim measure for using plutonium recovered from reprocessing plants prior to its eventual use in fast reactors; MOX allows useful energy to be recovered from plutonium which would otherwise remain in store and provides a convenient and proliferation resistant means of returning plutonium to the country of origin. However, in the last few years the prospect of the widespread introduction of commercial fast reactors has receded in most member states and the question arises of how best to manage the plutonium for the now extended period before it is required in fast reactors. The approach which is being developed in France and elsewhere is to reprocess LWR MOX assemblies after irradiation and to recycle the plutonium in a second generation of MOX assemblies and then perhaps a third and a fourth and so on. This is effective at managing separated plutonium stocks and also at extracting additional energy from the plutonium. But the isotopic quality of the plutonium is degraded with each irradiation cycle and this may at some point prevent any further recycling as LWR MOX. The question of how many generations of LWR MOX recycle are practical is one which, although it has been examined previously in the literature, has yet to be answered satisfactorily. This paper reviews the existing literature critically and provides a clearer picture of the assumptions underlying the past work and will highlight areas where past work has been deficient. From this comes a concise analysis of the technicalities of multiple MOX recycle and the areas of uncertainty which remain. The paper concludes that at present the question remains one for which there is as yet no satisfactory answer, largely because the basic nuclear data and lattice codes essential to define the technical limits are not yet sufficiently developed and validated.

1. WHY MULTIPLE RECYCLE?

Multiple recycle has always been taken for granted in the context of fast reactor fuel cycles, as it an essential feature of the fuel cycle. Fast reactor fuel cycles aim to extract the full 205 MeV fission energy from each ^{238}U atom after conversion to the more fissile ^{239}Pu by neutron capture. Due to limitations on fuel endurance, this cannot be achieved in a single step and multiple reprocessing and recycle of fast reactor fuel has therefore been considered an integral part of the fuel cycle. From the perspective of reactor physics, multiple recycle poses no particular problem in the fast reactor cycle, because all the plutonium isotopes are fissile (to a greater or lesser extent) in the fast spectrum and the balance between the various plutonium isotopes tends towards an equilibrium in which ^{239}Pu dominates, with the higher isotopes present in smaller amounts, the asymptotic composition depending on the balance between fissions and absorptions in the various isotopes.

Recycle of plutonium in thermal reactors, such as Light Water Reactors (LWRs), has always been regarded as an interim step on the way to substantiating fast reactor fuel cycles; it helps develop the necessary reprocessing and recycling technology, while allowing use of plutonium recovered from LWR reprocessing in the interim period before the fast reactor cycle is established. It also helps to manage the stockpile of separated plutonium. With the prospects of the large scale deployment of fast reactors having now receded in most countries, the role of LWR MOX in the interim has now become much more

important and because of the extended timescales on which fast reactors are now expected, the need to consider multiple recycle of LWR fuel has arisen.

Each complete cycle of MOX fuel manufacture, in-reactor irradiation, pond cooling and reprocessing occupies a period of about 10 years, which means that two such cycles will hold open the technology for a 20 year delay in the introduction of fast reactors. Failure to reprocess MOX assemblies and re-use the recovered plutonium in the thermal reactor MOX cycle would create strategic problems for LWR utilities, who would otherwise need to consider other interim storage/disposal options for the MOX assemblies.

All this assumes that fast reactors will be needed in the longer term. Although this may seem a remote possibility in the current political climate, it is clear that thermal reactors are ultimately not sustainable in the long term with finite uranium resources. If the view is taken that nuclear power can make a significant contribution to ever increasing energy requirements, the eventual need for fast reactors is undeniable. Moreover, fast reactors offer a major environmental benefit in that higher actinides are eventually converted to fission products which decay radioactively on much shorter timescales. No matter what geological disposal method is used for the waste products of a nuclear fuel cycle, the timescale for dispersion of radionuclides into the environment extends for at least several thousand years, by which time the fission products have largely decayed and it is such timescales for which the radiotoxicity estimates are relevant. Thus when the radionuclides buried in long term repositories eventually find their way back into the biosphere, fast reactors allow a reduction of up to two orders of magnitude in toxicity [1]. Notwithstanding the assumption of the eventual deployment of fast reactors, multiple recycle of thermal MOX of itself provides modest but worthwhile environmental benefits in terms of resource conservation and reduced toxic potential commitment [2] which strengthens the case for pursuing it. Because of the insensitivity of fast reactors to plutonium isotopic composition, it will be possible at any stage to transfer plutonium from thermal reactor multi-recycle into fast reactors.

2. PHYSICS OF MULTIPLE RECYCLE

2.1 Plutonium Fissions

Figure 1 shows the relative probabilities of the various plutonium isotopes to undergo fission in a fast reactor spectrum. Each bar shows the ratio of fissions to total neutron captures (fissions+absorptions) for the principal uranium and plutonium isotopes. Since the fission probabilities only differ to a modest extent between the odd and even plutonium isotopes, the fast reactor is not very sensitive to the source of the external plutonium used to initiate the fuel cycle.

Figure 1 also shows the corresponding fission probabilities for the same isotopes in a thermal reactor spectrum, such as that in a Pressurised Water Reactor (PWR) or Boiling Water Reactor (BWR). In this case the fission probabilities in the even plutonium isotopes are effectively zero and therein lies the major difference compared with the fast reactor cycle; in a thermal reactor spectrum the various plutonium isotopes are far from equivalent in terms of their contribution to the lifetime reactivity of fuel. In the thermal spectrum the even isotopes act as neutron absorbers and do not contribute to fissions. Their presence must therefore be compensated for by increasing the concentration of the odd isotopes. This is reflected in Table I which gives rough reactivity equivalence coefficients for the various plutonium isotopes in the fast and thermal reactor spectra according to an OECD study [3]. The difficulties associated with multiple recycle of thermal MOX arise from this lack of equivalence of the various isotopes combined with the degradation in isotopic quality in each recycle.

Because the probability of fissioning ^{242}Pu is essentially zero, the only means of removal is via neutron captures to higher actinides, which proceeds at a very slow rate. Therefore, unlike a fast reactor, the quantity of ^{242}Pu accumulates in each recycle and the fissile quality (defined as the fraction of ^{239}Pu + ^{241}Pu to total plutonium) decreases. The outcome is that in each succeeding phase of multiple the total plutonium requirement increases. This ultimately limits the number of such recycles which are practical.

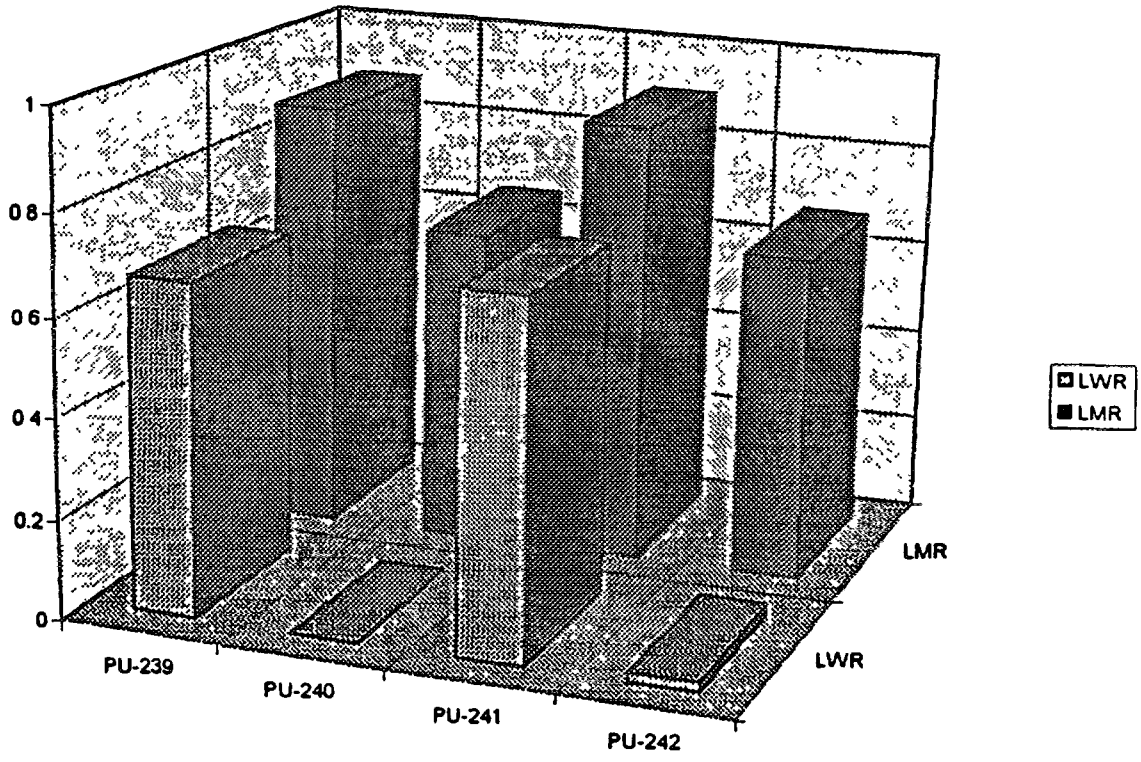

FIG. 1. *Fission Probability of Principal Plutonium Isotopes in LWR and Liquid Metal Fast Reactor (LMR) Neutron Spectra*

Table I **Indicative Reactivity Equivalence Factors of Plutonium Isotopesin LWR Spectrum Relative to ^{239}Pu (according to Ref. [1])**

^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
-1 0	+1.0	-0.4	+1.3	-1.4

This number is not, however, easy to determine, as there are a number of important variables which influence it, particularly the discharge burnup, the blending fraction of MOX to UO_2 in the reprocessing operations and the fuel design

2.2 Reactivity Behaviour

Figure 2 compares the reactivity characteristics of first and second generation PWR MOX assemblies with a UO_2 assembly that is equivalent in terms of lifetime reactivity. Both MOX assemblies have much flatter variations of reactivity with burnup than the UO_2 assembly. This is partly due to the different neutron absorption characteristics of MOX and partly due to the fact that ^{240}Pu, which is a neutron absorber in an LWR spectrum is partially converted to ^{241}Pu which is fissile. The important point to note is that the second generation MOX assembly curve is flatter than that of the first generation MOX. This is characteristic of MOX assemblies; the higher the initial loading of plutonium, the flatter the reactivity curve is with burnup. This has important implications for multiple recycle.

The main observation that needs to be made is that the reactivity of the fuel averaged over its lifetime in the core becomes insensitive to the initial plutonium loading at high plutonium concentrations,

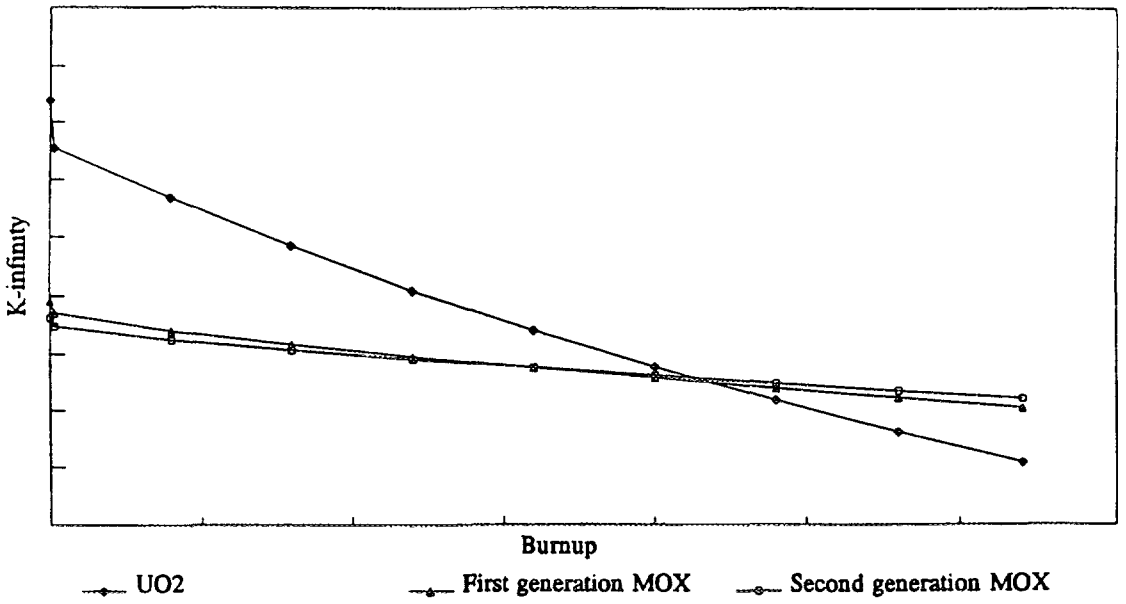

FIG. 2. Variation of k-infinity versus Burnup for UO_2, 1st and 2nd Generation MOX Assemblies

especially so if the isotopic quality of the plutonium is poor. The implication is that if the lifetime reactivity of the fuel is a little low for example, a large increase in the initial plutonium may be needed to correct it. As will be seen later, the reactivity predictions for MOX fuel with a high plutonium loading are presently very uncertain and this translates into a disproportionately larger uncertainty on the plutonium loading needed for lifetime reactivity equivalence. This behaviour is easy to understand when it is considered that with poor isotopic quality plutonium, the act of increasing the fissile plutonium loading to correct a shortfall in lifetime reactivity necessarily introduces more non-fissile plutonium which in turn necessitates additional fissile plutonium and so on. There must come a point when further increase of the plutonium is fruitless. This probably occurs at plutonium concentrations outside the range of practical interest, but it is fair to say that the precise point is not known with any confidence at present.

2.3 Void Coefficient

The void reactivity coefficient measures how much reactivity changes when the moderator density decreases when bubbles of steam form in an LWR. It is usual for the nuclear designer to ensure that the void coefficient is always negative, so that any voidage decreases reactivity and provides negative feedback. Provided a MOX assembly has only a modest plutonium concentration (less than about 10 w/o plutonium total), a negative void coefficient is guaranteed in all normal operating conditions. If the plutonium concentration is high enough, however, the void coefficient will become positive and this will ultimately define the effective limit. This can be understood most easily by noting that with total plutonium concentrations in excess of 10 w/o, a MOX assembly is beginning to resemble a fast reactor (where total plutonium concentrations in the range 15 to 25 w/o are generally used). Thus when the moderator is voided the spectrum tends towards that of a fast reactor and all the plutonium isotopes start contributing to fissions. Under these circumstances the void coefficient can be positive.

A recent OECD Study [3] conducted a comparison of void coefficient predictions for a MOX configuration with various total plutonium concentrations up to 14.4 w/o. The results showed reasonable agreement as to the magnitude of the void coefficient. In particular, there was agreement that with the specified plutonium isotopic composition, the void coefficient becomes positive at a total plutonium concentration of around 12 w/o. The value of this result lies not so much in the precise point at which the void coefficient changes sign, which depends on the plutonium isotopic characterisation, but in the fact that the various code systems are in reasonable agreement. This suggests that as far as determining the highest acceptable plutonium concentration, current codes can be relied on to give a reasonable indication.

2.4 Factors Determining Initial Plutonium Loading

In combination with the discharge burnup to which a MOX assembly is to be irradiated, the plutonium isotopic quality determines the initial plutonium loading. The source of plutonium for recycle as MOX is a major factor determining the practicability of multiple recycle in LWRs. The plutonium isotopic composition at discharge depends on the fuel type, whether UO_2 or MOX, and on the discharge burnup. For the reasons noted earlier, the plutonium isotopic quality from first generation MOX is worse than that of UO_2. That from any subsequent generations of MOX will be correspondingly worse. Similarly, plutonium isotopic quality degrades with increasing discharge burnup.

A major factor determining the plutonium isotopic quality in multiple recycle is the extent to which MOX and UO_2 assemblies are co-processed in the reprocessing plant. Due to the higher plutonium content of MOX assemblies and also the higher concentration of minor actinides, in current commercial reprocessing plants it will be necessary to reprocess UO_2 and MOX assemblies in a ratio of at least 3:1. Co-reprocessing in this way improves the quality of the plutonium recovered from MOX assemblies to something in between that of MOX and UO_2.

There is one further important point to note here. Although MOX assemblies supplied currently all have a moderator/fuel ratio identical to that of the co-resident UO_2 assemblies, the French are investigating the possibility of a dedicated MOX fuelled PWR with a higher than normal moderator/fuel ratio [4]. The greater moderation is closer to the optimum for plutonium fuel and enhances the negative moderator feedback coefficient. This design may allow the use of multi-recycle plutonium that would otherwise be unusable in a conventional LWR because the inherent moderator feedback is more favourable.

2.5 Uncertainties

An OECD study [1] completed recently included a series of reactor physics benchmark exercises to determine the degree of agreement between nuclear design codes when applied to LWR MOX configurations with poor isotopic quality and high plutonium concentrations. This is precisely the situation that is of interest here in considering multiple recycle. The reactivity benchmarks highlighted large discrepancies between the various solutions submitted, which if taken as indicative of the calculational uncertainty would be unacceptable for design purposes. Although some of the discrepancies seen can be explained by known limitations in some current nuclear design code packages, there remain significant underlying differences attributable to the nuclear data libraries that can only be resolved when experimental data to cover high plutonium loadings and poor quality plutonium.

Given the insensitivity of lifetime reactivity to initial plutonium loading noted earlier, which applies especially to high plutonium loadings, the degree of uncertainty in the nuclear codes is particularly significant. It is probably fair to say that the initial plutonium loadings are not known to within 1 or 2 w/o or more. This large uncertainty range impinges heavily on the technicalities of multiple recycle and is the single most important unknown at present.

3. MULTIPLE-RECYCLE SCENARIOS

3.1 Self-generated Recycle

The existing literature on multiple recycle in thermal reactors [3,5] concentrates on so called "self-generated recycle" scenarios in which a single thermal reactor is assumed to recycle only that plutonium that it generated itself. The scenarios assume an indefinite number of generations of MOX recycles and usually carry the analysis through to the point where equilibrium is attained. For example, in the PWR case considered in the OECD study, summarised in Table II the plutonium isotopic composition deteriorates in the first two generations of MOX recycle, but thereafter stabilises at the equilibrium value of 57% fissile/total that is sustainable indefinitely. Similarly in the BWR case the equilibrium plutonium composition corresponds to 59% Pu^{fiss}.

Table II **Summary of OECD Self Generated Multiple Recycle Scenario for a PWR (according to Ref. [1])**

MOX recycle generation	1	2	3	4	5
Total Pu in MOX (w/o)	4.72	5.83	6.89	7.51	8.05
Fissile fraction at discharge in MOX	0.659	0.618	0.595	0.583	0.573
MOX fraction in core	0.184	0.234	0.265	0.278	0.288

Assumptions:
PWR core with 3 batch 12 month refuelling giving low 30's GWd/t discharge burnup

But both these are for fuel cycles with a low discharge burnup of around 30 GWd/t. These are hardly relevant to scenarios that with 10 years for each cycle of fuel fabrication, irradiation, pond cooling and reprocessing and five or more such cycles will extend into the second half of the 21st century. With the higher discharge burnups more relevant to today's and future fuel management schemes, the isotopic quality at equilibrium would be considerably poorer and this may well restrict the number of multiple recycles possible.

The MOX fraction at equilibrium in such scenarios is relatively modest. For example the OECD plutonium report [3] quotes MOX core fractions varying from 18.4% in the first generation to 28.8% at equilibrium. In high burnup scenarios more relevant to the current and future situations, the self-generated MOX fraction is considerably lower, less than 20%. This happens because of the combined effects of needing a higher initial fissile inventory to reach high burnups and the degradation of plutonium fissile quality which accompanies higher burnups.

An important element in analyses of multiple recycle scenarios is the variation of the initial plutonium content of the MOX fuel through the various MOX generations. Because the isotopic quality degrades in each generation up to equilibrium, the total plutonium content rises correspondingly. For example, in the OECD report [3], the initial plutonium loading varies from 4.72 w/o Pu^{tot} (3.11 Pu^{fiss}) in the first generation to 8.05 w/o Pu^{tot} (4.61 Pu^{fiss}) at equilibrium. As seen earlier, the precise relationship between initial plutonium loading and isotopic quality is not well defined at present because of limitations with current nuclear design methods. This crucially affects the self-generation scenarios, as it is the key determinant of the self-generated MOX fraction. Curiously, Ref. [5] assumes a very different variation of plutonium loading in each successive generation; the first generation of MOX has 4.0 w/o Pu^{fiss}, and each subsequent generation has 4.1, 4.2, 4.3 w/o etc Pu^{fiss}. This assumption is less penalising than that of Ref. [3] even though the discharge burnup assumption (50 GWd/t) is considerably less favourable for the multiple recycle scenario. In spite of the current uncertainties, this appears to be a considerable underestimate, though it should be said that the main purpose of Ref. [5] was to examine the discharge isotopics of various MOX fuels and not on the details of the multiple recycle scenario.

3.2 Open Recycle

Though the concept of a self-generated recycle scenario is valid theoretically and is useful especially in strategic, logistic and environmental analyses of idealised fuel cycles, its relevance to realistic situations is questionable. The main criticism is the implicit assumption that there is a dedicated reprocessing plant that only reprocesses the fuel for the reactor in question, or equivalently, that commercial reprocessor's plants could be scheduled to operate in such a manner as to effectively simulate this situation. In practice, this is unlikely to be the case. When MOX fuel is reprocessed in current commercial reprocessing plants, even with dilution with UO_2 assemblies, it is possible that the overall throughput will be reduced in order to satisfy various safety constraints. Any such throughput limitations would apply even if a single MOX assembly is input, and this would make it very important to the reprocessor to ensure that whenever MOX assemblies are scheduled for reprocessing the MOX component is made as high as possible. Therefore a more realistic scenario to examine is one in which the mixing

Table III Example of Open Recycle Scenario (according to Ref. [3])

MOX recycle generation	1	2
Total Pu in MOX at loading (w/o)	7.00	9.82
Fissile Pu in MOX at loading (w/o)	4.67	5.70
Total Pu in MOX at recycle (w/o)	4.66	6.61
Fissile Pu in MOX at recycle (w/o)	2.35	3.04

Assumptions :
PWR core with 4 batch 12 month fuel cycle giving low 40's GWd/t discharge burnup
UO_2 and MOX assemblies recycled in 4:1 ratio
Post-irradiation cooling time 10 years

ratio of UO_2 to MOX corresponds to the 3:1 or 4:1 ratios potentially available from commercial reprocessors. This is the open recycle scenario, where the plutonium recycled into a reactor does not all necessarily originate from the same reactor.

As an example of an open recycle scenario, Table III summarises a case in which a PWR operating on a 1/4-reload 12 month fuel cycle with 43 GWd/t discharge burnup uses both first and second generation MOX. The plutonium for the second generation MOX is assumed to originate from co-reprocessing of UO_2 and first generation MOX assemblies in a 4:1 ratio. The UO_2 enrichment is 3.7 w/o and, using reactivity equivalence coefficients similar those quoted by the OECD [3], a 7.0 w/o total plutonium content is necessary in the first generation MOX assemblies to ensure the equivalent reactivity lifetime.

A total plutonium content of 9.8 w/o is needed for the second generation MOX assemblies. This should only be considered a very rough estimate, as the OECD equivalence coefficients are not strictly valid for other than the much lower plutonium contents and better quality plutonium isotopics for which they were derived. Nevertheless, it is clear that a high plutonium concentration approaching 10 w/o is needed in the second generation MOX because of the deterioration of plutonium isotopic quality. This is close to the 10 to 12 w/o region where the moderator void coefficient is expected to become positive. The analysis was not taken to the third generation MOX because the uncertainties implicit in the prediction of the initial plutonium concentration and because it is very likely that the void coefficient would in any case prove to be positive. The third generation MOX is therefore assumed to be stored until disposed of in some unspecified way or recycled to provide feedstock for fast reactors. The self-generated MOX fraction for this scenario is roughly 15%, the precise figure depending on whether any external plutonium feed is introduced to top up the MOX fraction to the 33% limit assumed to apply to standard PWRs.

This example contrasts with those presented in Refs. [2] and [5], which imply that indefinite multiple recycle is practicable and suggests that a more realistic scenario will only allow two recycles in LWRs before the practical limitations intervene. The difference stems from the 4:1 blending ratio assumed, in combination with the higher discharge burnup. It should be noted that 43 GWd/t is itself a modest burnup by today's standards and will probably be well overtaken in the decade or more that it will take before second recycle scenarios are established. This highlights the fact that truly realistic multiple recycles should really take account of the evolution of fuel management schemes during the long periods of time covered and not restrict themselves to idealised scenarios such as those considered here.

The above result should not be taken as implying that two recycles is the absolute limit, as there is clearly scope in the open recycle scenario to blend down with other plutonium sources in order to improve the isotopic quality of multiple recycle plutonium. Obvious candidates would be to blend with ex-military plutonium or to blend with plutonium derived from reprocessing campaigns in which no MOX is included. Flexibility in interchanging plutonium may therefore be the key to multiple recycle.

Overall, given the uncertainties in the current understanding of the basic physics, two recycles in LWRs seems reasonably assured, but to proceed further it is likely that a strategy of further blending will be necessary unless the multi-recycle is restricted to dedicated reactors with assembly designs optimised specifically for the purpose.

4. MATERIALS BALANCE

This section considers briefly the materials balance implication of multiple recycle in LWRs, which is relevant to plutonium management and environmental impact issues.

4.1 Plutonium

An important element of multiple recycle is the question of how much plutonium is generated or consumed. Self-generation recycle scenarios, if they proceed indefinitely, are by definition self-sufficient in plutonium and neither generate nor destroy it. However, indefinite multiple recycle is a theoretical abstraction and the cycle will in practice be interrupted at some time. At this point the working plutonium inventory must be attributed to the electricity generation. Thus the net plutonium production per GWye output depends on the point at which the recycle scenario is interrupted.

In contrast, open recycle scenarios have the potential to be net plutonium burners if the MOX fraction is in the region of 40%, the precise figure depending on details of the scenario. This figure arises from balancing the net plutonium production in the UO_2 component of the core with the net plutonium destruction in the MOX component. Because of the in-growth of fresh ^{239}Pu and the accumulation of ^{242}Pu in MOX assemblies, the plutonium content at discharge is invariably a significant fraction of that loaded initially. Thus, even though the plutonium balance in the MOX assemblies always destroys plutonium, because the conversion ratio in an LWR is well below 1.0, there is nevertheless a significant residual plutonium loading at discharge which needs to be accounted for.

4.2 Higher Actinides and Toxic Potential

An important feature of multiple recycle in LWRs is that there is a modest accumulation of higher actinides in the waste destined for geological disposal compared with a once through fuel cycle. This arises from the greater opportunity which multiple recycle allows for transformation of plutonium by neutron absorptions to higher actinides. This feature is highlighted in Ref 2, and causes the toxic potential associated with multiple recycle to increases by a modest amount for the first 300 years after discharge compared with a once through cycle. The biggest contribution to this increase comes from medium half-life isotopes which have largely decayed by 300 years, as have the bulk of the fission products. After this time the toxic potential is dominated by the plutonium committed to geological disposal (unless used at some point in a fast reactor), because of its long half life. Since all multiple recycle scenarios generate less plutonium per GWy of electrical energy generated than a once through cycle, the plutonium inventory implications are beneficial. In the scenario considered in Ref. [2], a 40% reduction in toxic potential is realised in the long term, consistent with other published results [6].

In the event that the plutonium from LWR recycle is eventually used in a fast reactor cycle, the ultimate toxic potential can be, as noted earlier, reduced by two orders of magnitude, as virtually all the plutonium isotopes (and also many of the higher actinides) are eventually fissioned in the fast spectrum.

5. CONCLUSIONS

The current status of multiple recycle in LWRs has been reviewed. This is a complicated area from both the logistics and the technicalities for which is it difficult to draw conclusions which are generally valid to all situations. Thus the practicability of multiple recycle depends on the detailed characterisation of the plutonium which is available for recycle and the extent of any blending of plutonium

from UO_2 and MOX assemblies in the recycle operations. Although the fundamental technology is well understood in principle, a serious handicap at present is the lack of validation evidence in support of reactor physics calculations applied to LWR MOX fuel with high initial plutonium contents and poor isotopic quality plutonium. The large spread in reactivity predictions seen in a recent international benchmark analysis highlights the need to acquire relevant validation information as a priority.

With the present limited physics understanding, it has been shown here that practical multiple recycle scenarios will most probably allow at least two recycles of plutonium as LWR MOX. Further recycles may require blending of plutonium from other sources to overcome the fundamental limitations arising from the need to maintain the void reactivity coefficient negative.

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EVALUATION OF THE MAXIMUM CONTENT OF A MOX-FUELED PRESSURIZED WATER REACTOR VERSUS ISOTOPIC COMPOSITION WITH RESPECT TO THE VOID COEFFICIENT

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Abstract

The study is within the framework of the feasibility of 100 % MOX recycling in a Pressurized Water Reactor. The objective is to determine the limit content of total plutonium in a MOX fuel with respect to the serious accident of core total draining, during which the reactivity effects must remain negative. Six isotopic vectors, from a very degraded plutonium to a plutonium with a high content of fissile nuclei, are studied. The very conservative result of 12 % for the whole vectors allows to confirm, a posteriori, the dimensioning of recent factories intended for the production of MOX fuel.

1. INTRODUCTION

The reprocessing of spent fuels, method chosen by France, enables a better management of the back end of the fuel cycle, providing to recycle the separate fissile matters : uranium, plutonium, and to solve the nuclear waste problem : minor actinides and fission products.

Initially the plutonium had to feed the fast neutron reactors, but for economical reasons the place of these ones among the other power plants is postponed. The FBR delay (several decades) increases the mean term interest for the recycling in water reactors with regard to the economical impact and to the impact on the balance of nuclear matters. In fact, the quality of the produced plutonium is deteriorated due to the decay of the 241 isotope into 241 americium (20 % in 5 years), thus a lost of fissile matter together with a poisoning of the fuel and an increase of its gamma activity.

The plutonium recycling in the PWRs started concretely in France with the introduction of six assemblies in the SENA reactor : 4 CEA-BN-RBU in 1974 and 2 FRAMATOME in 1976. The operation continued at an industrial scale in the Saint Laurent B1 reactor. The plutonium coming from UO_2 is loaded in a MOX form (Mixed OXide of uranium and plutonium) in only 30 % of the assemblies for control and safety reasons. The total plutonium content in the MOX is limited to 5.3 % and the burnup fraction to 39 GWd/ t, so that the mean consumption does not exceed 16 % in three irradiation cycles. The production of the UP2-800 factory being of about 8 tons / year, the 30 % MOX recycling is insufficient to absorb it, even if the number of concerned reactors is appreciably increased. Moreover, the recycled plutonium degrades itself in fissile isotopes and its reuse in PWR has still to be demonstrated. Furthermore, the UO_2 and MOX cohabitation makes local power problems inducing an expensive zoning at manufacturing level in the plutonium assembly. The present recycling remains limited whereas the performances and the manufacturing potential of plutonium fuel increase. So there is an interest to design an exclusively MOX loaded reactor, that also reduces the number of units concerned by the recycling.

Here and now, numerous studies, including this one, are carried out to design all-MOX PWR cores : modification of fuel, of moderation ratio, increase of control means efficiency, improvement of physical parameters of the core, limitation of damages to structures and vessel, multi-recycling capability ...

The penalizations due to the plutonium loading are directly related to the content of this one in the MOX. The calculations made by CEA and partners have shown that a load of about 8 % of first generation total plutonium is necessary to ensure a yearly cycle duration (280 Jepp) in a PWR 900

managed by quarter of core and exclusively MOX loaded (unloading at 43 000 MWd / t). The multi-recycling, even with dilution of a first generation plutonium, obliges inevitably to increase the content owing to a decrease in fissile nuclei and an increase in absorbent even nuclei. A second cycling without mixing in PWR requires a minimal content of total plutonium of about 10 %.

Within the framework of a 100 % MOX PWR feasibility study, the voiding is a dimensioning accident : it is necessary to determine the limit content in plutonium beyond which the core reactivity increases with respect to the nominal operating conditions. We will limit the study to the global draining. In another stage, the effects of a local draining, which necessitate tridimensional calculations and a preliminary definition of new control means, will be studied. The multiplicity of available isotopic compositions leads to carry out a parametric study versus several vectors (6). That is a generic study analyzing the behaviour of a parameter which could limit the penetration of the MOX in power plants, that is not a feasibility demonstration for the 100 % MOX recycling in the PWRs. The results show that the plutonium limit content varies from 12.5 % (vector with high concentration of fissile nuclei) to 15 % (degraded vector). These values are conservative and include the uncertainties related to calculations and to nuclear data.

2. DEFINITIONS AND OBJECTIVES

The voiding is a serious accident during which there is a loss of primary coolant caused, for instance, by the rupture of one of the legs of the circuit. The reactivity effect is defined by :

$$\Delta\rho(pcm)* = 10^5\ Ln\left(\frac{k\ effective\ without\ water}{K\ effective\ no\ min\ al}\right) \qquad [E.2/1]$$

* 1 pcm = 10^5 Δ k / k

The safety requires that this value is negative.

The plutonium content in the MOX is defined by : $T = \frac{mass(Pu + Am241)}{mass(Pu + U + Am241)}$

The limit content T_l corresponds to the value for which Δρ is equal to zero.

While modelling as well as one can the physical phenomena, the most conservative calculation options will be taken to determine the envelope values which may be used as reference in the feasibility studies.

We will search for a linear expression of the form :

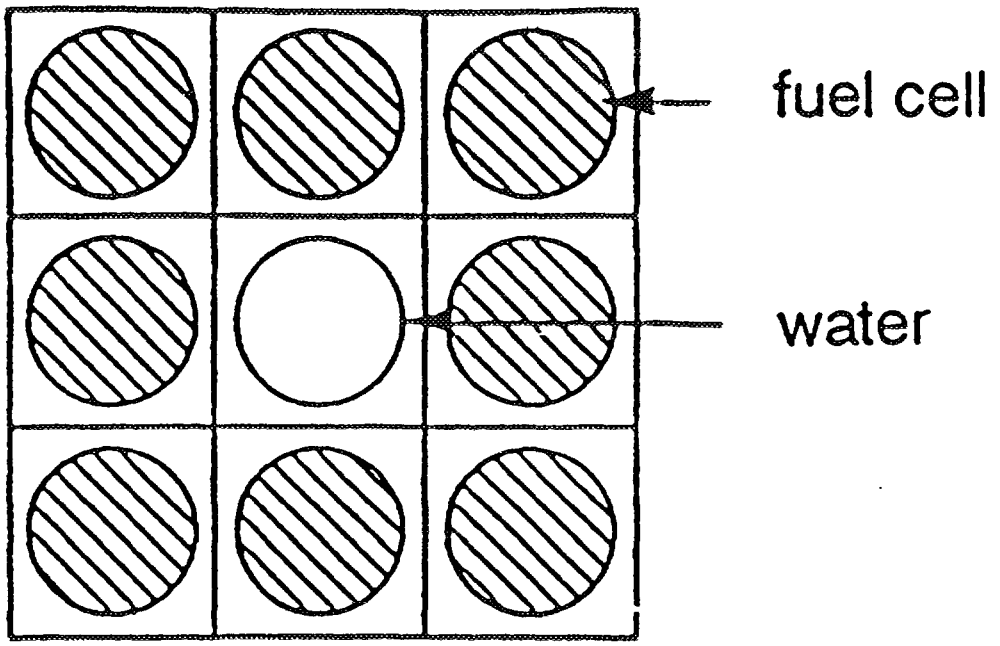
$$T_{limit}(\%) = a\,C_{Pu238} + b\,C_{Pu239} + c\,C_{Pu240} + d\,C_{Pu241} + e\,C_{Pu242} + f\,C_{Am241}$$

C_x being the mass percentage of isotope x. Once the limit content calculated for six realistic isotopic vectors, one only will have to solve a linear system of six equations with six unknowns to determine the a, b, c, d, e and f factors.

3. MODELLING. CALCULATION PLAN

The reference reactor in our calculations is a PWR 900 managed by quarter of core in yearly cycles (280 Jepp) with a mean burnup fraction at unloading of 43 GWd/ t. This choice is done for homogeneity purpose with the studies already performed elsewhere on the 100 % MOX PWR.

In order to limit the volume and the cost of calculations, the transport calculations will only be carried out on a supercell (figure F.3/1) representative of the PWR assembly with the correct moderation ratio (~ 2).



3/1. Supercell used in the transport calculation and modelling the PWR assembly.

The void effect represents the deviation between two $k_{effective}$ related to $k_{infinite}$ of the supercell by : $k_{eff} = k_{inf} / (1 + M^2 B^2)$. The leaks $M^2 B^2$ must be evaluated in both conditions. The $k_{infinite}$ and the migration area M^2 related to the fuel nature and to the moderation ratio are given by the transport calculation. According to the CEA's experience about the PWR core calculation, the mean buckling B^2 in nominal condition is empirically fixed to :

$$B^2_{\text{non drained}} = 2.5 \star B^2_{\text{geometric}}$$

with : $$B^2 geometric = \left(\frac{\pi}{He}\right)^2 + \left(\frac{J_o}{Re}\right)^2$$

where : He : extrapolated height of the core
Re : extrapolated radius of the core

In voided condition, there is practically no more thermalization and we can admit we are in fundamental mode, i.e. : $B^2_{\text{non drained}} = B^2_{\text{geometric}} = 3.24 \times 10^{-4}\ cm^{-2}$ (PWR 900).

The sensitivity to the buckling choice will be analyzed.

The transport calculations are carried out with the APOLLO2 code [R.3/1] which integrates the most recent results in reactor physics and in calculation methods. Moreover it includes a sophisticated model for the processing of resonances of heavy nuclei and isotope mixtures. The nuclear data come from the JEF 2.2 evaluation (CEA 93 tape). The used evolution chain includes 99 isotopes, 77 of which are fission products. The following nuclei are self-shielded in solved and non solved domains : U 235, U 238, Pu 238, Pu 239, Pu 240, Pu 241, Pu 242, Am 241, Zr natural. Ten equivoluminal self-shielding zones, with separate evolution, are represented in the fuel. The transport equation is solved through the collision probability method in the multicell approximation. The processing of interface currents is done with the UP_1 option (uniform in space and linearly isotropic in angle) for voided condition. The validity of the multicell calculation Pij has been tested in this case by comparison with an exact two-dimension probability calculation.

The qualification of voiding calculations through APOLLO2 code in the water lattices is going on at CEA, action supported by the partners EDF and FRAMATOME [R.3/2]. CEA has also participated to the OECD/NEA international Benchmark about the plutonium recycling in PWRs [R.3/3] in which there was a local void practice.

4. FORMULATION OF THE LIMIT CONTENT T_l

It is the content for which the reactivity $\Delta\rho$ in the expression [E.2/1] is equal to zero. During a progressive voiding of the core, three situations [illustrated in Figure 4/1] can occur.

Six realistic isotopic compositions are selected, from a very degraded plutonium (B) to a plutonium with a high content of fissile nuclei (M), and the cooling, manufacturing, storage periods are taken into account in Table 4/1.

For each of the six compositions, and for five contents (5, 10, 12.5, 13.75, 15 %), we carry out a transport calculation with the APOLLO2 code on the supercell F.3/1 with an evolution from 0 to 60 000 MWd/t, at a power of 38.3 MW/t and a boron concentration of 500 ppm. The calculations of $k_{effective}$ used in the voiding effect evaluation are carried out with concentrations of 150 MWd/t (saturated xenon) and 43 000 MWd/t (unloading, $\overline{BU}$ presence of fission products). The calculations are carried out with critical buckling, option usually taken for the calculation of cross sections in the core studies.

For conservative concern, we attempt to maximize, in algebraic value, the reactivity $\Delta\rho$ (E.2/1) i.e. to obtain the highest keff in drained condition and the lowest one in nominal condition. The leaks being defined, about 4 000 pcm in nominal condition and 23 000 pcm in drained condition, we choose to take $k_{infinite}$ at 43 000 MWd/t (minimum fissile matter) for the condition with water - 0 ppm of boron - and $k_{infinite}$ at 150 MWd/t (maximum fissile matter) for the condition without water. The evolution of the reactivity variation due to voiding versus plutonium content, for each isotopic vector, is shown in Figure 4/2.

The limit content is obtained in searching the zero of a 4th order polynomial, interpolating the void effect values between the five contents of total plutonium : 5 %, 10 %, 12.5 %, 13.75 % and 15 %. Table 4/2 gives the result of these calculations, as well as the corresponding values of void effects, recalculated by APOLLO2.

It will be noted that the rank of the limit contents is opposite to the one of the fissile plutonium contents, but the higher these ones, the lower will be the content necessary to ensure the cycle length.

Then, we search for a limit content expression of the form :

$$T_{limit}(\%) = a\,C_{Pu238} + b\,C_{Pu239} + c\,C_{Pu240} + d\,C_{Pu241} + e\,C_{Pu242} + f\,C_{Am241}$$

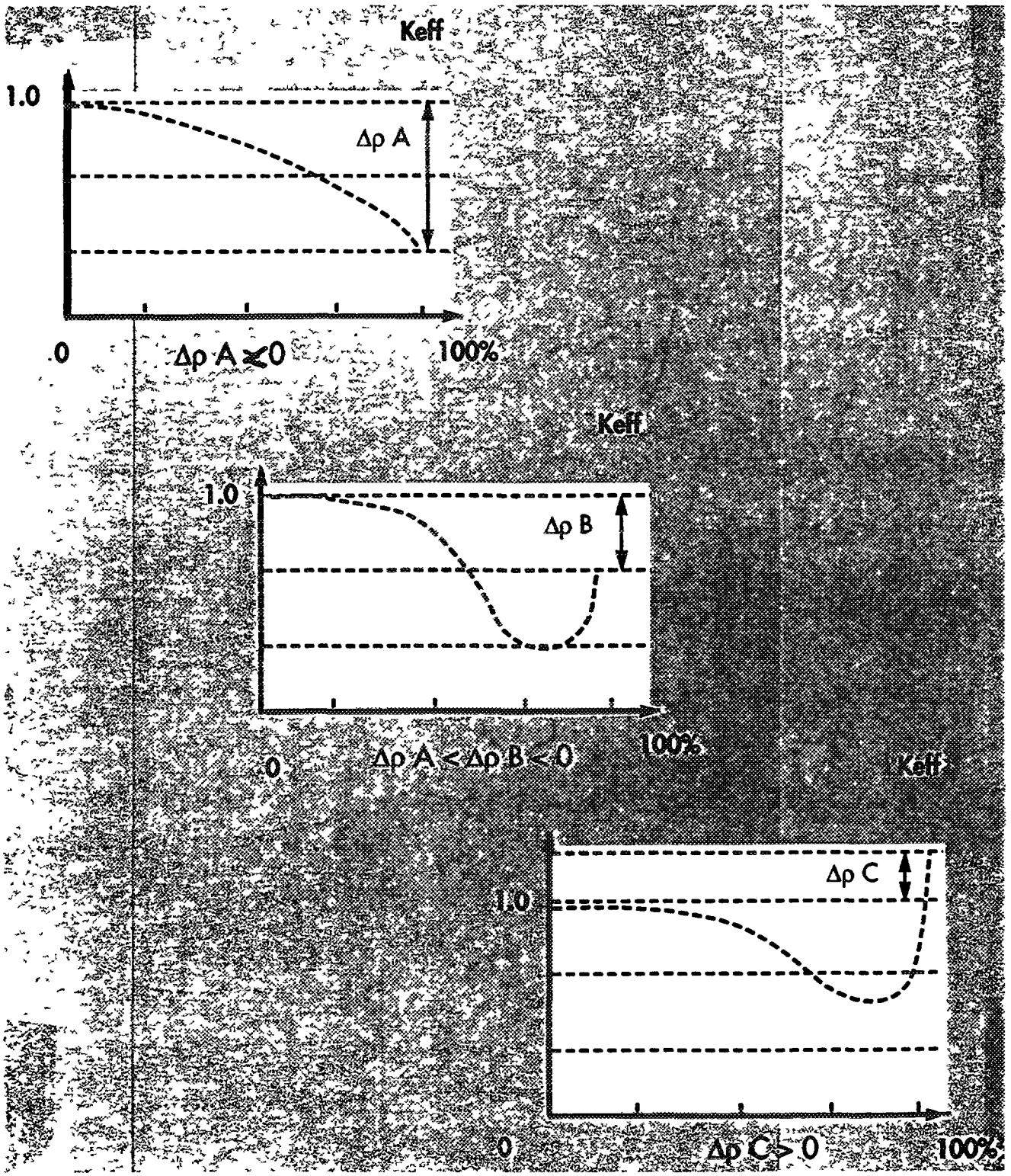
solving a linear system of six equations with six unknowns. The important contribution of U238 of the MOX to the void effect is explicitly taken into account by $1 - T_l$. We obtain the following coefficients :

a	b	c	d	e	f
0.108828	0.117737	0.196200	0.010116	0.222978	0.119437

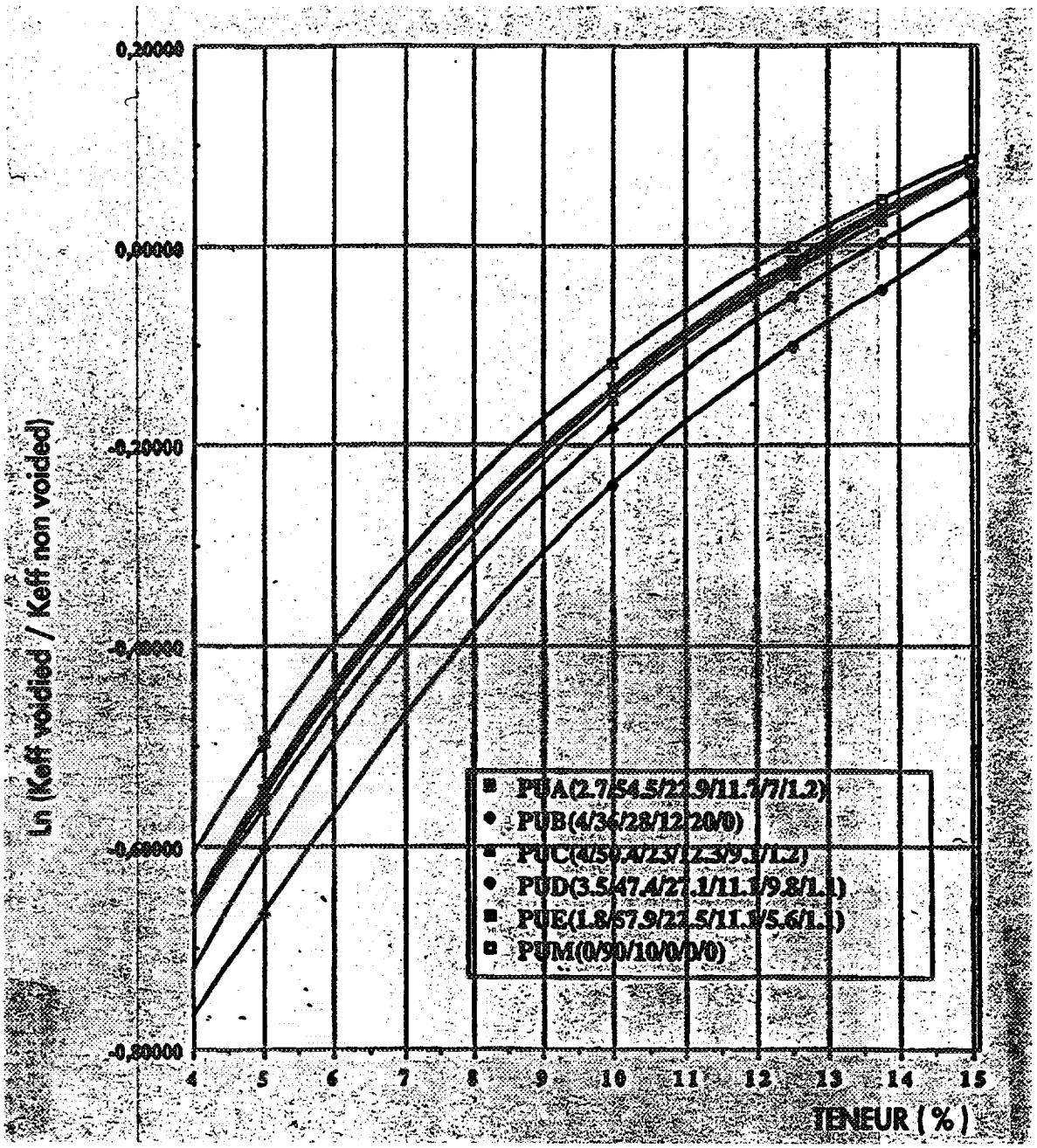
Verifications carried out on isotopic compositions away from the range of the 6 compositions of the system led to observe absolute deviations on the limit content up to 1 %. Such a linearity default may be attributed on one hand to the adopted conservatism, on the other hand to the fact the system is not well conditioned : the isotopic weights of the Pu 238 and Am 241 are not equilibrated as regard to other nuclei.

From that comes the idea to search for a formula with 4 coefficients, from the most differentiated compositions : A, B, D, M. Then we get :

$$T_1\,(\%) = 0.1156845\ C_{Pu239} + 0.214839\ C_{Pu240} + 0.0222565\ C_{Pu241} + 0.220139\ C_{Pu242}$$



4/1. $k_{effective}$ evolution during a progressive voiding of the core



4/2. Evolution of $Ln \frac{Keff_{voided}}{Keff_{non-voided}}$ *versus plutonium content.*

For the six reference vectors, the deviation between the two formulas does not exceed 0.1 % in absolute value (the most important contributions come from the isotopes Pu 239, Pu 240 and Pu 242). Further verifications were carried out on six other very different vectors Si. Table 4/3 shows the results.

The maximum deviation observed in the formula application does not exceed 1605 pcm (the void effects are generally of about several tens thousands of pcm), that represents approximately an error of 0.4 % in absolute content. One can assert that, within a very large range of isotopic compositions, the plutonium limit content in the MOX, as regard to the accident of total voidage of the core, is higher or equal to 12 %.

Table 4/1. Isotopic compositions selected for the study.

Origin of the Pu	Cooling	Storage	No	Pu238	Pu239	Pu240	Pu241	Pu242	Am241	Pu fiss.
Equilibrium cycle PWR 3.7 % (U5/U) 42 GWj/t-1/4	3 years	2 years	A	2.7	54.5	22.9	11.7	7	1.2	66.2
Benchmark OECD 1993 [R.3/3]	3 years	2 years	B	4	36	28	12	20	0.0	48.0
Equilibrium cycle PWR 4.5 % (U5/U) 55.5 GWj/t-1/6	3 years	2 years	C	4	50.4	23	12.3	9.1	1.2	62.7
MOX first generation 45 GWj/t-1/4	7 years		D	3.5	47.4	27.1	11.1	9.8	1.1	58.5
Equilibrium cycle PWR 3.25 % (U5/U) 33 GWj/t-1/3 *	3 years	2 years	E	1.83	57.93	22.5	11.06	5.6	1.08	68.99
Shield Plutonium			M	0.0	90	10	0.0	0.0	0.0	90.0

* standard first generation

Table 4/2. Limit contents obtained by interpolations from 5 calculated contents, and precision.

Composition	Pu A	Pu B	Pu C	Pu D	Pu E	Pu M
Limit content (%)	13.026	14.748	13.179	13.707	12.924	12.558
Recalculated effect (pcm)	+ 1.3	- 397	+30	- 10	+ 1.3	- 48

Table 4/3. Precision tests on the application of the 4-term formula.

	Isotopic composition PU8/PU9/PU40/PU41/PU41/AM41	Obtained limit content	void coefficient calculated by APOLLO2
Composition S1	5/51/12/10/22/0	13.5436 %	+ 306 pcm
Composition S2	4.5/35.5/30.2/14.4/14/1.4	13.9974 %	- 1216 pcm
Composition S3	4/42/27/11/15/1	14.2063 %	- 289 pcm
Composition S4	3/54/15/10/18/0	13.6546 %	+ 1291 pcm
Composition S5	2/60/26/5/6.5/0.5	14.0691 %	+ 1605 pcm
Composition S6	2.5/65/10/13/9/0.5	11.9385 %	- 570 pcm

5. ANALYSIS OF THE GLOBAL VOIDAGE IN A 100 % MOX PWR

The voiding is a complex phenomenon whose effects are related to a spectrum variation (Figure 5/1) disturbing drastically the competition between productions and absorptions of some isotopes having strong resonances. The following brief analysis will illustrate the phenomena.

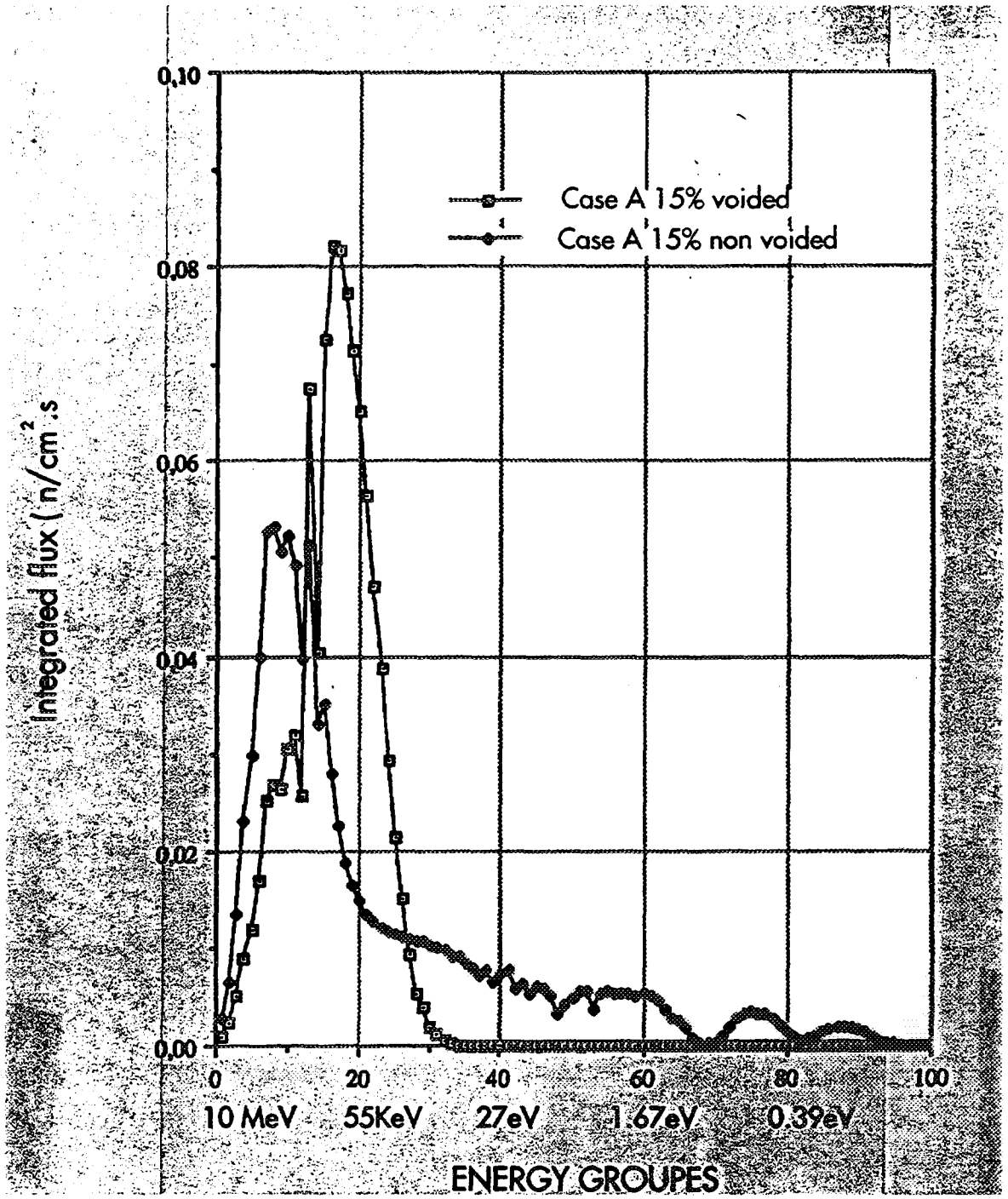
We choose the compositions A (realistic) and M (extreme). The voiding is carried out at 43000 MWd/t (unloading $\overline{BU}$, presence of fission products, very absorbent in thermal), on the supercell F.3/1. Figure 5/2 shows the evolution of infinite multiplication factor. The variation direction of the reactivity changes only from 60 % of void for the 15 % content, and from 80 % of void for the 10 % content. Generally, the situation becomes worrying only beyond 2/3 of the voiding, that could eventually allow to conceive a palliative.

The variations of contribution of each isotope of composition A versus the void fraction are shown on Figures 5/3 : 10 % content, and 5/4 : 15 % content.

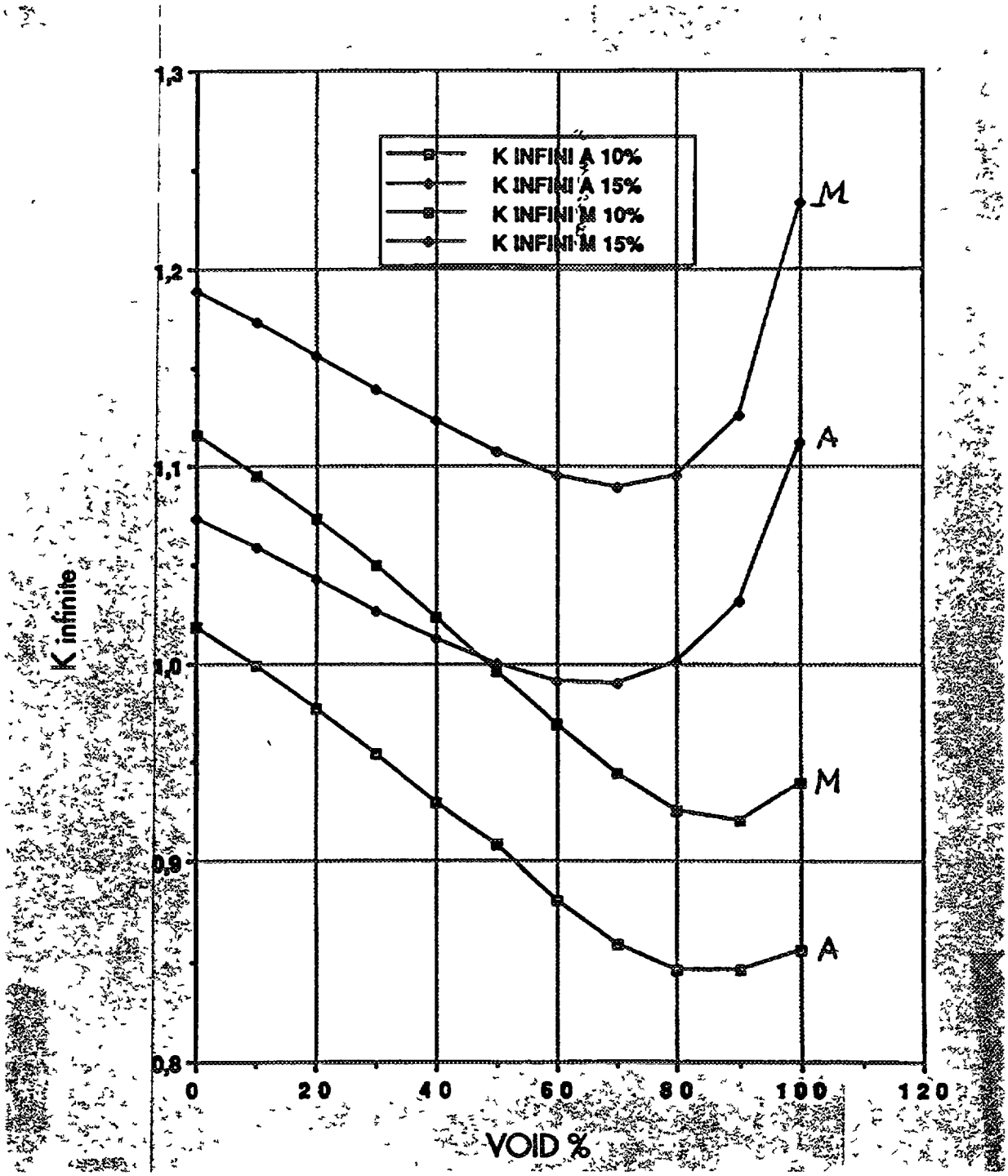
The most important contributions come from Uranium 238 (always negative and as a function of the content), from Plutonium 240 (always positive and not very depending on the content for the chosen vectors), from Plutonium 239 (negative, or close to zero for high concentrations), from Plutonium 242 (positive or equal to zero, very depending on the isotopic concentration).

The highly negative contribution of Uranium 238 is due to an important increase of absorptions in the 10 MeV - 1 KeV energy range (even though absorption cross sections are not very high in this energy range the number of absorptions is boosted by a very high increase of the neutron flux), which compensates amply the vanishing of the resonant capture (5 to 200 eV) when the spectrum becomes harder, due to voiding.

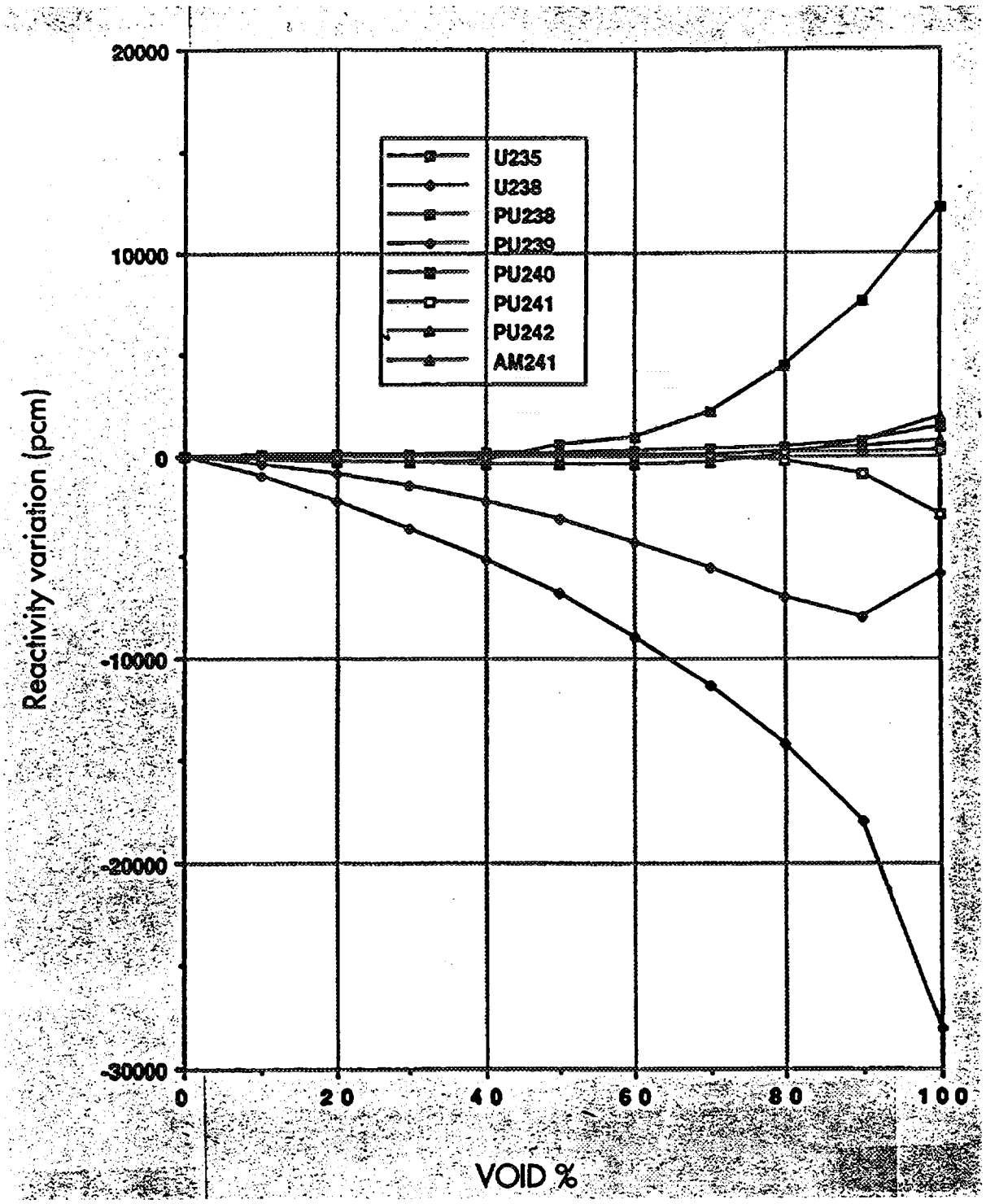
The Plutonium 240 has a capture centered on the thermal domain with a large resonance at 1 eV. The spectrum hardening reduces to zero this capture while increasing the productions over 1 MeV (decrease of $\alpha = \sigma_c / \sigma_f$ and increase of ν : number of neutrons emitted per fission), thus a highly positive contribution.



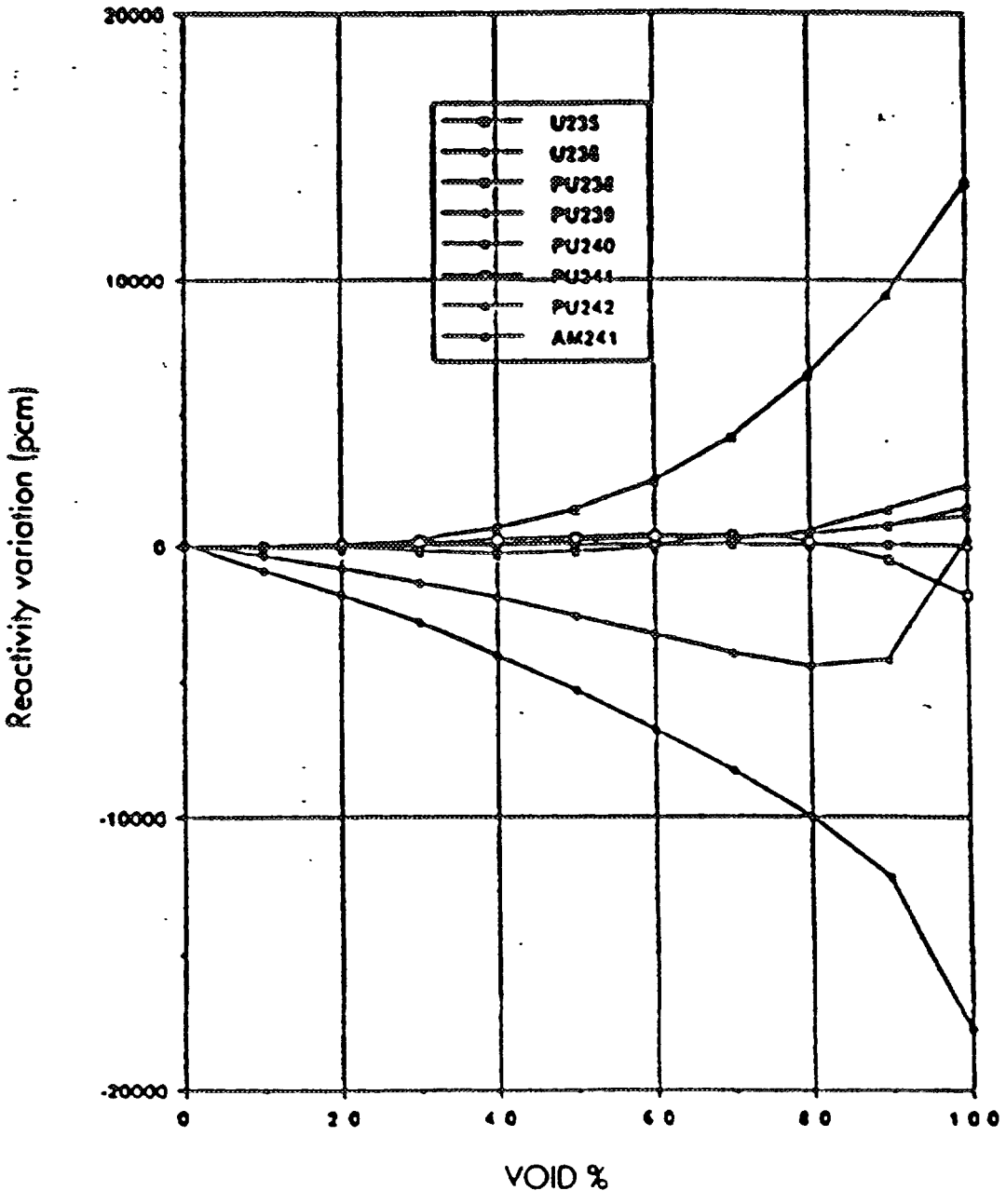
5/1. Variation of the spectrum in the assembly during a global voidage. Case : Pu A, content 15 %, 0 MWd/t.



5/2 Evolution of $k_{infinite}$ versus the void fraction during the voiding (at 43 GWd/t)



5/3. Evolution of cumulated contributions of main isotopes to the voiding coefficient versus the void fraction. Case A 10 % at 43 GWd/t.



5/4. Evolution of cumulated contributions of main isotopes to the voiding coefficient versus the void fraction. Case A 15 % at 43 GWd/t.

6. CONCLUSION

This generic study, carried out with the purpose of improving the use of plutonium in the PWRs, enabled us to determine a limit for the total plutonium content in the MOX fuel, as regard to the total voidage of the core. This conservative limit varies from 12.5 % (vector with high content of fissile plutonium : 90 %) to 15 % (vector with very degraded plutonium : 48 % fissile Pu). We remark the more the plutonium is degraded, the higher is the limit content.

The following linear formula allows us to approximate the limit content for any vector, as a function of mass concentrations of only 4 isotopes :

$$T_1\,(\%) = 0.1156845\, C_{Pu239} + 0.214839\, C_{Pu240} + 0.0222565\, C_{PU241} + 0.220139\, C_{Pu242}$$

The absolute precision is ± 0.4 %.

This formula can be used for PWR multi-recycling studies, where numerous isotopic compositions coming from plutonium mixtures are concerned.

A brief analysis of the voiding effect is carried out at assembly level, indicating the behaviour of the main responsible nuclei : U 238 (stabilizer) Pu 240 (destabilizer) ... We note that problems occur only beyond 60 % of void.

The contribution to the feasibility studies for PWR cores exclusively MOX loaded is continued by the search for content limits related to a cooling accident (opening of a valve in the secondary circuit), very penalizing in this type of core due to the very negative moderator coefficient and to the reduced control means.

ACKNOWLEDGEMENTS

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VENUS: A TOOL FOR THE RESEARCH OF THE NEUTRONIC BEHAVIOUR OF PU AND U FUEL

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Abstract

The Venus International Programme (VIP), executed at the VENUS facility, aimed at validating reactor codes in order to meet safety requirements enforced by licensing authorities. The VIP programme has validated the reactor codes DOT and WIMS (GOG and TWOTRAN) for reactivity and fission rate distribution calculations in PWR and BWR MOX configurations. The VIPO programme has been performed to cope with future developments of the nuclear fuel cycle. The tendency of going to higher burn-ups causes the need to investigate a possible positive void coefficient at high plutonium enrichments. The programme has validated the DORT, TORT, WIMS and KENO reactor codes for criticality calculations and KENO for fission rate distribution calculations so far. Work is going on to validate other codes as well. The VIPEX programme, which will be started end of 1995 or beginning of 1996, aims at measuring parameters that are mainly of interest for reactor operation. These parameters comprise β_{eff}, control rod worth, flux tilt, Americium effect and influence of moderator density.

1. INTRODUCTION

This paper describes the VENUS facility and the most recent programmes that have been executed at it. The aim of the VIP programme is to validate codes that are able to calculate a core with MOX-assemblies as well as a core with standard UO_2-assemblies, in order to meet safety requirements enforced by the licensing authorities. The VIPO programme has been set up for investigation of the void coefficient in highly enriched MOX assemblies in view of future developments of the fuel cycle that require higher burn-ups. This programme mainly focussed on safety issues. The VIPEX programme will mainly focus on reactor operation, but partially also on safety.

Some examples of comparisons between experimental and calculational values are given and discussed.

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 maintains fuel rods in a vertical position.

Criticality is reached by raising the water level within the vessel: via a series of pipes and pumps, water is led to the vessel from a storage tank which contains water when the reactor is not in operation. Reactivity control is thus obtained by controlling the water level in the vessel or by means of absorbing rods if experiments are performed at nominal water level.

Since the neutron flux is very low, no water circulation is needed in order to keep the fuel rods at low (room) temperature.

Whilst the water level goes up in the vessel, air is injected simultaneously into the jacket (a closed stainless-steel circular cavity located inside the vessel) and the two dump-tanks (i.e. two closed tanks each as large as the vessel itself) connected to the vessel. The dump-tanks and the jacket are kept under pressure, allowing the water level in the open vessel to go up whilst maintaining the water level constant both in the jacket and in the two dump-tanks (see figure 1).

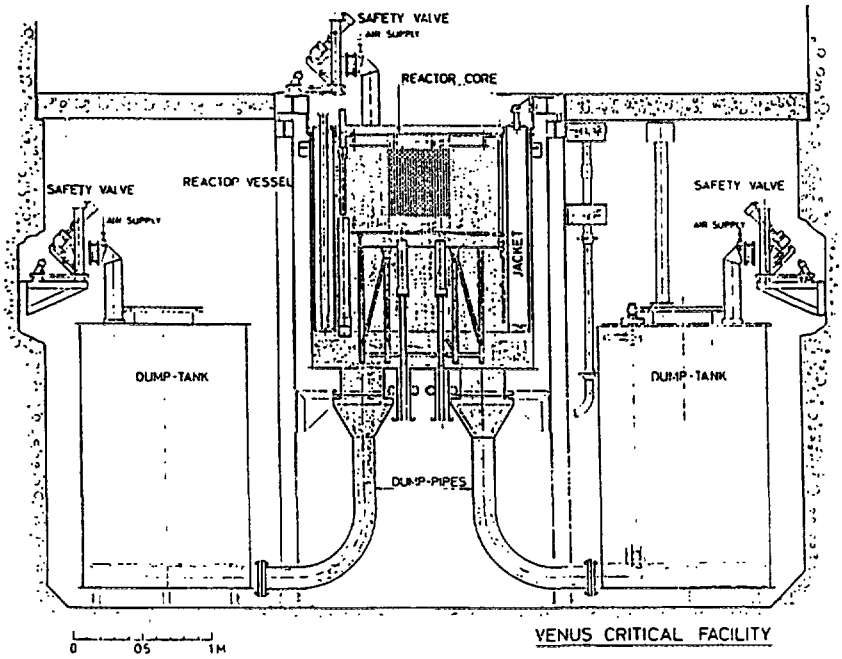

FIG. 1. Cross-section of the VENUS reactor

The reactor shut-down is induced by emptying the vessel. This can be done by opening the "safety-valves" and allowing the air to escape from the jacket and the dump-tanks, water is thereby transferred very rapidly into the jacket and the dump-tanks.

The VENUS reactor is characterized by a high experimental flexibility, featured by:

- direct access and manual handling of individual fuel pins
- easy loading of reactor
- easy removal of the reactor grids
- special (dismountable) pins and removable grid-parts
- easy water density simulation (up to 300°C)
- reactor control through water level variation or regulating rod
- boron poisoning (up to 2000 ppm)
- special nuclear measuring devices compatible with the grids (e.g. fission chambers)

Moreover, the reactor allows for experiments to be carried out with a high accuracy due to:

- strict tolerance for fuel pin geometry
- fuel pins with the same dimensional characteristics as LWR fuel

- strict tolerances of the reactor grid geometry (0.05 mm on pitch)
- no perturbation of the investigated configuration
- no local power perturbation due to fuel pin scanning
- no local perturbation of the fine structure due to the use of dismountable fuel pins

3. MEASURED PARAMETERS AND UNCERTAINTIES

The following parameters are measured at the VENUS reactor:

- the critical water level h_c
- the reactivity coefficient $\delta\varrho/\delta h$
- the axial fission rate distribution
- the horizontal fission rate distribution
- spectral indices F5/F9, F8/F9, C8/F9
- fission rate distribution inside fuel rod
- detector response

In the near future also β_{eff} measurements and tomography on fuel pellets will be performed.

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. This height was chosen in order not to be disturbed by the intermediate grid of the reactor (the upper water level had to be at least one extrapolation length below the grid), while on the other hand the level should not be too low for reasons of precision of the measurements.

The random uncertainty of the value of the critical level is 0.02 cm, the systematic uncertainty is about 0.07 cm. The random uncertainty is due to the reading of the water level and the temperature correction. The systematic uncertainty is mainly due to the determination of the lower level of the active part of the MOX and Gd fuel.

The comparison of the same or similar configurations will only be affected by the random uncertainty.

With the help of the critical level measurements also changes in reactivity can be measured. These changes can be due to:

- Am effect
- changes in moderator density
- replacements of absorber rods (Gd, B4C, AgInCd)

The reactivity coefficient is measured to make a link between the uncertainty of the critical level measurement and the uncertainty of the reactivity. Normally a value for $\delta\varrho/\delta h$ 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 axial and horizontal fission rate distribution can be used directly for validation purposes.

Moreover, the axial fission rate distribution is used to determine the buckling. In this way a measure of the axial leakage is obtained, which is used for validation of two-dimensional codes. The axial buckling is measured with an uncertainty of 2 to 4%. The axial fission rate distribution is measured with an uncertainty from point to point of approximately 2% for UO_2 rods and 3% for MOX rods. The higher uncertainty for MOX is due to the higher background correction.

For the horizontal fission rate distribution the uncertainty from pin to pin is estimated to be 1% for UO_2 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_2 and MOX), an additional systematic uncertainty of 1.4% has to be taken into account due to different fission yield and self-shielding corrections for U and Pu.

In some validation cases pin-to-pin 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 spectral indices can be measured. A spectral index is the ratio of two different reaction rates. Spectral indices that are measured at the VENUS are F5/F9, F8/F9 and C8/F9. Random uncertainties on the measured values are between 2 and 2. 5%. Since the applied measurement method is in a qualification stage, an extra systematic contribution of 2. 5% is added at the moment.

Radial fission rate distributions inside a fuel rod have been measured in the past. At present an effort is done to establish a new measurement method for this. Tilt factors will also be measured by this method. No details can be given with respect to the uncertainties of the envisaged method.

4. RECENT PROGRAMMES PERFORMED AT THE VENUS REACTOR

4.1 The VIP programme

From 1990 to 1992 the Pu recycling in LWR's was investigated. The programme, called VIP (VENUS International Programme), used fuel with high Pu and Gd content. The aim of the VIP programme was the validation of reactor codes with respect to MOX-fuel for both PWR's and BWR's, in order to meet safety requirements enforced by the licensing authorities. It focused on the criticality and fission rate distribution calculation. A comparison of some experimental and calculated data [1,2] is given here.

The programme was divided into two stages:

- VIP - BWR co-sponsored by Toshiba, Hitachi, SCK-CEN and BN.
  Three mock-ups were considered:
  mock-up 1: All UO_2: 8 x 8 subassembly
  mock-up 2: All MOX: 8 x 8 subassembly
  mock-up 3: Island MOX: 8 x 8 subassembly

- VIP - PWR co-sponsored by Mitsubishi, BNFL, SCK-CEN and BN.

  mock-up 1: All MOX 17 x 17 subassembly
  mock-up 2: MOX- Gd 17 x 17 subassembly

Figures 2 and 3 show examples of the BWR and PWR configurations, respectively.

Table 1 shows the results of the k_{eff} calculations for different VIP configurations by the codes DOT, GOG and TWOTRAN. The calculated k_{eff} values agree reasonably well with the experiment. Only for the gadolinium case a significant deviation can be observed, due to the absence of a specific treatment of Gd fuel [1] .

Figure 4 shows a typical result of the fission rate distribution of the BWR ALL MOX configuration, calculated with DOT. Figure 5 does the same for a WIMS calculation. Table 2 outlines the main results of the fission rate distribution calculations.

Table 1 **Results of k_{eff} calculations for the experimental critical level (1.00000 ± 0.00025) in the VIP programme**

Configuration	DOT	GOG	TWOTRAN
BWR ALL UO_2	1.006	0.9910	0.9971
BWR ALL MOX	0.9982	0.9943	1.0006
BWR I-MOX	0.996		
PWR	0.9998	0.9972	1.0011
PWR Gd	1.015	0.9842	0.9886

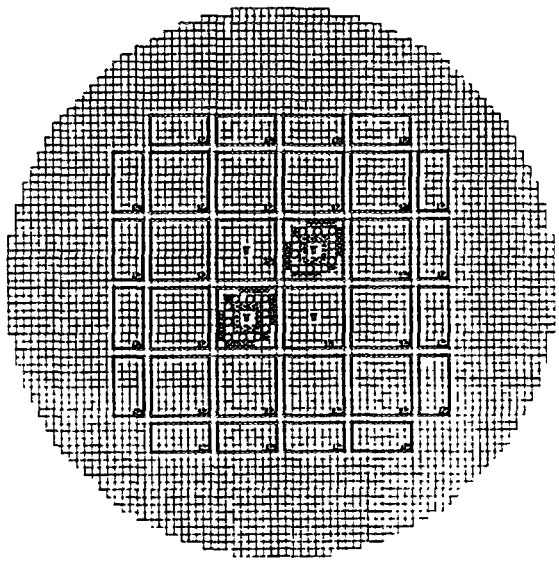

Figure 2. Schematic view of VIP BWR configuration

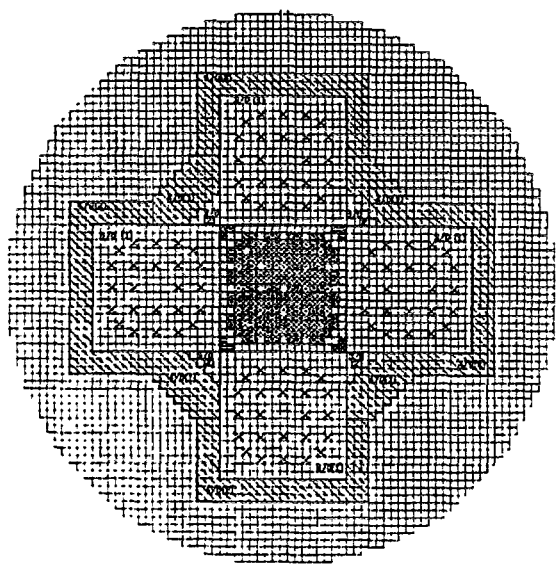

Figure 3 Schematic view of VIP PWR configuration

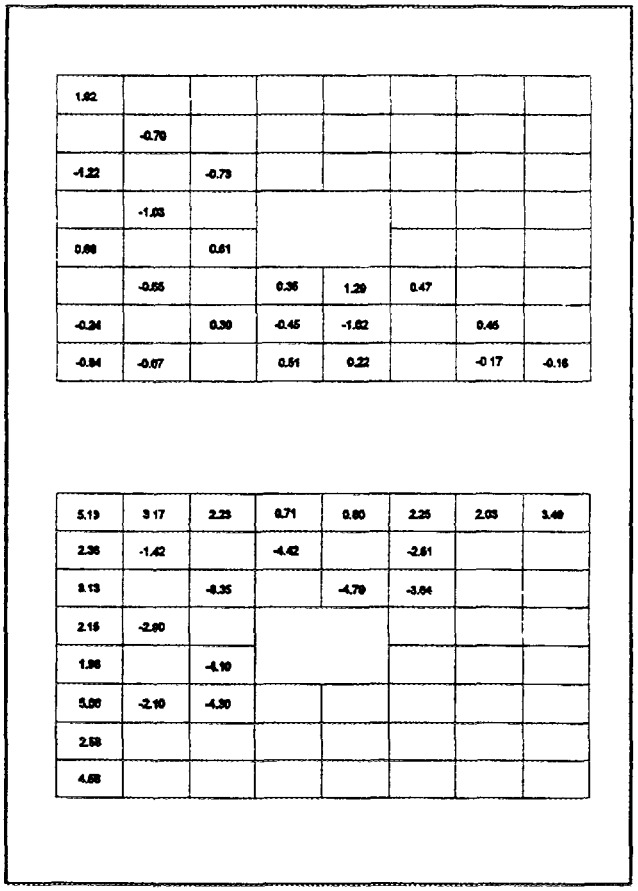

Figure 4 (C-E)/E values of fission rate distribution in VIP BWR ALL-MOX configuration calculated by DOT

Table 2 **Standard deviation of (C-E)/E values (%) in the VIP programme. The first value in each cell is the standard deviation in the UO_2 assembly, the second is for the MOX assembly**

Configuration	DOT	GOG	TWOTRAN
BWR ALL UO_2		1.2 2.6	1.2 2.8
BWR ALL MOX	0.8 3.7	1.1 1.1	0.8 2.2
BWR I-MOX	1.4 2.1		
PWR	1.4 1.7	1.1 1.5	1.1 1.1
PWR Gd		1.0 1.4	1.1 1.6

The uncertainty of the experimental values is 1.4% from pin to pin for MOX rods and 1.0% for UO_2 rods. Since the number of measured pins is about 50 with the majority being MOX rods, one may expect a standard deviation of 0.2 to 0.3%. The values listed in Table 2 are above this value, indicating that there are other than statistical influences only, but on the whole they are very satisfactory.

4.2 The VIPO programme

Since 1993, the void coefficient in LWR's was investigated. Calculations had 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), co-sponsored by EDF, BNFL, NFI, MHI, SCK-CEN and BN, was devoted to the measurements of the perturbation caused by a void bubble in an 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.

Measured and calculated criticality and fission rate distributions [1,2] are compared here.

Figure 6 shows an example of a VIPO configuration. Table 3 shows the results of the k_{eff} calculations by the codes DORT, TORT and KENO.

The DORT calculations could only be performed for the non-voided configurations since it is a two-dimensional code. The TORT and KENO results are very satisfactory. The DORT results show somewhat higher deviations.

Within the VIPO programme the horizontal fission rate distributions of two configurations have been calculated at the moment, the H-MOX configurations without void and with 10 by 10 void. Since the calculations for a configuration with void demand three-dimensional modelling, the nonvoided configuration has been calculated with both two- and three-dimensional codes for a good comparison.

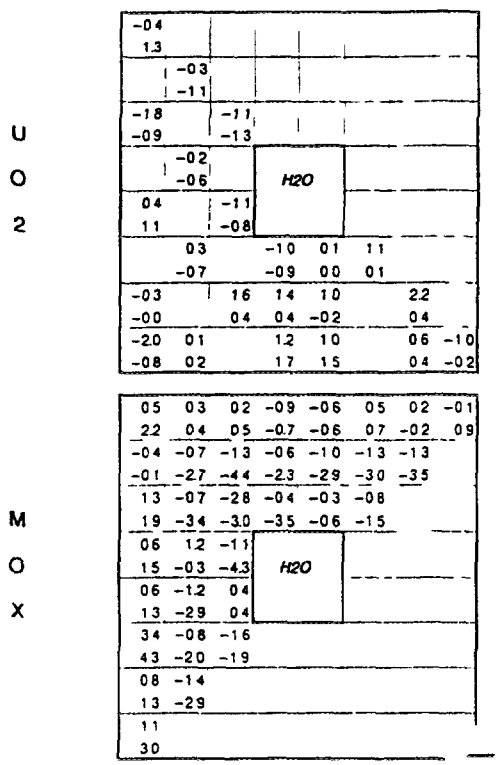

Figure 5 As fig 4. but calculated by GOG (upper value) and TWOTRAN (lower value)

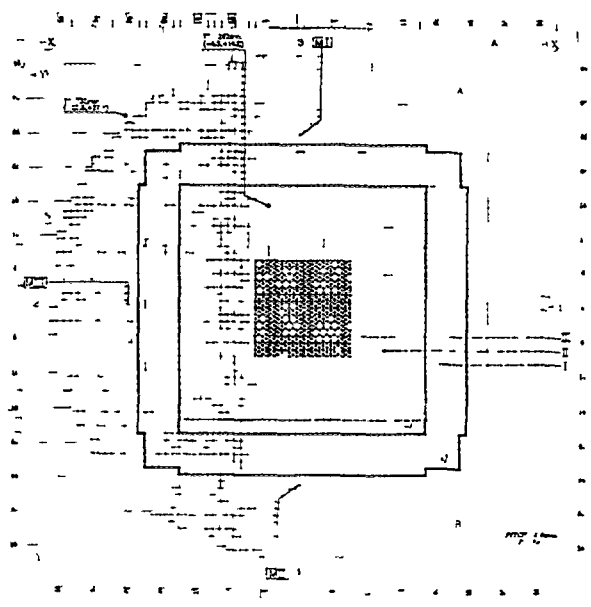

Figure 6 Schematic view of the VIPO configuration

Table 3 **Results of k_{eff} calculations for the experimental critical level (1.00000 ± 0.00025) in the VIPO programme**

Configuration	DORT	TORT	KENO
H-MOX no void	1.0074	1.0010	0.99673
H-MOX no void	1.0090	1.0024	0.99716
H-MOX 10 by 10 void		1.0023	0.99674
H-MOX 10 by 10 void		1.0014	0.99799
H-MOX 14 by 14 void		1.0020	0.99391

Figure 7 shows a typical example of the comparison between calculations and experiment, a KENO calculation of the H-MOX configuration with 10 by 10 void.

Table 4 lists the range and the standard deviations of the (C-E)/E values.

Although a little bit beyond the statistical expectation, the calculational results for both configurations are rather stable. No particular problems are observed at the UO_2-MOX border, where one could expect most problems.

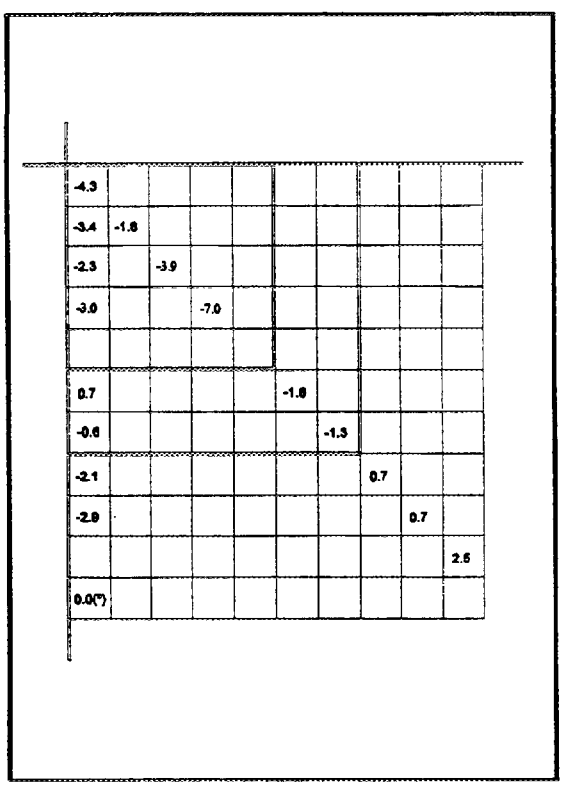

Figure 7. (C-E)/E values of fission rate distribution in voided VIPO configuration calculated by KENO

Table 4 Range and standard deviation of (C-E)/E values (%)

Configuration	TWOTRAN	KENO
H-MOX no void	-2.0 to + 2.0 (range) 1.22 (standard dev.)	-3.5 to + 1.8 1.44
H-MOX 10 by 10		-7.0 to + 2.5 2.31

5. FUTURE PROGRAMMES TO BE PERFORMED AT THE VENUS REACTOR

The VIP programme essentially concentrated on licensing problems that were rather urgent for some facility operators in the early 90's. The VIPO programme looks more to the future, where possible safety problems are investigated related to the use of high burn-up MOX fuels.

In the near future the VIPEX programme (VIP Extension) is planned at the VENUS reactor. This programme will focus specifically on plant operation with MOX fuel assemblies. Parameters that will be measured in the framework of this programme will be:

- Am effect.
  A configuration with a 17 by 17 MOX assembly from the VIP programme will be reloaded rod by rod and the critical height will be determined. This will be compared with the critical height that has been measured 4 years ago in exactly the same configuration.

- β_{eff}
  The smaller β_{eff} for Pu fuel will have its implications for reactor control. This parameter will be measured by small changes of the critical mass. Other measurement methods are evaluated at the moment for checking the critical mass change results.

- control rod worth.
  Due to the less thermal neutron spectrum of MOX the anti-reactivity of the control rods decreases. The control rod worth will be measured by introducing part by part a control rod (or several control rods) in the MOX configuration.
- In connection with this overmoderated MOX configurations are studied as well. These configurations will have a more thermal neutron spectrum, thus increasing the control rod worth. Although less MOX will be present in these configurations, a higher fission rate per rod is expected due to the higher fission cross-section at lower neutron energies.

- flux tilt.
  The difference in fission rate will be determined inside a rod at the edge of the MOX assembly. Due to the large difference between the neutron spectra in the MOX assembly and the UO_2 assembly, the fission rate inside the border rods will vary a lot, probably causing limitations on operation.

6. CONCLUSIONS

With the VIP and VIPO programmes highly accurate experimental data for MOX and UO_2 fuel element mock-ups have been achieved with the SCK-CEN critical facility VENUS.

The VIP programme has demonstrated the validity of DOT, GOG and TWOTRAN for calculating fission rate distributions in present-day fuel assemblies.

The VIPO programme has demonstrated the validity of KENO Va for calculating fission rate distributions in MOX fuel assemblies with and without voids within 2. 5% precision.

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MEASUREMENT AND ANALYSIS OF MOX PHYSICAL PROPERTIES

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Abstract

A programme of physical properties measurements has been carried out on MOX fuel manufactured using the Short Binderless Route (SBR) by BNFL and on MOX fuel manufactured using the MIMAS process by Belgonucleaire. The programme includes the following work: - Determination of the melting point of MOX fuel. - The measurement of the thermal expansion of MOX fuel. - Determination of the thermal diffusivity of MOX fuel. This paper will describe the programme of measurements and summarise the results obtained as well as analysing the results in comparison with previous published work, where applicable.

1. INTRODUCTION

A programme of fuel properties measurements for unirradiated uranium dioxide and uranium/plutonium mixed oxide (MOX) fuel has been carried out. The programme covered the measurement of thermal expansion, melting point and thermal diffusivity, together with appropriate pre- and post-test characterisations. This paper reviews the results of these measurements.

2. SAMPLE FABRICATION

The unirradiated MOX fuel samples were manufactured by BNFL using the Short Binderless Route (SBR). This involves blending UO_2 and PuO_2 in a high energy attritor mill and conditioning the powder in a speroidiser prior to pellet pressing and sintering. The UO_2 samples were manufactured at BNFL Springfields using depleted UO_2 powder produced via the Integrated Dry Route (IDR) and contained CONPOR pore former. The MOX fuel was manufactured at Sellafield from depleted IDR UO_2 powder blended with PuO_2 powder and also contained CONPOR. Hyperstoichiometric MOX fuel, with an O/M ratio of 2.02, was produced by controlling the oxygen potential of the furnace atmosphere during the final sintering of the samples.

In addition to these UO_2 and MOX samples fabricated by BNFL, MOX samples were prepared from a fuel rod which had been manufactured by Belgonucleaire. This MOX fuel was manufactured by the MIMAS process, which involves the addition of UO_2 and PuO_2 powders to form a master blend which is then added to UO_2 to form the MOX powder of the correct enrichment prior to pelleting.

A more complete description of the characterisation of the fuel samples is given Table 1.

Table 1: Fuel Variant Nominal Characteristics

Characteristic	UO_2	SBR MOX (O/M=2.00)	SBR MOX (O/M=2.02)	MIMAS MOX
Pu/(U+Pu) (%)	-	9.91	9.91	9.64
Pu Agglomerate Size (μm)	-	< 100	< 100	< 214
O/M Ratio	2.00	2.00	2.02	2.00
Density (%TD)	95.05	94.45	94.48	93.81
Diameter (mm)	8.202	7.699	7.678	8.203
Length (mm)	9.384	8.940	8.904	11.830
Mean Pore Size (all, μm)	2.6	2.0	2.0	1.7
Mean Pore Size (> 5μm)	7.1	7.7	7.6	8.1
No. of Pores > 100μm (%)	0.03	0	0.03	0.03
Grain Size (μm)	7.6-8.1	8.0-8.1	8.0-8.1	0-12

Figures 1a - 1c show ceramographs of archive UO_2, SBR $(U,Pu)O_2$ and SBR $(U,Pu)O_{2.02}$, respectively. In addition, Figures 2a and 2b show α-autoradiographs of the SBR $(U,Pu)O_2$ and SBR $(U,Pu)O_{2.02}$, respectively.

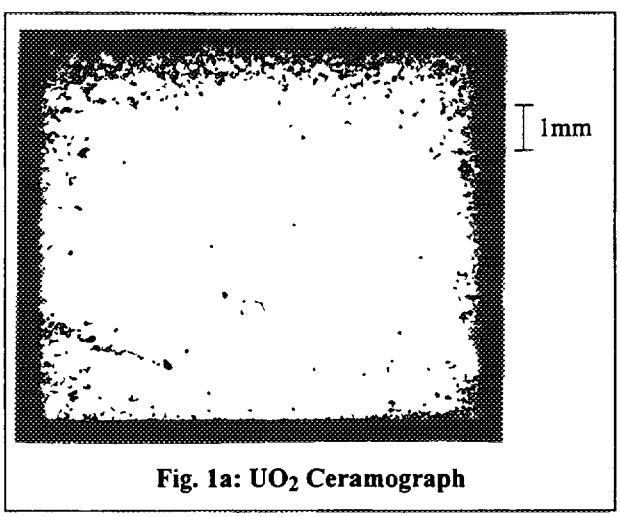

Fig. 1a: UO_2 Ceramograph

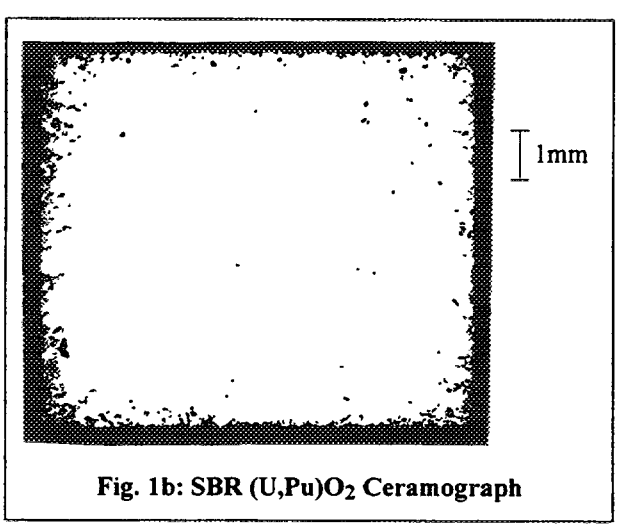

Fig. 1b: SBR $(U,Pu)O_2$ Ceramograph

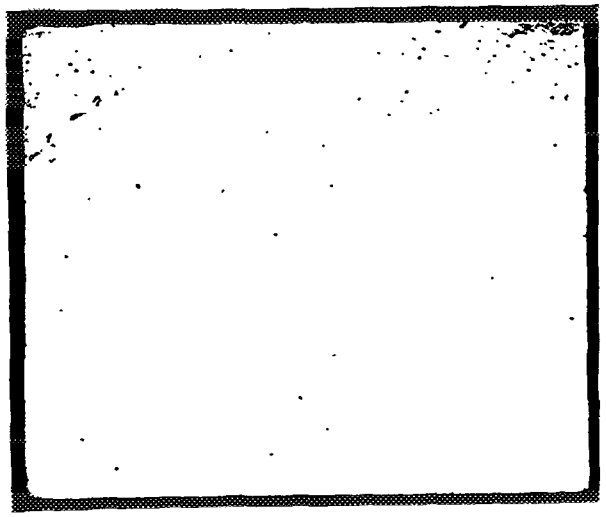

Fig. 1c: SBR $(U,Pu)O_{2.02}$ Ceramograph

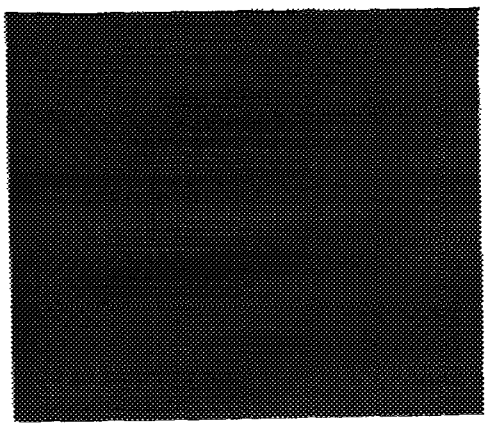

Fig. 2a: SBR $(U,Pu)O_2$ α-Autoradiograph

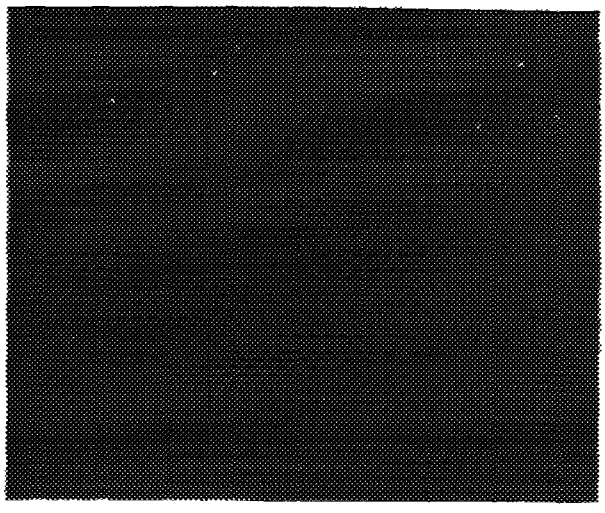

Fig. 2b: SBR $(U,Pu)O_{2.02}$ α-Autoradiograph

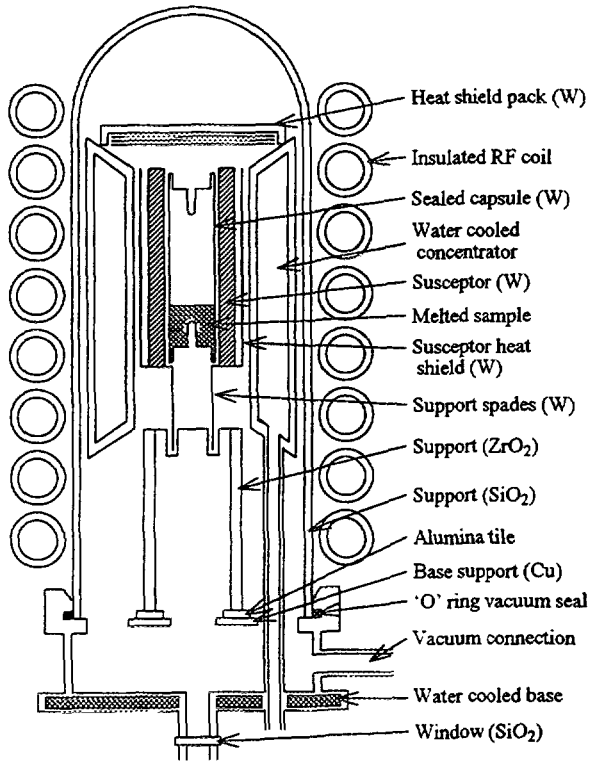

Fig. 3a: Melting Point Apparatus

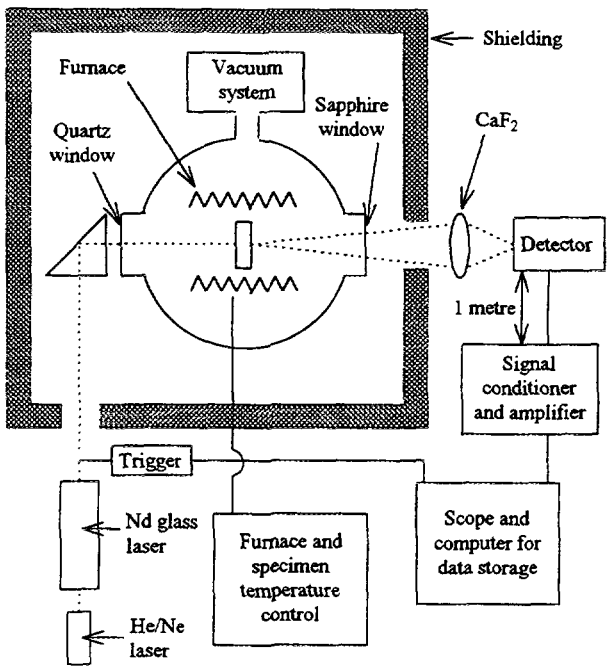

Fig. 3b: Thermal Diffusivity Apparatus

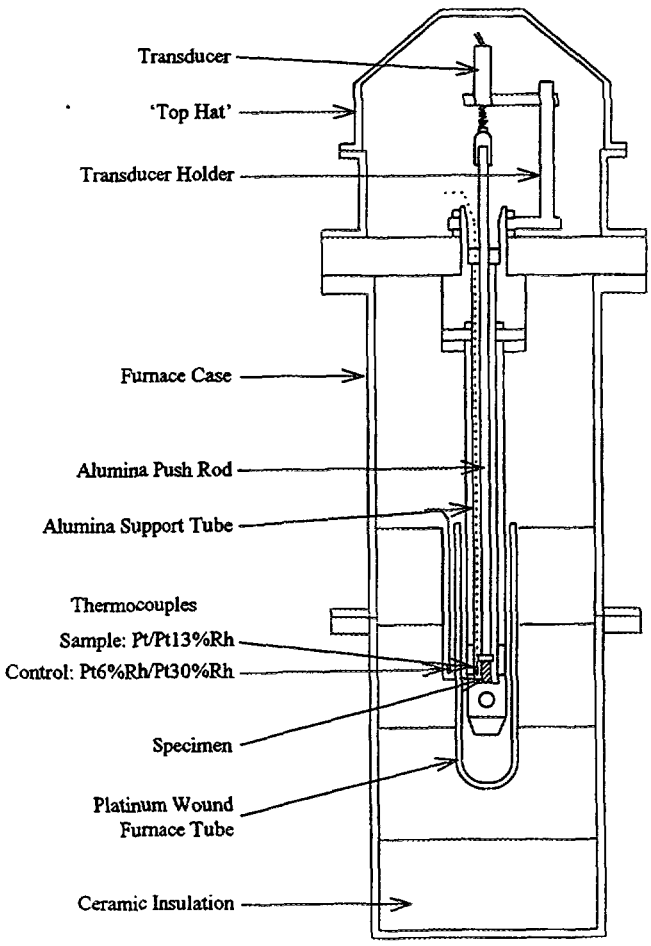

Fig. 3c: Thermal Expansion Apparatus

3. METHODOLOGY

The melting points of unirradiated stoichiometric UO_2 and MOX, and hyperstoichiometric MOX were measured using the thermal arrest method. In these experiments, the temperature of the fuel samples in a sealed tungsten capsule, as determined using an infra-red pyrometer, are measured as a function of time whilst the capsules are heated using an RF induction furnace. The apparatus is shown in more detail in Fig. 3a. The melting point of the sample is marked by a pause in the temperature rise which is the 'thermal arrest'. Calibration of the technique was made using pure molybdenum and tantalum samples.

The thermal diffusivity of unirradiated stoichiometric UO_2 and MOX, and hyperstoichiometric MOX was measured by the laser flash method. With this method, thin samples of the fuel were sealed in a vacuum and heated to the test temperature by a furnace. The time taken for a heat pulse, generated by a pulse from a laser, to travel through the sample is then measured in order to determine the diffusivity of the sample. The apparatus is shown in more detail in Fig. 3b. Measurements were made in the range 400 - 1600°C by cycling the temperature of the samples up and down in 200°C intervals and measuring the diffusivities on both legs of the temperature cycle. Each type of fuel was subject to one cycle up and down in temperature with the exception of the hyperstoichiometric fuel, where three additional samples were subjected to additional heating cycles with peak temperatures of 1000°C, 1400°C and 1600°C.

The thermal expansion measurements were performed on single pellets of unirradiated stoichiometric UO_2 and MOX, and hyperstoichiometric MOX. The pellets were heated by a furnace in an argon atmosphere and the expansion of the pellets in the vertical direction measured by a transducer via an alumina push rod. The apparatus is shown in more detail in Fig. 3c. The rig was calibrated using pure molybdenum pellets. Measurements were made of the thermal expansion of the fuel samples at temperatures in the range 150°C - 1230°C by cycling up to the peak temperature and down again twice for each sample.

4. THERMAL DIFFUSIVITY

The results of the thermal diffusivity measurements for two stoichiometric MOX samples are shown in Fig. 4 as a typical example of the measurements obtained. It can be seen that reproducibility

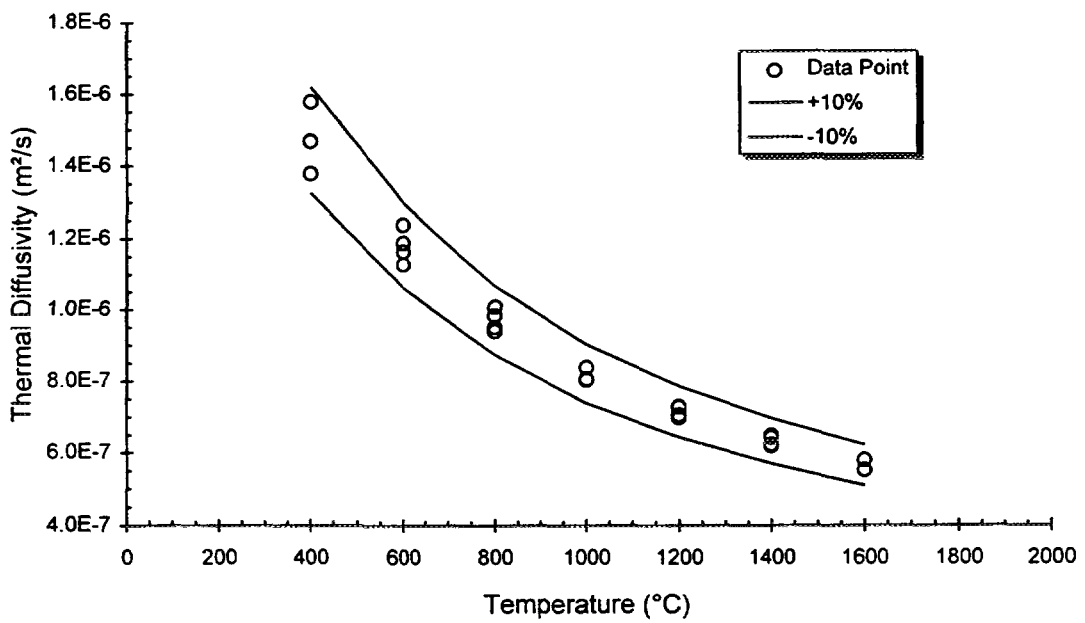

Fig. 4: Result of Thermal Diffusivity Measurements on Stoichiometric MOX Samples

of the data for multiple measurements at each temperature is good during both heating up and cooling down for these stoichiometric samples, as the dispersion of the thermal diffusivities at each temperature is less than 10%. From this result, it can be confirmed that changes in the specimen characteristics, such as stoichiometry and microstructure, during the measurements must be small.

The thermal conductivity was derived from these results by using the following equation:

$$k = \alpha \cdot C_p \cdot \rho$$

where k is the thermal conductivity in W/m/K, α is the thermal diffusivity in m^2/s, C_p is the specific heat capacity in J/kg/K, and ρ is the density in kg/m^3. The specific heat capacity was derived using the equation recommended in MATPRO Version 11[5]:

$$C_p = \frac{K_1 \cdot \theta^2 \cdot \exp\left(\frac{\theta}{T}\right)}{T^2 \cdot \left\{\exp\left(\frac{\theta}{T}\right) - 1\right\}^2} + K_2 T + \frac{(O/M)}{2} \cdot \frac{K_3 E_D}{RT^2} \cdot \exp\left(\frac{-E_D}{RT}\right)$$

where C_p is the specific heat capacity in J/kg/K, T is the temperature in K, O/M is the oxygen to metal ratio, R is the gas constant (8.3143 J/mol/K), θ is the Einstein temperature in K, and K_1, K_2, K_3, and E_D are constants.

The specific heat capacity of the MOX samples was calculated from those of pure UO_2 and PuO_2 assuming that the contribution from each constituent was in proportion to its weight fraction.

The theoretical density of the samples used in this study was around 95%, although there were small differences between the samples. The thermal conductivity of each sample was corrected for a density of 95% using the Maxwell-Euken's equation:

$$k = \frac{\rho}{100 + \beta(100 - \rho)} \cdot \frac{100 + 0.5(100 - 95)}{95} \cdot k_{95}$$

where k is the thermal conductivity of a test sample with a density of ρ% TD, β is a constant and k_{95} is the thermal conductivity of a sample with a density of 95% TD.

Fig. 5 shows the thermal conductivity of UO_2 derived using this method from the thermal diffusivity measurements. This figure also shows the thermal conductivities recommended by Martin [1], Washington [2], Brandt et al. [3] and Kosaka [4]. The data presented here can be seen to agree with these recommendations, and to have the same temperature dependence in the range used in the tests (400 - 1600°C). This confirms the validity of the measuring technique and the reliability of the data.

Fig. 6 shows the thermal conductivity of stoichiometric MOX along with the recommendations due to Martin [1] and MATPRO version 11 [5]. The data presented here can be seen to fall between these two recommendations.

In Fig. 7, the result of Gibby's study [6] on the effect of PuO_2 content on the thermal conductivity of $(U,Pu)O_2$ solid solutions is shown along with the data from this study. This data shows that the thermal conductivity of the stoichiometric MOX sample (9.9 wt% Pu/U+Pu) was approximately 10% less than that of UO_2 in the range 400 - 1600°C. The same dependence of thermal conductivity on Pu content is observed in Gibby's data, and while the absolute values of the data in this study are a little smaller than Gibby's data at the lower temperatures (400 - 600°C), it agrees very well with Gibby's data at the higher temperatures (800°C - 1200°C).

In the case of the thermal diffusivity measurements on the hyperstoichiometric MOX, a slight increase in the diffusivity was observed in the cooling cycle relative to the heating cycle, although the diffusivity in the cooling cycle agree reasonably well with the data for stoichiometric MOX. It is believed that the heating cycle up to high temperature moved the O/M ratio closer to 2.00. Based on several measurements for hyperstoichiometric samples with different peak temperatures, it was show that the change in O/M ratio was negligible under 1200°C, and hence, only the data for the heating cycle below 1200°C is used in this evaluation. In Fig. 8, the thermal conductivity of the hyperstoichiometric MOX is compared with the results for the stoichiometric MOX, along with the recommendation for stoichiometric MOX due to Martin [1]. It can be seen that the thermal conductivity of the hyperstoichiometric sample was smaller than that of the stoichiometric sample by about 10% in the range 400 - 1200°C.

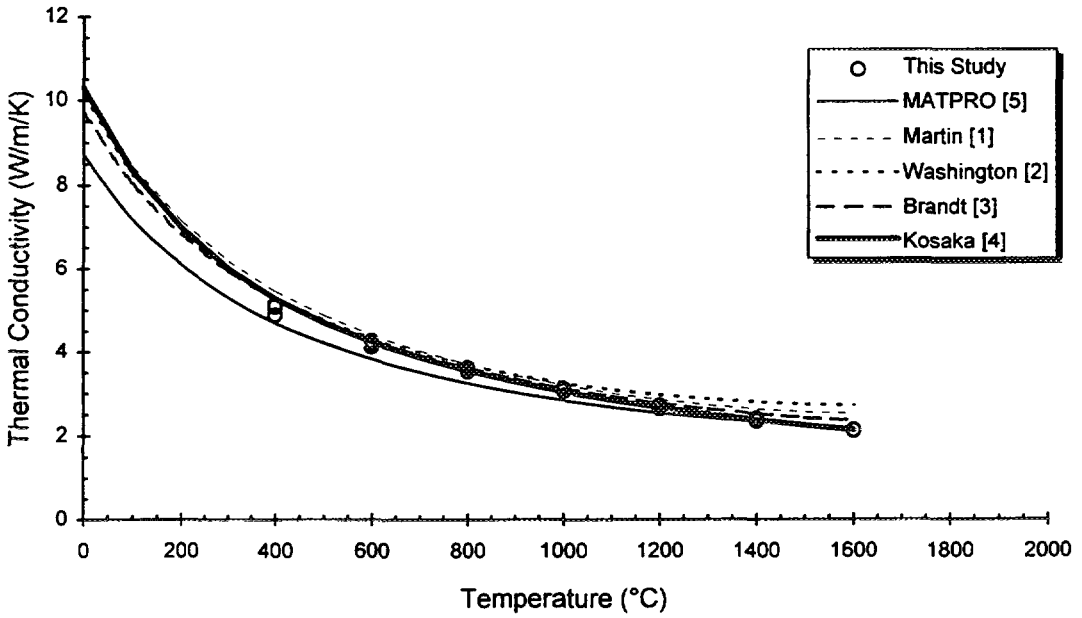

Fig. 5: Temperature Dependence of Thermal Conductivity of UO_2

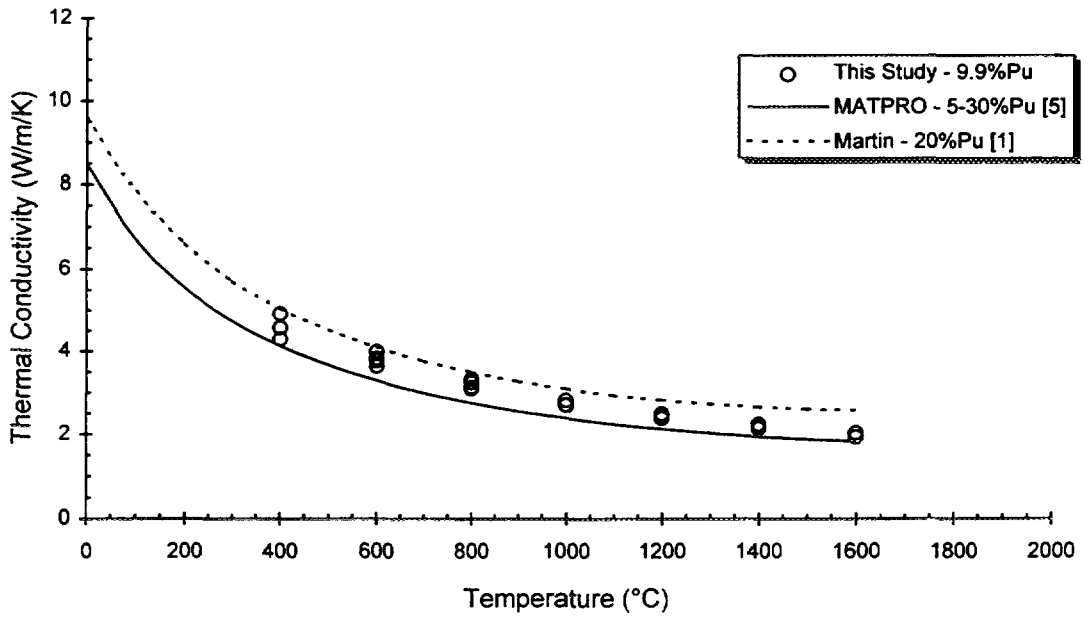

Fig. 6: Thermal Conductivity of Stoichiometric MOX

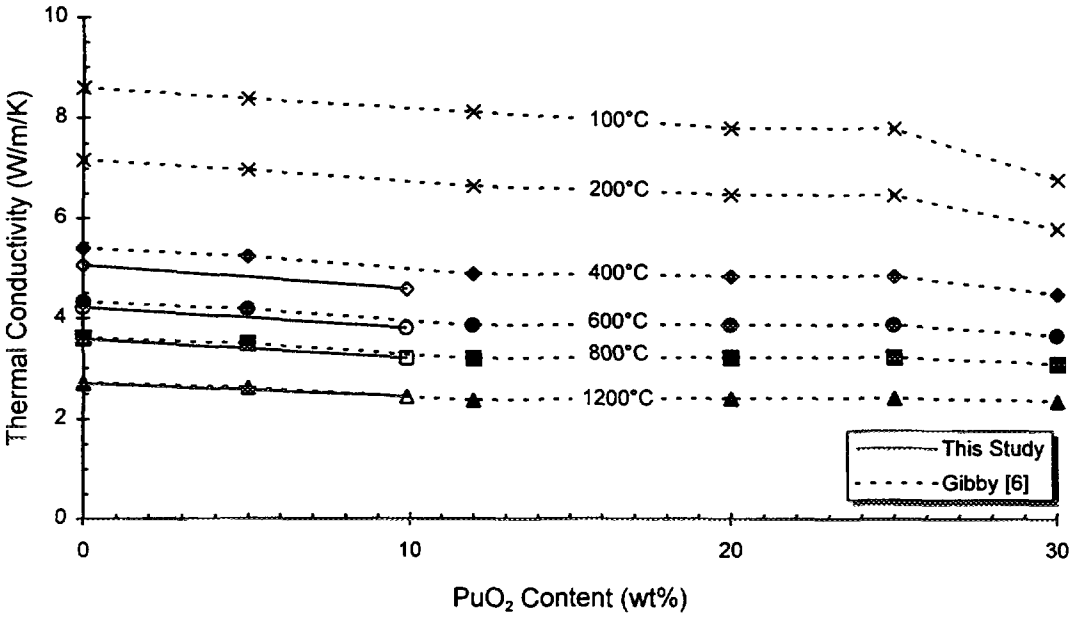

Fig. 7: Thermal Conductivity of MOX as a function of PuO_2 Content

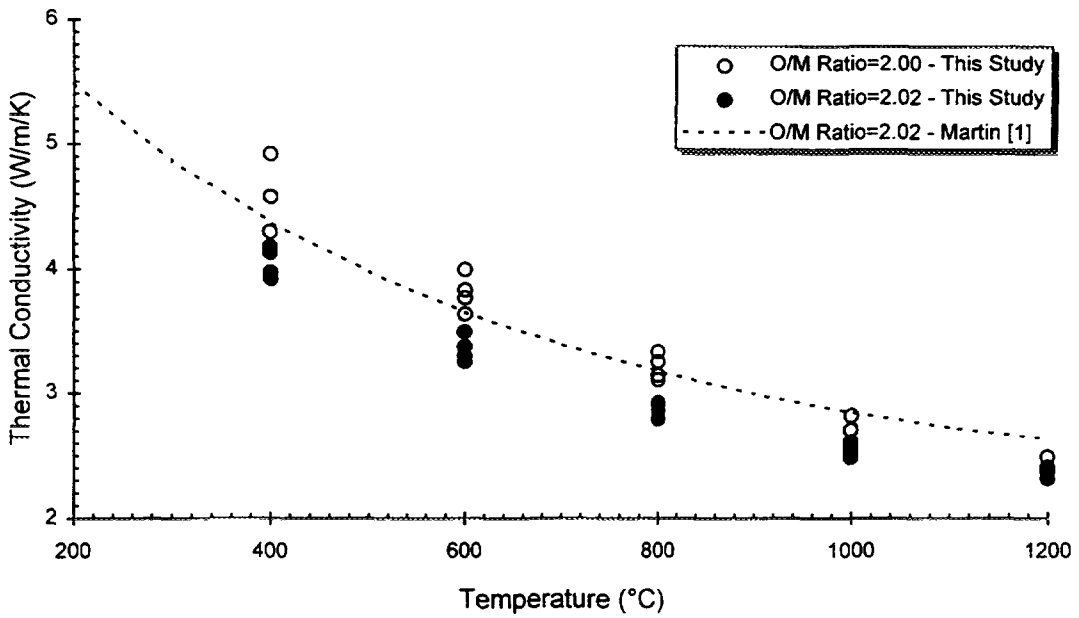

Fig. 8: Thermal Conductivity of Stoichiometric and Hyperstoichiometric MOX

5. MELTING POINT

In this study, the average melting point of the UO_2 sample was found to be 2849°C, which agrees well with the value 2840°C for unirradiated UO_2 which is given by MATPRO version 11 [5] referring to Brassfield et al. [7] and Lyon et al. [8]. Hence, the data reported here is in good agreement with their results, which gives confidence in the accuracy of the measurement technique.

In Fig. 9, the liquidus and solidus temperatures of the stoichiometric MOX samples are shown as a function of the Pu content along with those measured by other investigators, namely Adamson [9], Lyon et al. [10] and Aitken et al. [11]. While the solidus temperature of the stoichiometric MOX was higher than that recommended by Adamson [9], the solidus temperature was found to be consistent with the solidus recommended by Adamson.

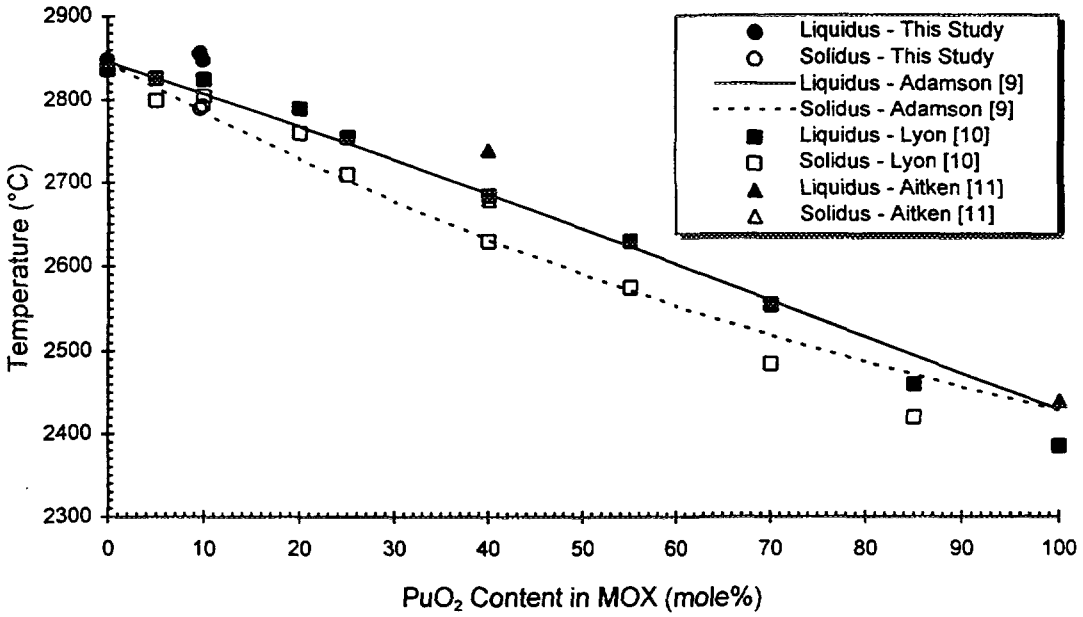

Fig. 9: Liquidus and Solidus Temperatures of MOX

As can be seen in Fig. 9, there was no obvious difference between the average liquidus and solidus temperatures of stoichiometric SBR MOX and those of MIMAS MOX. From these results, it would appear that the MOX pellet fabrication process does not affect the melting point of the product.

The average solidus temperatures of the stoichiometric and hyperstoichiometric (O/M = 2.02) MOX samples was lower than that of the stoichiometric MOX samples by about 70°C. This implies that the degradation of the solidus due to a deviation of the O/M ratio of 0.02 from stoichiometry is around 70°C.

6. THERMAL EXPANSION

The thermal expansion coefficient at a particular temperature (θ°C) was normalised to zero at 200°C according to the following formula:

$$\frac{\Delta L_\theta}{L_{25}} = \frac{L_\theta - L_{200}}{L_{25}} \times 100$$

where $\Delta L_\theta / L_{25}$ is the normalised thermal expansion coefficient at θ°C, L_θ is the sample length at θ°C, L_{200} is the sample length at 200°C and L_{25} is the sample length at 25°C.

Fig. 10 shows the temperature dependence of the thermal expansion coefficients of UO_2 and stoichiometric MOX with the recommended values for UO_2 due to Martin [12] and MATPRO version 11 [5]. It can be seen that the averaged thermal expansion of the stoichiometric MOX sample is almost equal to that of the UO_2 samples, and in good agreement with the recommendations of Martin [12] and MATPRO version 11 [5] at temperatures up to 1200°C.

The thermal expansion of the hyperstoichiometric MOX samples is compared with that of the stoichiometric MOX samples in Fig. 11. This shows that there is virtually no observable difference in the thermal expansion resulting from a deviation in stoichiometry of 0.02.

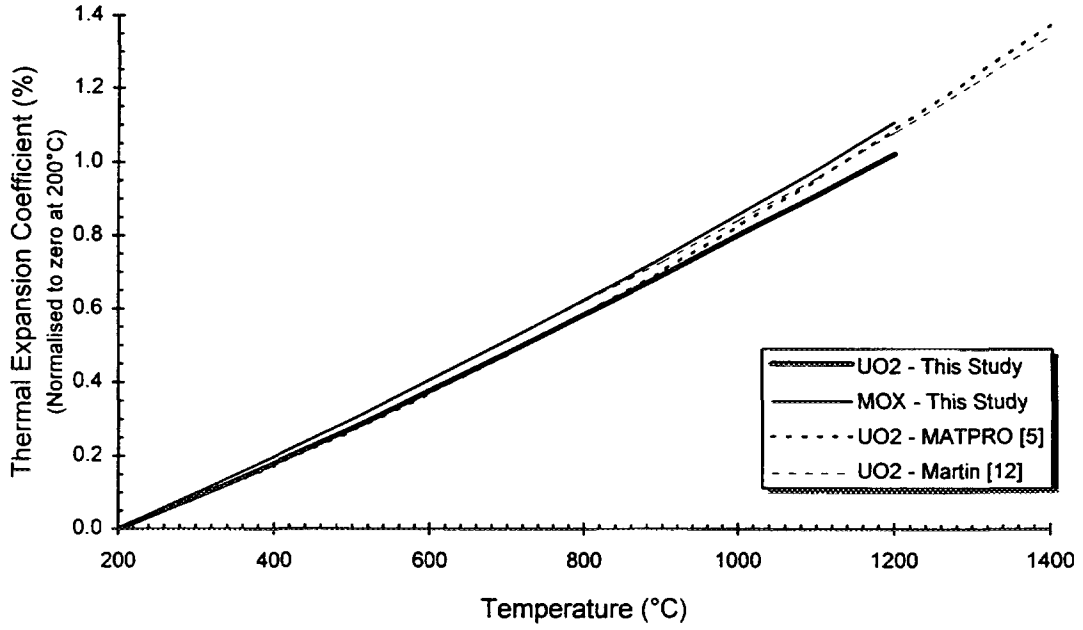

Fig. 10: Thermal Expansion of Stoichiometric UO_2 and MOX

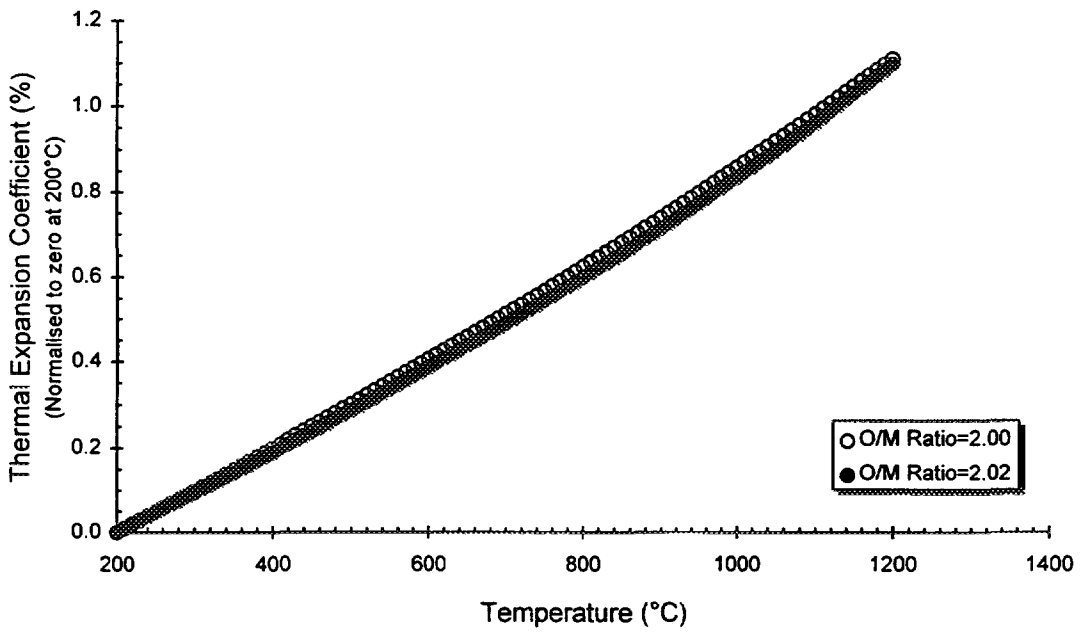

Fig. 11: Thermal Expansion of Stoichiometric and Hyperstoichiometric MOX

7. CONCLUSIONS

In this study, the thermal diffusivity, melting point and thermal expansion of stoichiometric UO_2, stoichiometric and hyperstoichiometric SBR MOX (9.9 wt% Pu/U+Pu), and the melting point of stoichiometric MIMAS MOX (9.6 wt% Pu/U+Pu) have been measured.

The following conclusions can be drawn from this study:

a) The thermal conductivity of stoichiometric UO_2 and MOX is confirmed to be in good agreement with previous published data, and that of MOX was approximately 10% less than that of UO_2 at temperatures between 400 and 1600°C.

b) The thermal conductivity of hyperstoichiometric MOX (O/M ratio = 2.02) was smaller than that of stoichiometric MOX by about 10% in the range 400 - 1600°C.

c) There were virtually no differences in the observed thermal expansions of stoichiometric UO_2, stoichiometric and hyperstoichiometric MOX in the temperature range 150 - 1200°C.

d) The melting point of stoichiometric UO_2 and MOX was in good agreement with those of other investigators.

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NEUTRONIC FEASIBILITY OF PWR CORE WITH MIXED OXIDE FUELS IN THE REPUBLIC OF KOREA

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Abstract

Neutronic feasibility of a PWR core with mixed oxide (MOX) fuels has been investigated as part of the feasibility study for recycling spent fuels in Korea. A typical 3-loop PWR with 900 MWe capacity is selected as reference plant to develop equilibrium core designs with low-leakage fuel management scheme, while incorporating various MOX loading. The fuel management analyses and limited safety analyses show that, safely stated, MOX recycling with 1/3 reload fraction can be accommodated for both annual and 18 month fuel cycle schemes in Korean PWRs, without major design modifications on the reactor systems.

1. INTRODUCTION

Nuclear generated electricity plays vital role in Korea by accounting for 40 % of total electric power generation in 1993, and this trend will continue in the years to come. We have firm plan of building 14 more units (10 PWRs and 4 PHWRs) by the year 2006 besides 9 operating units (8 PWRs and 1 PHWR) as of 1994. These nuclear units will result in an accumulation of more than 7,000 tHM of spent nuclear fuels in Korea by the year 2006. The lack of proper store place for and the rapidly increasing amount of spent nuclear fuels have made their management one of the national issues. Possible reuse of spent nuclear fuels could mitigate spent fuel storage problem, and contribute to the recycling of resources and the protection of natural environment through the reduction of radioactive wastes. In this regards, two types of spent fuel recycling scheme could be considered in Korea; recycling of spent fuels into PWRs and PHWRs. The technology development of the Direct Use of spent PWR fuel In CANDU (DUPIC) has been under progress since 1992. The neutronic feasibility of a PWR core with mixed oxide (MOX) fuels has been investigated as part of the feasibility study for recycling spent fuels. This paper describes preliminary results of the work.

Although many countries such as France [1-4], Germany [5-8], Belgium [1], and Japan [9-10] have continuously developed and matured the technologies to recycle the plutonium as MOX in thermal reactors, there exist many technical questions to be answered on our part because we do lack technical experience and our PWRs are going toward 18 month cycle operation. For the MOX feasibility study, a typical 3-loop PWR is selected as reference plant to develop equilibrium core design for which MOX fuels are assumed to be partly loaded.

This study has comprised of several stages. The first step was to design the MOX fuel assembly; determination of the equivalent plutonium content in MOX fuel using both simple graphic method and multicycle scoping analysis, and optimal allocation of MOX fuel rods to flatten the peak rod power. The second step was to develop equilibrium core designs with low-leakage fuel loading strategy, and analyze the changes in neutronic characteristics. Since Korean PWRs adopt 18 month cycle, both 12 and 18 months cycle schemes are covered, while incorporating three scenarios of up to 40 % MOX loading fraction. Finally preliminary safety analyses have been performed for several typical reactivity-related accidents such as rod ejection accidents and steam line break accident.

2. OPTIONS FOR RECYCLING SPENT FUELS IN KOREA

The Korean nuclear program is one of the largest in the world. The nuclear installed capacity has been continuously enlarged since 1978 when Korea witnessed its first nuclear power generation. As of

1994, 9 nuclear power plants including one pressurized heavy water reactor (PHWR : CANDU) are in commercial operation, accounting for nearly 40 % of total electricity generation. According to the long-term plan, 14 more units (10 PWRs and 4 PHWRs) will be on line by the year 2006. Table I shows the summary of current status and plan for nuclear power plants in Korea

The continuous expansion and development of nuclear power program increase the cumulative amount of spent fuels and therefore plutonium discharged from nuclear reactors. The quantities of heavy metal in spent fuel unloaded from a PWR(900MWe) and a PHWR(700MWe) are estimated to be about 20 tones and 90 tones annually. Therefore more than 7,000 tHM of spent nuclear fuels (4,000 tHM from 18 PWRs and 3,000 tHM from 5 PHWRs) are expected to be accumulated from 23 nuclear units in Korea by the year 2006 as shown in Fig. 1. This amount of spent nuclear fuels roughly translates into 50 tones of plutonium which represent significant amount of semi-domestic energy resource, and thus cannot simply be ignored.

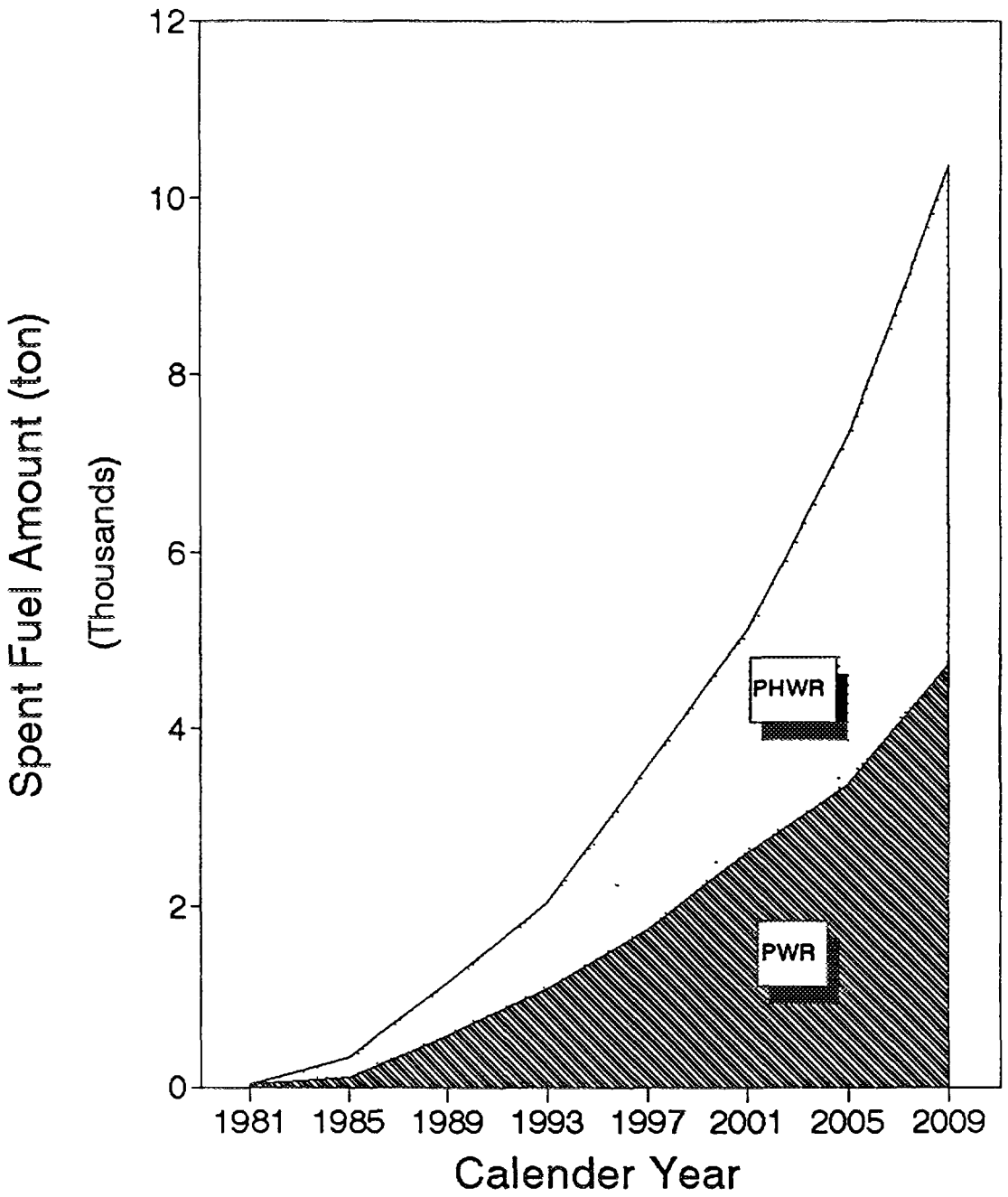


Fig. 1. Estimation of Cumulative Spent Fuels Discharged from Korean PWRs

The lack of proper store place for and the increasing amount of spent nuclear fuels have made their management one of the national issues. Recycling of spent fuels can provide Korea with many attractive benefits; it will help to mitigate spent fuel storage problem, and contribute to the recycling of resources and the protection of natural environment through the reduction of radioactive wastes. In this regards, two types of recycling could be considered in Korea; recycling of spent fuels into PWRs and PHWRs. Taking into account the very specific situation of Korea, the technology development of the Direct Use of spent PWR fuel In CANDU (DUPIC) has been under progress since 1992. Since the use of MOX in LWRs is well established technology which have been chosen by many European and Japanese utilities, it can be implemented without significant R&D effort. Therefore, neutronic feasibility of a PWR core with mixed oxide (MOX) fuels has been investigated as part of the feasibility study for recycling spent fuels.

3. MOX FUEL ASSEMBLY DESIGN

3.1. Equivalent Plutonium Content

It is well known that neutronic characteristics of plutonium isotopes are quite different from those of uranium isotopes. Due to slower reactivity change of MOX as function of burnup, it is necessary to determine the plutonium content of MOX fuel equivalent to the UO_2 fuel. The concept of equivalence adopted in our study states that both fuels should provide the same cycle length for equilibrium cores. Thus the equivalent plutonium content could be varied with different fuel cycle operations. We assumed that the reference uranium cores are in operation with annual cycle strategy with 3.5 w/o U^{235} enriched fuel, and 18 months cycle strategy with 4.0 w/o U^{235} enriched fuel.

According to the linear reactivity model, the equilibrium cycle length of the core loaded with the constant enriched fuel is given by [11]

$$B_c = \frac{\rho^0}{A} \cdot \left(\frac{2}{n+1} \right) \tag{1}$$

where B_c = equilibrium cycle length,
ρ^0 = initial reactivity of feed fuel,
A = rate of reactivity change per unit of burnup,
n = number of regions in the core.

Keeping the same number of regions in the core for both UO_2 and MOX fuels, the following is the relation to result in the same equilibrium cycle length for UO_2 and MOX cores regardless of MOX loading fractions:

$$\frac{\rho^0_{UO_2}}{A_{UO_2}} = \frac{\rho^0_{MOX}}{A_{MOX}} \tag{2}$$

where $\rho^0_{UO_2}$ and ρ^0_{MOX} = initial reactivities of uranium and MOX fuel,

A_{UO_2} and A_{MOX} = rates of reactivity change per unit burnup of uranium and MOX fuel.

$\rho^0_{UO_2}/A_{UO_2} (= \rho^0_{MOX}/A_{MOX})$ in Eq.(2) means the fuel burnup at zero reactivity. Using this

simple relationship, we can find the equivalent MOX fuel to UO_2 fuel by identifying the MOX reactivity curve crossing the point $(0, \rho^0_{UO_2}/A_{UO_2})$ in the reactivity-burnup plot precalculated for selected plutonium contents, which is called the graphic method. The equivalent plutonium contents of MOX fuel for 3.5w/o and 4.0w/o U^{235} enriched uranium fuels turned out to be 3.1w/o of plutonium fissile mixed with natural uranium and 4.0w/o of plutonium fissile with depleted uranium respectively.

The validity of the graphic method was confirmed through multicycle scoping calculations with FLOSA code[12]. It is shown that Eq.(2) holds for wide range of fuel management parameters such as reload batch size, fraction of MOX loading, and the number of MOX assemblies on the core periphery. Fig. 2 shows the change of equivalent plutonium contents for different fraction of MOX loading.

3.2. Optimal Zoning in MOX Fuel Assembly

In case MOX fuel assemblies are partly loaded in the core, it is important to control the power peaking of the peripheral rod within MOX fuel assembly. The peripheral rods are strongly affected by neighboring UO_2 fuel assemblies. Therefore zoning in MOX fuel assembly is necessary in order to flatten the power distribution over MOX fuel assembly. We developed optimal zoning with MOX rods having three different plutonium contents as shown in Fig. 3.

4. FUEL MANAGEMENT STUDIES FOR MOX PART-LOADED CORES

4.1. Fuel Management

For this study, a typical 3-loop PWR with 900 MWe capacity is selected as the reference plant to develop equilibrium core design with low-leakage fuel loading scheme. Since 900 MWe class PWRs adopt

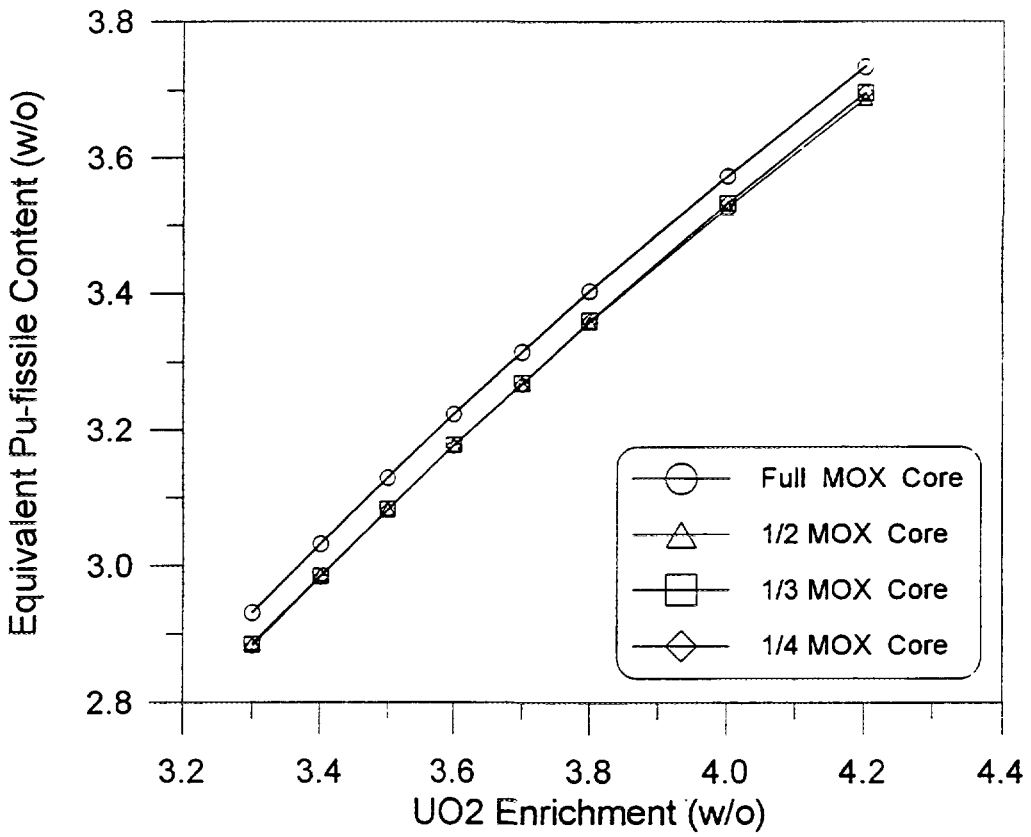


Fig. 2. Equivalent Plutonium Content vs. UO_2 Fuel Enrichment

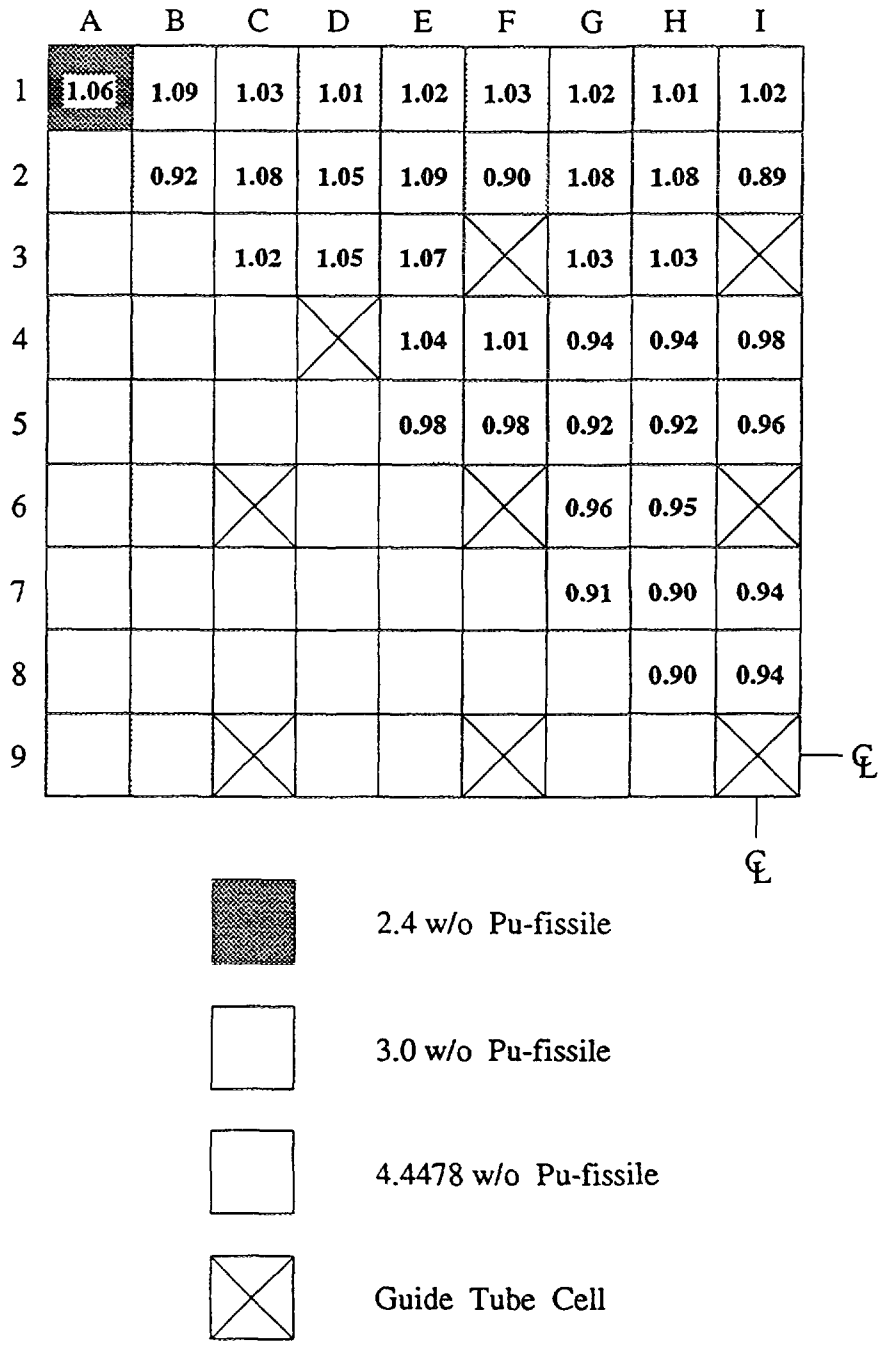
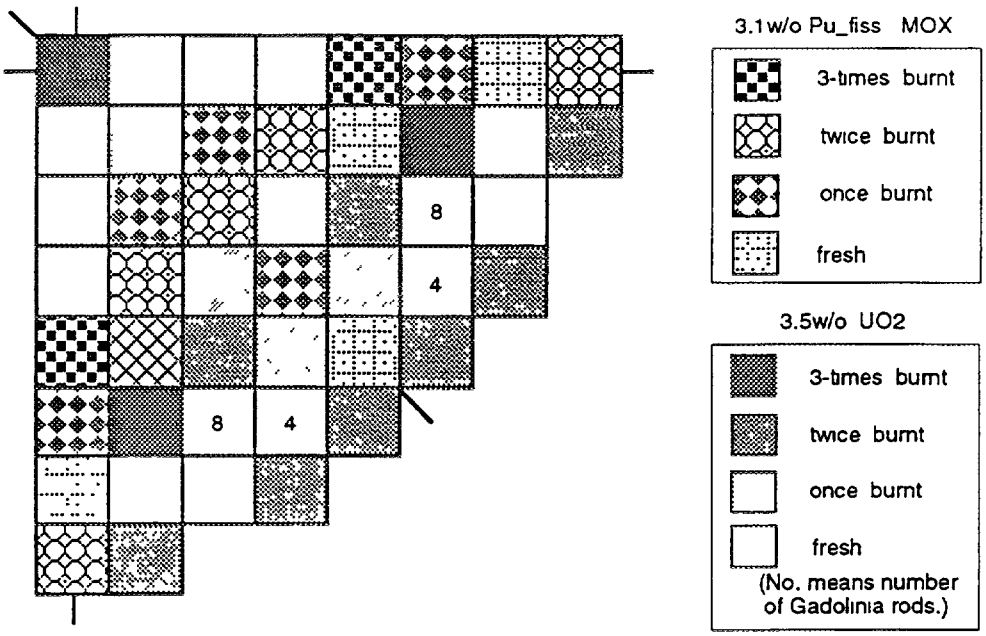


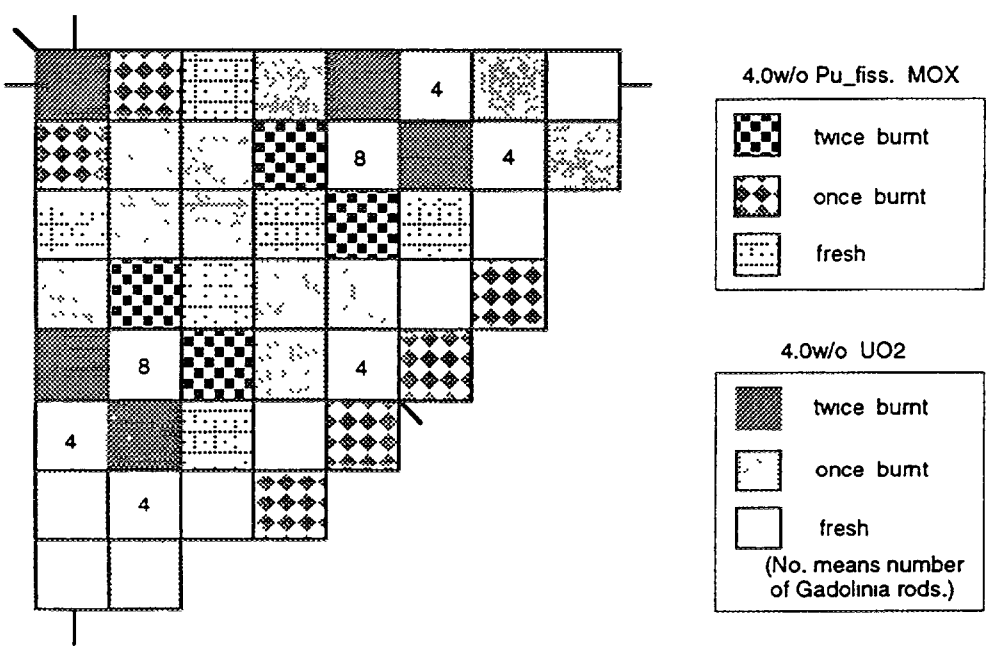
Fig. 3. Zoning and Relative Power Distribution of MOX Assembly

18 month cycle, both annual and 18 month cycle schemes are covered, in which MOX fuels are assumed to be partly loaded. We considered three scenarios of up to 40 % MOX loading in the annual operating cycle in contrast to only 1/3 MOX fraction in 18 month cycle. Loading patterns of MOX equilibrium cores for both cycle schemes are shown in Fig. 4. The low-leakage loading strategy made most of fresh fuel assemblies occupy inboard locations, and some fresh UO_2 assemblies require gadolinium burnable poisons for controlling peak pin power.

The fuel cycle characteristics for MOX and UO_2 cores are summarized in Table II. The MOX cores represent 1/3 MOX loading for both annual and 18 month cycles. The cycle lengths of annual and 18 months MOX cores are 12.0 and 16.53 MWD/MT which are very close to those of UO_2 cores, 12.22 and 16.60 MWD/MT. This clearly demonstrates again the validity of our method to determine the equivalent plutonium content of MOX fuel.



Annual Cycle



18 month Cycle

Fig. 4. Loading Patterns of MOX Equilibrium Cores for Annual and 18 Month Cycles

Table I. Status of Nuclear Power Plants in Korea

Plant Name	Reactor Type (Supplier)	Capacity (MWe)	Starting Date	Current Cycle at 95.7.1
Kori 1	PWR (W)	587	1978.4.29	14
Wolsung 1	PHWR (AECL)	678.8	1983.4.22	-
Kori 2	PWR (W)	650	1983.7.25	11
Kori 3	PWR (W)	950	1985.9.30	9
Kori 4	PWR (W)	950	1986.4.29	9
Yonggwang	PWR (W)	950	1986.8.25	9
Yonggwang 2	PWR (W)	950	1987.6.15	8
Ulchin 1	PWR (F)	950	1988.9.10	7
Ulchin 2	PWR (F)	950	1989.9.	6
Yonggwang 3	PWR (CE)	1000	1995.4.	1
Yonggwang 4	PWR (CE)	1000	1996.3.	-
Wolsung 2	PHWR (AECL)	700	1997.6.	-
Ulchin 3	PWR*	1000	1998.6.	-
New PHWR 1	PHWR	700	1998.6.	-
Ulchin 4	PWR*	1000	1999.6.	-
New PHWR 2	PHWR	700	1999.6.	-
New PWR 1	PWR*	1000	2001.6.	-
New PWR 2	PWR*	1000	2002.6.	-
New PWR 3	PWR*	1000	2003.6.	-
New PWR 4	PWR*	1000	2004.6.	-
New PWR 5	PWR*	1000	2005.6.	-
New PWR 6	PWR*	1000	2006.6.	-
New PHWR 3	PHWR	700	2006.6.	-

* : will be Korean Standard Type.

Table II. Fuel Cycle Characteristics of UO_2 and MOX Cores

Fuel Cycle Type	Annual Cycle		18 Month Cycle	
Core Characteristics	UO2 Core	MOX Core	UO2 Core	MOX Core
Number of Fuel Assemblies in a Core				
MOX Fuel Assembly	-	52	-	56
UO_2 Fuel Assembly	157	105	157	101
Number of Feed Fuel Assemblies				
MOX Fuel Assembly	-	16	-	20
UO_2 Fuel Assembly	48	32	64	44
Fuel Assembly Specification				
Fissile Pu Content in MOX (w/o)	-	3.1	-	4.0
U^{235} Enrichment in MOX (w/o)	-	0.71	-	0.225
U^{235} Enrichment in UO_2 (w/o)	3.5	3.5	4.0	4.0
Cycle Length (MWD/MtM)	12.22	12.00	16.60	16.53
Fuel Burnup (MWD/MtM)				
MOX Fuel Batch Average Burnup	-	39.74	-	45.53
MOX Assembly Maximum Burnup	-	41.65	-	47.23
UO_2 Fuel Batch Average Burnup	37.59	35.45	40.79	38.35
UO_2 Assembly Maximum Burnup	43.65	41.25	50.03	47.55

4.2. Neutronic Characteristics of MOX Core

The power distributions of MOX cores were not significantly different from those of uranium cores. The axial power distributions of MOX cores at HFP at several burnup stages, however, are slightly bottom skewed from those of uranium cores, which resulted from more negative moderator temperature coefficient in MOX core.

A major effect of MOX loading is the hardening of thermal neutron spectrum in the core which alters various core physics parameters, mainly reactivity-related ones. Some important core physics parameters are compared between 1/3 MOX and UO_2 cores in Table III. Because of hardened spectrum, the reactivity worth of soluble boron, control rod and xenon are proportionally reduced with increased MOX loading. The consequence of smaller soluble boron worth is reflected in the larger critical boron concentrations, which may require some changes in the boron systems. The MTC and ITC become more negative with MOX loading, while the Doppler coefficient is hardly influenced. In all cases. the differences between MOX and UO_2 cores are more apparent at beginning-of-cycle.

Since control rod worth is reduced, the shutdown margins of MOX cores are generally reduced. The magnitude of reduction, however, depends more on the loading pattern than the MOX fraction to some extent. The required minimum shutdown margin is 1.77% $\Delta k/k$. This limit is still maintained for all MOX cases.

4.3. Preliminary Safety Analysis

Preliminary safety analyses were performed for 1/3 MOX equilibrium core in the annual operating cycle. Considered are only those accidents which are most influenced by the change of core physics

Table III. Important Core Physics Parameters of UO_2 and MOX Cores

Fuel Cycle Type	Annual Cycle		18 Months Cycle	
Core Characteristics	UO_2 Core	MOX Core	UO_2 Core	MOX Core
Boron Concentration				
Refueling CB,ARI(k < 0.95)	>1571	>2187	>2066	>2674
Shutdown (k = 0.98) with ARI, HZP	862	971	1313	1372
Shutdown (k = 0.98) with ARI, HFP	1878	2131	2374	2564
To control at HZP, ARO, (k = 1.0)	1638	1819	2100	2213
To control at HZP, ARI, (k = 1.0)	640	680	1056	1010
To control at HFP, ARO, (k = 1.0)				
0 MWD/MtM, No Xenon	1401	1519	1907	1908
240 MWD/MtM, Equilibrium Xenon	1072	1155	1541	1520
Moderator Temperature Coefficient at HFP (pcm/°C)				
BOC / EOC	-18/-54	-28/-59	-10/-56	-23/-60
Isothermal Temperature Coefficient at HZP, BOC (pcm/°C)	-1.46	-15.65	1.54	-11.45
Doppler Temperature Coefficient at near EOC (pcm/°C)	-3.93	-4.04	-3.96	-4.06
Boron Worth at HFP (pcm/ppm)				
BOC / EOC	-8.2/-9.5	-6.5/-7.8	-7.2/-8.8	-5.7/-7.0
Xenon Worth (pcm)				
BOC / EOC	2839/2895	2519/2674	2721/2848	2338/2545
Total Control Rod Worth (pcm)				
BOC / EOC	8660/8920	7421/8170	8004/8350	7262/7690
Shutdown Margin (%Δρ)				
BOC / EOC	4.54/3.55	3.45/2.97	4.26/2.85	3.47/2.63

characteristics induced by MOX loading; control rod ejection accidents at zero power and full power, and steamline break accident (or the most limiting cooldown event). Control rod ejection analyses at zero power and full power initial conditions have shown acceptable consequences because the smaller reactivity of the ejected rod compensate the adverse effect of the effective delayed neutron fraction and prompt neutron lifetime. Steamline break accident analysis also resulted in favorable consequences.

5. CONCLUSIONS

The continuous expansion and development of nuclear power program in Korea increase the cumulative amount of spent fuels discharged from nuclear reactors. Recycling of spent fuels can provide Korea with many attractive benefits; it will help to mitigate spent fuel storage problem, and contribute to the recycling of resources and the protection of natural environment through the reduction of radioactive wastes. As part of the feasibility study for recycling spent fuels, neutronic feasibility of a PWR core with mixed oxide (MOX) fuels has been investigated. The fuel management study and restricted safety analyses show that, safely stated, MOX recycling with 1/3 reload fraction can be accomodated for both annual and 18 month fuel cycle operations in Korean PWRs, without major design modifications on the reactor systems. Therefore, if internationally agreed, the real feasibility of recycling plutonium as MOX fuel can be demonstrated by trial loading of very small number of MOX assemblies in a commercial PWR.

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SESSION 4
PLUTONIUM DISPOSAL

COULD WEAPON-GRADE PLUTONIUM BE AN ASSET FOR MANAGING PU INVENTORIES?

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Abstract

Due to the temporary shortage of MOX fuel fabrication facilities, the stockpile of separated civilian grade Pu (CPu) is predicted to increase up to the turn of the century. An additional quantity of weapon grade Pu (WPu) will be progressively isolated at the same period. Both CPu and WPu surplusses require disposition as soon as feasible.

Although non-proliferation concerns, established national policies, public acceptance problems and other considerations largely complicate the aspect of the use of WPu, it is worth examining the advantages which could result from a synergetic management of :

- LWR grade Pu to which AGR grade Pu might be associated
- WPu
- GCR grade Pu which should be considered as a Pu variety situated between the two first ones as far as their physical and neutronic characteristics are concerned.

Two scenarios of integrated managements of the CPu varieties and WPu are being considered. They indicate several technical and economical advantages but also important problems to be resolved, mainly from the non-proliferation point of view. In that respect, it is concluded that, although no reasonable perspective exists to resolve these problems easily (or at all), the advantages justify an effort of the international community to consider how it could be implemented.

1. INTRODUCTION

Disposition of separated Pu has attracted wide attention from political authorities, mainly in the perspective of non-proliferation and safeguard concerns. A lot of alternatives are being considered :

- utilization as MOX(1) fuel in LWRs, FNRs and ATRs using commercially established technologies;
- utilization in CANDUs and HTRs, being studied in conceptual projects based on earlier R & D results;
- unconventional options based either on existing power plants (LWRs, FNRs and CANDUs), on power plants developped earlier (HTRs and MSRs) or on new concepts based on accelerator-driven sub-critical reactors;
- ultimate disposal after conditioning for insolubilisation and diversion resistance.

(1) : A list of abbreviations and acronyms is given in the Appendix

Emphasis in this paper will be on the first alternative, since it leads to the earliest possible disposition of separated Pu inventories. Indeed, MOX is being or has been utilized, at commercial scales, in 6 of the 30 countries operating nuclear reactors, representing 41% of the installed nuclear capacity in the world. Additionally 7 other countries, totalling a further 44% of the installed capacity, have conducted demonstration irradiations of MOX fuel.

More details on the assumptions and considerations underlying this presentation are contained in [1].

2. COMPARISON OF Pu UTILIZATION ALTERNATIVES

Table I provides the disposition capacity over the next 10 and 20 years using each reactor type, if all the reactors of that type were loaded with a maximum of Pu fuel, for two installed capacity assumptions :

- the reactors connected to the grid and under construction;
- the same plus 10 GWe;

Table I. Pu UTILIZATION CAPACITY (t Put) OF EACH POWER REACTOR TYPE

Installed generating capacity considered	current + under construction		same + 10 GWe	
Period	10 yr	20 yr	10 yr	20 yr
LWR				
current MOX (33%)	3300-4400	9000-10000	3300-4400	9000-10000
100% MOX core	3300-4400	17000-20000	3300-3400	17000-20000
alternative Pu fuel	0	0-7000	0	0-7000
CANDU				
MOX fuel	28-80	170-230	70-200	400-540
alternative Pu fuel	0	0-410	0	0-580
FNR	14-43	90-120	50-150	300-400
ATR	2	13	2	13-110
HTR	-	-	0	negl

The ranges reflect the ranges assumed for disposition rate in each system and for lead times before industrial utilization.

It shows that MOX utilization in LWRs is the most expeditious solution to eliminate Pu stockpiles, as already concluded in [2]. The calculations indicate that, even if only 10% of the LWRs were loaded with MOX fuel, as currently applied (i.e. MOX fuel constituting one third of each reload), an inventory of 200 t Pu could be utilized within 6 to 8 years and an inventory of 400 t Pu within 9 to 11 yrs. It justifies to consider only the MOX in LWRs alternative in this paper of limited scope.

This does not prejudice recognition that alternative systems have intrinsic advantages and that, Pu not being a fungible commodity, national context may influence the choice of the Pu disposition scenario(s).

3. Pu INVENTORY

3.1. Status at end 1994

As a result of studies conducted by FEX [3] and of data published by Japan AEC [4], the separated CPu inventory at end 1994 can reasonably be evaluated at :

- 120 t Put stored as raw material, in good agreement with the 112 t Put predicted [5] by a model developped by the IAEA [6], taking into account the simplifying assumption adopted in both assessments;

- 12 t Put stored as MOX and Pu bearing experimental material.

3.2. Future arisings

When THORP will reach steady state operation, a total of 29 t Put will be separated per year. When Rokkasho-mura will be in operation (i.e. after 2004), the arisings will go up to 38 t Put per year.

To these quantities of CPu should be added the WPu which will be available as a result of the START agreements and can be considered now as stored in the form of low-alloyed Pu metal. In general, publications indicate rounded figures of 100 t Pu from the Russian arsenal and 50 t Pu from the American arsenal which should be separated from warheads within the START commitments.

The so calculated arisings (Fig.1) should be considered upper boundary, since it is based on the current La Hague, Sellafield, Tokai-mura and Mayak reprocessing plants operating at rated capacity and on the future Rokkasho-mura and Krasnoyarsk reprocessing plants to be on schedule. Given the cancellation of post-baseload committments by German utilities, the schedule shifts observed in startup of new reprocessing plants and the reduced capacity factor usually experienced during the first years of operation of novel plants, the actual Pu arisings might be significantly lower.

3.3. Availability of recipient reactors

Although the quantities look large, Table I indicates that disposition of 750 t Put over 15 years can be achieved by loading

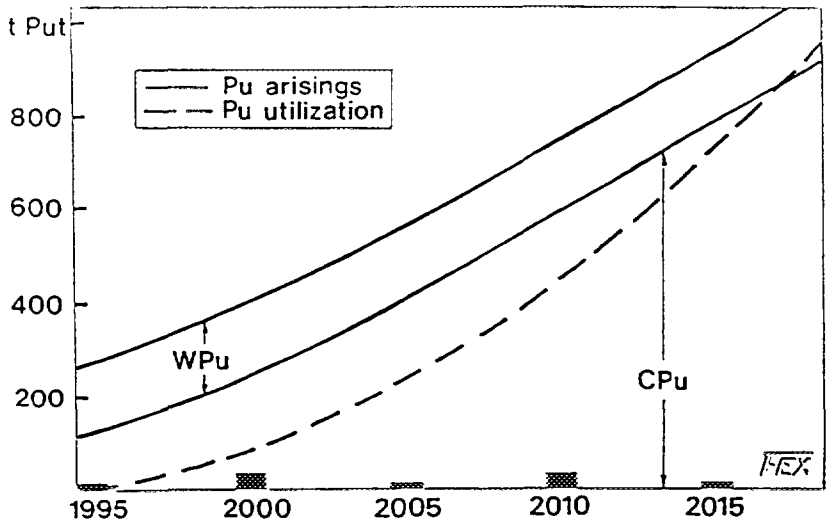

Fig. 1. Cumulated Pu arisings and utilization since end 1994

MOX fuel in all existing and today foreseen FNRs and ATRs and 7 to 10% (23 to 32 GWe) of the LWRs (1/3 MOX reloads). Since 43% of the LWR capacity is installed in countries possessing commercial experience of MOX fuel utilization, only 6 to 23% of the LWRs in those countries would need to be loaded with MOX fuel to reduce the stockpile to zero by 2010. Currently, there are 34 reactors licensed to use MOX fuel and 25 others in the licensing process, representing a total of 54 GWe. Furthermore, other reactors, such as CANDUs, and advanced systems, such as 100% MOX cores in LWRs, offer interesting perspectives for Pu utilization and will probably also contribute to Pu disposition, thereby reducing the number of LWRs to be loaded with MOX fuel.

It can be concluded that availability of reactors is, in principle, not the limiting factor for Pu disposition.

4. MOX FABRICATION

4.1. Fabrication capacity

Table II provides an indicative overview [4,5,7,8,9,10] of the existing and planned MOX fabrication capacity available for LWR, ATR and FNR fuel. It should also be considered upper boundaries. Due to the large investment required and the decommissioning fund to be constituted, implementation of any additional MOX fabrication facility requires assurance that operation at, or close to, rated capacity is possible over a long period of time. The fate of the Hanau MOX plants has not provided industry with confidence in that respect.

4.2. Pu processing capacities

Based on Table II, the existing fabrication capacity is able to fuel 12 GWe of FNRs. This is much more than the 4.1 GWe in operation and under construction, of which a total of 1.7 GWe in France, Japan and 2.4 GWe in Kazakhstan and Russia. For BN 800 FNRs

Table II. MOX Fabrication capacity (tHM/yr)

FUEL	1995	2000	2005
LWR	100	300	410
ATR	10	40	50
FNR	21	22	80

in Russia, no definite planning has been communicated; nevertheless, two first BN 800s have been included in the above figures. Anyway, the contribution to disposition of the Pu stockpile is relatively minor.

In PWR fuels [11,12], the Pu content varies presently between 4.2 and 6.0% Put FA average, with a representative value of 5.3% Put. In the future, MOX discharge burnup will be increased from the current 36 GWd/t to the planned 41-45 GWd/t, a design basis already implemented for the MOX fuel loaded in 1995 in the Belgian PWRs [13]; the Pu content will then rise to a representative value of 6.4% Put. In the next century, when target burnup will increase further and the available CPu from reprocessing LWR fuels will have a fissile content of 62-65% instead of the current 70%, the average Pu content of MOX will reach 7 to 9% Pu t [11,12,13,14].

BWR MOX fuel is now designed for discharge burnups similar to PWR fuel and the average Pu contents range from 4.2 to 8% Put [15,16], depending mainly on the FA design.

Based on these considerations and on Table II, the LWR MOX fabrication plants, which have processed 20 t Put before end 1994, will use as fuel in the future :

- 70 t Put in the period 1995-2000;
- 140 t Put in the period 2001-2005;
- 160 t Put in the period 2006-2010.

ATR fuel production is not limited by fabrication capacity but by NPP fuelling requirements.

4.3. Evolution of the Pu stockpile

Figure 1 provides an overview of the Pu arisings and utilization resulting from the assumptions outlined in Sections 3 and 4. The Pu stockpile should reach its maximum shortly after the year 2000 and should be eliminated by 2017 if only CPu is being utilized and shortly after 2020 if WPu is also utilized.

These conclusions are only valid if Pu obligations and ownership liabilities do not prevent fabrication capacities and MOX fuelling opportunities to be adequately utilized. Since fabrication capacity is the bottleneck for many years to come and fabrication for foreign customers is a commercially established practice, it is unlikely that institutional obligations or regulations will have a major influence in restricting Pu disposition.

However, most MOX manufacturing plants are not designed and/or licensed to use Pu of isotopic composition corresponding to WPu. Some plants are not even provided with a cross-blending step of the feed powders, which thus enables to homogenize Pu feeds of different origins. This complicates the logistics of Pu disposition, but will probably not influence its disposition rate.

The assumed rate of Pu disposition depends on the fabrication capacity assumptions and on the MOX discharge burnup targets, but the prospect of Pu stockpile exhaustion is not much affected. If a 40 tHM/yr additional LWR fuel fabrication capacity starts commercial operation in the year 2000, the exhaustion of the Pu stockpile would take place one to two years earlier. If extended burnup of MOX fuel were implemented 4 years earlier than assumed, the exhaustion of the stockpile would take place one year earlier.

5. Pu CHARACTERISTICS

5.1. Types of Pu

While WPu is relatively uniform in isotopic composition, CPu has a large variety of compositions depending on the reactor in which it was generated and the burnup of the spent fuel it was reprocessed from (Table III). The cumulated quantities of each type of Pu can be estimated (Fig.2) with a fair accuracy. The disposition requirements will be dominated by LWR Pu up to the year 2000, because WPu will not yet be available in significant quantities and thereafter because reprocessing of spent LWR fuel will largely exceed the other Pu arisings.

Isotopic composition has an impact on Pu storage, MOX fabrication and MOX utilization in NPPs.

5.2. Radioactivity

The alpha activity of Pu comes mainly from Pu 238 and Am 241. The heat generation resulting therefrom is a limiting feature for Pu storage as well as for MOX fabrication, storage and transportation.

The gamma activity of Pu consists essentially in the high energy low intensity gammas from the Pu 236 daughter products, the medium energy high intensity gammas from U 237 (resulting from alpha decay of Pu 241) and the low energy high intensity gammas from Am 241. The activity increases with aging : the differences

Table III. REPRESENTATIVE ISOTOPIC COMPOSITION OF Pu (rounded %)

Pu type NPP GWd/t	Pu 238	Pu 239	Pu 240	Pu 241 + Am 241	Pu 242
WPu	0.0	94	5.5	0.5	0.0
GCR 5-6	0.2	69	25	4.9	1.2
AGR 18-24	0.6	54	31	10	5
PWR 33-35	1.6	58	25	10	5.5
43	2.6	56	24	12	6.3
52	2.7	50	28	11	8
BWR 30	2.8	55	23	14	5

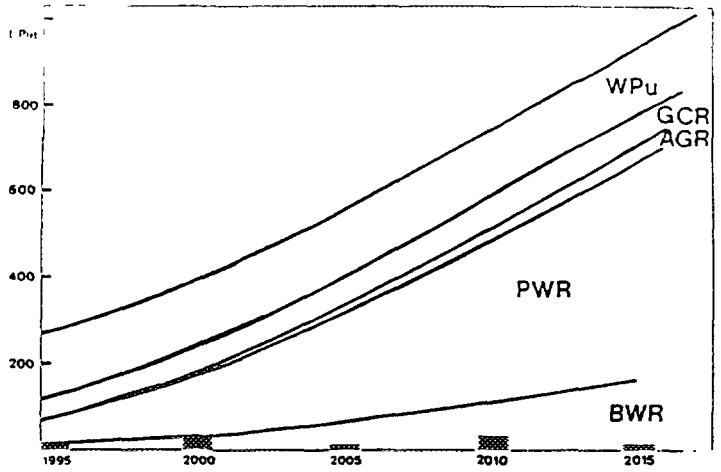

Fig.2. Cumulated Pu quantities requiring disposition

between Pu types after 5 or 10 years ageing is important (Table IV). The gamma activity impacts fuel fabrication, transportation and handling at the reactor site.

The neutron activity of Pu results from spontaneous fissions and from alpha-n reactions in the oxide. Pu 238, Pu 240, Am 241 and Pu 242 are the emitters, with a moderate increase of neutron activity after 5 yr aging for LWR Pu. This radioactivity is very difficult to shield and results in considerable detriment for fuel fabrication, storage, transportation and handling at the reactor site.

Table IV. RELATIVE GAMMA DOSE RATES OF THE DIFFERENT TYPES OF Pu AFTER 5 AND 10 YEAR AGING

Type	WPu	GCR	AGR	PWR	BWR
5 yr	1.0	4.9	9	11	12
10 yr	1.3	8	13	17	20

As concerns radioactivity, WPu and GCR Pu are gentle materials to process and utilize, compared to the other CPus and especially if aging is to be taken into account as the result of temporary imbalance between Pu arising and MOX fabrication capacity.

5.3. Nuclear characteristics [17]

Although all Pu isotopes and Am are fissile in fast neutron fluxes, only Pu 239 and Pu 241 have significant fission cross-sections within the neutron spectrum typical of LWRs. The relative reactivity lifetime value of the various isotopes (if 1.0 is attributed to Pu 239, typically : -0.18 for Pu 238, -0.3 for Pu 240, +1.2 for Pu 241, -1.7 for Pu 242 and -1.8 for Am 241) leads to requiring higher Pu contents for MOX fuel with LWR Pu than with WPu or GCR Pu. The harder neutron spectrum resulting therefrom reduces

the boric acid and control rod worths. Moreover, the quality of LWR Pu is very susceptible to aging : after 10 years storage (as PuO_2 feed material and/or MOX fuel) WPu has lost 1% of its reactivity value, CGR Pu 7%, current PWR Pu 24% and PWR Pu from high burnup spent fuel 31%. The Pu contents of MOX fuel must further be increased accordingly.

The reactivity evolution with burnup, which is much flatter for MOX than for UOX fuel, is only slightly better with WPu and CGR Pu than with LWR Pu. It does not constitute a distinct advantage for any Pu type. Similarly, the 1% higher fission energy of Pu 241 than the Pu 239 can be overlooked.

The interplay of the various Pu isotopes with high absorption resonances in the epithermal energy range (5 000 barns at 0.3 eV for Pu 239, 100 000 barns at 1 eV for Pu 240 and 2 000 barns at 0.26 eV for Pu 241) constitutes a challenge for the calculational methods. In this respect, MOX fuels fabricated from WPu or GCR Pu are much easier to design and license than MOX fuels from LWR or AGR Pu. However, larger inventories of Pu isotopes shift MOX moderator temperature and fuel temperature (Doppler) coefficients to more negative values. The latter is instrumental in early termination of RIAs (e.g. CR ejection in PWRs and CR drop in BWRs); given the attention devoted recently to RIA licensing limitations, LWR and AGR Pu have a definite advantage over WPu. A higher moderator temperature coefficient is a mixed blessing : it improves core stability and stretch-out operation capability, but renders the core more susceptible to accidents involving injection of cold water in the core (e.g. steam-line break in PWRs and turbine trip in BWRs). MOX fuel with high Pu contents and especially high Pu 240 content (such as high burnup MOX fuel fabricated from aged AGR Pu) may exhibit positive void coefficients [18], thus raising new licensing issues. The fraction of delayed neutrons, 0.0022 for Pu 239 and 0.0054 for Pu 241 (close to U 235, also for delayed neutron half-lives), provides for a better control and safety behaviour to MOX from LWR Pu than from WPu.

This glimpse of nuclear design and licensing features reveals pros and cons for each type of Pu feed. A global assessment is that WPu is simpler to manage and therefore better suited for less experienced teams while, on the other extreme, LWR Pu is more sophisticated to design for, but provides improved operational behaviour and licensing margins (unless these are eroded by design uncertainty margins). However, aging deteriorates rapidly the assets of LWR and AGR Pu.

5.4. Spent MOX fuel

During reactor operation, the radioactivity inventory of MOX fuels, at the same power and burnup, is dominated by the fission products. The differences in fission yields between Pu 239 and Pu 241 are small. In accident conditions and for short times after shutdown and unloading, the radioactivity of MOX fuel is almost unaffected by the type of Pu utilized. For interim storage, spent fuel transportation, reprocessing and high level waste (or spent fuel) disposal, the Pu type from which the MOX was fabricated plays an important role, due to the different inventories [19,20] of Pu 238 (which dominates radiotoxicity the first 10-20 years after discharge), Am 241 (which dominates thereafter up to 1 000 years), Cm 244 and Pu 241 (both of which fade out 100 years after discharge). Consequently, for what concerns spent fuel management, WPu is the most gentle type to utilize and LWR Pu is the worst.

6. EXTERNALITIES

The choice of a Pu management scheme cannot be based only on technical, safety and economic considerations, as was the case in the 1960s and 1970s, when Pu was considered primarily as a resource and, hence, as a tradable good. Progressively, non-proliferation, national policy and public acceptability implications have emerged as prime concerns : this resulted in widespread impediments to the free flow of all nuclear goods and services, but particularly Pu. Emphasis shifted from considering it as a resource to booking it as a liability with obligations attached to it and quite a large uncertainty about the future evolution of regulations. Practically, due to the absence of a Pu market, each Pu owner is now forced to care for the disposition of his own Pu in his own system.

For LWR Pu, commercial utilization of MOX started in the mid-1970s and, mainly from the 1980s, expands continuously. Nevertheless, postponement or cancellation of the FBR industrial programmes together with shortage of LWR MOX manufacturing capacity are the main causes of the increase of LWR Pu stockpiles up to the early 2000s. The resulting deterioration of the Pu isotopic composition will degrade MOX fuel fabrication and utilization conditions and might necessitate very expensive Am separation.

AGR Pu starts arising this year, since the operation of THORP has begun. It is also a Pu type with unfavourable aging characteristics. Although MOX was loaded, on a small demonstration level, in WAGR during the 1960s and performed well and notwithstanding the favourable nuclear and safety attributes of graphite moderated cores loaded with MOX fuel, neither the GCR NPPs nor the AGR NPPs can contribute to Pu disposition. The AGR FA design and the fuel route at NPPs for defuelling-refuelling renders adaptation to MOX fuel impracticable. The GCR fuel is metallic with magnox cladding : the manufacturing infrastructure is not available for Pu fuel and compatibility of magnox with metallic Pu fuel is, at best, questionable. In principle, oxide fuel in AGR type cladding can serve as design basis for alternative GCR fuel, but the time required to demonstrate the new fuel design, first with enriched U, then with MOX demo FAs, would shift the start of commercial implementation to ten years from now, i.e. too late, since all CGR stations still in operation are scheduled to close successively in the period 2001-2006 [21]. Only the unique PWR plant operating in the UK and contemplated future PWRs (unlikely to start operation before 2002 and be receptive to MOX fuel before 2004) can provide for the disposition of the accumulating UK stockpile.

GCR Pu has historically been utilized in FNRs, but a large stock has been left unused and will continue to increase (Fig.2) as long as GCRs continue operation. Since GCR Pu deteriorates at a reduced rate with aging, disposition is not a matter of urgency. Nevertheless, for economical (cost of Pu storage) and political reasons, the GCR Pu cannot remain stored for many further decades. It represents, in Europe today, close to 50% of the stockpiled separated Pu.

WPu practically does not deteriorate with aging and, in that respect, long term storage would not be a problem. But leaving military stockpiles neatly stored, ready to be incorporated on short notice in even more efficient warheads than the ones they were recuperated from, is not in the spirit of START. Disposition of WPu, or at least early denaturation to non-weapon Pu quality, is therefore a matter of urgency. The dismantling of warheads and

transformation of low-alloyed Pu metal to PuO_2 in yet non-existent facilities of adequate capacity, results, unfortunately, in WPu not being available for incorporation in MOX fuel before the turn of the century. Furthermore, acute safeguard concerns and feelings that WPu is a national treasure will incline the concerned countries (Russia and USA) to a policy of processing their WPu only in national facilities, at least to a point where WPu is denaturated, e.g. by blending with CPu; but only Russia has and continues to produce separated CPu. It is obvious that no one of these two countries will denaturate its WPu, unless the other country does the same at the same rate. Therefore, availability of WPu to fuel NPPs may even be delayed further. Finally, public acceptance may even be more of a problem for MOX made from WPu and this should not be overlooked.

In short, at the start of next century, most of the Pu utilized by then (Fig. 1) will be LWR Pu and the other Pu types (Fig. 2) will still be stockpiled, half of it being WPu, 30% LWR Pu, 20% GCR Pu and some % AGR Pu.

Notwithstanding severe regulations (a.o. obligations resulting from prior consent rights), flexibility of Pu management has been achieved : the administrative paperwork, safeguard precautions and commercial conditions to allow fungibility between CPu lots, even if present at separate locations, have been developped and implemented. Pu owners and MOX manufacturers are routinely taking advantage of this fungibility to improve Pu management. For the reasons explained before, extension of fungibility to WPu will require determination, innovation and time.

Restrictive regulations have increased the transportation cost of Pu and MOX and administrative restrictions or public acceptance problems have resulted sometimes in unscheduled delays. But, altogether, movability of Pu and MOX is being maintained. Transportation of WPu has also taken place on a large scale in military conditions, but will need to be reorganized into civilian practices.

In summary, there is an imbalance between the arisings and the rate or even the possibility of utilization of each Pu type in isolation. Management of CPu has been resolved by fungibility practices. The obstacles to eliminate for extending fungibility to WPu are important. Only if the incentive to manage rationally CPu and WPu together is important, would it be worthwhile to tackle the problems and seek for solutions.

7. INCENTIVE TO INTRODUCE WPu IN A GLOBAL Pu MANAGEMENT PLAN

Am 241 buildup has emerged as a measure or indicator of the inconvenience of Pu aging. Attention has been focussed so much on this indicator that projects are being launched to purify aged Pu by stripping Am. It reduces part of the radioactivity of aged Pu, but exclusively in the soft energy range, which is only a problem in the MOX fabrication plant and a minor problem in the future, as most recent plants are designed to handle Pu with 3% [22] or even 4% [23] Am 241 contents. Purification also restores part of the reactivity loss due to aging. However, it is costly and the economics of MOX fuel are already the subject of severe criticism. Additionally, the separated Am constitutes an additional HLW stream and a particularly noxious one since, as mentioned in Section 5.4., Am dominates radiotoxicity of spent fuel from 10 to 1000 years after reactor discharge. It could be argued that R & D programmes

are being pursued to transmute minor actinides : as long as those programmes have not produced conclusive results and industrial implementation is not guaranteed, Am would better be left in the Pu and irradiated in the MOX fuel, than shelved for an uncertain future at unknown additional cost.

Although a complete analysis would be necessary, this presentation will, for simplicity's sake, utilize the standard "Am content" measurement stick to glance at potential advantages of a globalized management of all Pu stocks. Moreover, only two scenarios will be illustrated : separate utilization of each Pu type and blending some CPu with WPu.

The first scenario assumes that blending CPu and WPu is not possible and that LWR Pu would be utilized first, then AGR Pu, thereafter GCR Pu and, finally, WPu. The average age and Am content of each Pu type is calculated at the most critical time at which it is processed (Table V), namely in the early 2000s for LWR Pu, when fabrication capacity should start to reduce the separated Pu inventory, and in the early 2010s for AGR Pu, when the UK Pu inventory can be utilized at industrial scale in national NPPs (either new PWRs replacing the shut down GCRs, or prolonged life GCRs with redesigned fuel concept). In this scenario, WPu disposition would take place in the late 2010s and an interim storage of important quantities of WPu in metallic or, even better, in oxide form will have to be undertaken. This constitutes a real weak aspect of the unavoidable interim storage of WPu [24]. In that respect, mixing WPu with CPu is favourable for the non-proliferation aspect, since it complicates plutonium diversion (quantities are bigger, radioactivity is higher).

In the second scenario, the WPu is supposed to become available for processing into MOX in the early 2000s and to be blended in a 1/1 proportion to CPu, to both denaturate the WPu as soon as possible and to cope with the aging of the most critical batches of CPu. Table V illustrates that this notional scenario improves the aging situation over scenario 1. It shows that coordinated management of the global Pu stockpile (or free-trade type fungibility of Pu amongst owners) is capable of providing for a more efficient and safe management of the Pu arisings.

Table V. AVERAGE AGE AND Am CONTENT OF Pu STOCKS AT THE MOST CRITICAL TIME FOR MOX FABRICATION AND UTILIZATION

Scenario	1		2	
Pu type	Age(yr)	% Am	Age(yr)	% Am
WPu	50-60	0.4-0.5		
GCR	34-40	3.9-4.2	35-40	2.2-2.3
AGR	14-15	4.9-5.1	17-18	3.0-3.1
PWR	5-8	2.1-3.2	9-12	2.0-2.4
BWR	5-8	3.0-4.5	9-12	2.7-3.3

Scenario 1 assumes that each Pu stock is undiluted with other Pu types and utilized sequentially.

Scenario 2 assumes that WPu becomes available soon after 2000 and, while stock lasts, it is blended to CPu in a 1/1 ratio, i.e. 50% CPu-50%WPu.

In the real world, all the Pu batches will not be of the average quality of CPu types illustrated in this presentation; some will be worse. The actual individual and national context of each Pu owner will prevail in the constraints he faces and the flexibility he has to manage his Pu arisings. Reluctance and problems to insert WPu in the civilian utilization of MOX are the major obstacles to optimize disposition of the Pu arisings. The difficulties should not be a reason to prejudge that adequate solutions cannot be found : the economic, environmental and safety benefits are large enough to try, even if full success is still not certain.

8. CONCLUSION

The temporary imbalance between CPu arisings and MOX fabrication capacities will result in separated Pu stockpiles building up until the first decade of the 2000s and then progressively being exhausted. During this interim period, the aging of CPu deteriorates the manufacturing and utilization conditions of MOX fuel. The inventory of separated Pu requiring disposition will increase by the WPu obtained from dismantling of warheads. While it prolongs the period before exhaustion of the separated Pu stockpile, a coordinated management of the CPu and WPu would result in big advantages on disposition of Pu stockpiles by accelerating denaturation of WPu and by improving fabrication and utilization of MOX fuel.

Institutional policies, public acceptability problems, safeguard concerns and stalling of progress on WPu disposition prevent the Pu stocks from being processed and utilized in the most economic and safe manner. The international organizations (IAEA, OECD, NATO, EC) can help resolve the issues by organizing working groups, conducting studies, publishing popularization and technical documentation and promoting coherence amongst the member states.

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APPENDIX : ABBREVIATIONS AND ACRONYMS

AEC : Atomic Energy Commission
AGR : Advanced GCR
ATR : Advanced Thermal Reactor : heavy water moderated, light water cooled calandria type (Japan)
CANDU : Pressurized Heavy Water Reactor (Canada)
CPu : Civilian grade Pu
CR : Control Rod
EC : European Commission
FA : Fuel Assembly
FEX : Nuclear Fuel Experts, sa
FNR : Liquid Metal (cooled) Fast Neutron Reactor
GCR : Gas Cooled Reactor (Magnox and UNGG)
HM : U + Pu + Am
HTR : High Temperature Gas (cooled) Reactor
IAEA : International Atomic Energy Agency
LWR : Light Water (cooled and moderated) Reactor
MA : Minor Actinides (Am, Cm, Np, etc...)
MOX : Mixed Oxide (U, Pu)O_2
MSR : Molten Salt (cooled and refuelled) Reactor
NATO : North Atlantic Treaty Organization
NF : Nuclear Fuel
NPP : Nuclear Power Plant
OECD : Organization for Economic Cooperation & Development
ORNL : Oak Ridge National Laboratory
Put : total Pu, i.e. all the Pu isotopes and the Am 241 resulting from decay of Pu 241
RIA : Reactivity Initiated Accident
RPu : Reactor grade Pu
START : Strategic Arms Reduction Treaty
UOX : Uranium Oxide (UO_2, generally U enriched)
WAGR : Windscale AGR
WPu : Weapon grade Pu

A ONCE THROUGH SCHEME FOR WEAPONS GRADE PLUTONIUM DISPOSITION IN LWRS: PROLIFERATION AND CRITICALITY ASPECTS

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Abstract

We investigate a novel scheme for burning weapons grade plutonium (W-Pu) in conventional light water reactors with a view to optimising the destruction rate of plutonium and increasing the proliferation resistance of the resulting spent fuel. The main feature of this scheme is the use of a fuel based on W-Pu and weapons grade uranium (W-U) in an inert matrix. The extended irradiation (þ2000 days) of this fuel leads to a high destruction rate (95%) of the Pu. Proliferation resistance of the spent matrix fuel is guaranteed by the very high levels of ^{238}Pu in the total plutonium (þ30%). The technicalities of how such inert matrix based fuel pins, with lifetime approximately 2000 days, may be used together with standard pins, with lifetime approximately 900 days in a PWR is described together with the criticality changes which may be expected through the use of fuel pins based on the inert matrix. On a timescale of a few hundred years the levels of ^{238}Pu, with half-life of 88.75 years, will decrease to around 5%. High proliferation resistance of the spent matrix based fuel can be recovered by re-irradiating the matrix for a further period of 1000 days. The process of re-irradiation and decay can be repeated, *without reprocessing*, for as long as the spent matrix is considered a proliferation risk.

1. INTRODUCTION

The relaxation in tension between the United States and the former Soviet Union, witnessed over the past few years, has given rise to tens of thousand of nuclear warheads on both sides which have been declared excess to current needs. According to the U. S. National Academy of Sciences report [1] on *Management and Disposition of Excess Weapons Plutonium* published recently, the excess amounts to 50 tonnes of weapons grade plutonium (W-Pu) in the U.S.A. At a recent NATO Advanced Research Workshop on *Managing the Plutonium Surplus: Applications and Options* [2] Russian papers consider the Russian excess of W-Pu to be 100 tonnes. At present the W-Pu and highly enriched uranium (W-U) are present in warheads and will have to be transferred for dismantling. In this process a major problem will arise with regard to proliferation of nuclear weapons to national powers or sub-national groups by theft or unauthorised diversion.

One option is to dispose of this material by 'burning' it in nuclear reactors and this is the subject of the present paper. The problem with this approach is the fact the even the 'ashes' resulting from burning of W-Pu (i.e. the spent fuel) can be used to make a nuclear explosive [3]. Following irradiation in the reactor, the spent fuel can be removed from the reactor, dissolved, and chemically processed to separate the plutonium which could then be used to construct a nuclear device. The compositions of different types of uranium and plutonium [3,4] are given in Tables I and II.

Although more difficult than with W-Pu, reactor plutonium (R-Pu) can be used to construct a nuclear explosive. It should be noted that although the critical mass of R-Pu is greater than that of W-Pu (see Table III), *the critical mass of R-Pu is less than that for weapons grade uranium (W-U).* In addition, heat generation in R-Pu is about a factor five higher than in W-Pu and the spontaneous emission of neutrons is about a factor 6 higher than in W-Pu. In Table III a summary of critical masses, heat production and neutron emission rates are given for various isotopes and isotope mixtures.

Table I: Composition (%) of different types of uranium.

	^{235}U	^{238}U
Natural Uranium	0.7	99.3
Weapons Grade Uranium (W-U)	93.5	6.5

Table II. Composition (%) of different types of plutonium.

	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
MOX Grade Plutonium (R-Pu)*	1.34	62.77	23.48	8.31	4.1
Weapons Grade Plutonium (W-Pu)	0	94	5.3	0.7	0

* Plutonium recovered from uranium pressurised water reactor fuel with burnup 33GWd/tonne

Table III. Critical masses, spontaneous neutron emission and heat generation rates of various isotopes and mixtures [3,4].

Isotope	$t_{1/2}$ (y)	Bare critical mass (kg)	Spont. Neutron Emission Rate $(kg\ s)^{-1}$	Isotopic Power $(W\ kg^{-1})$
^{235}U	7.04×10^8	49	0.4	$\cong 0$
^{238}U	4.47×10^9	-	15	$\cong 0$
^{238}Pu	87.75	9/15*	2.32×10^6	560
^{239}Pu	2.41×10^4	10/15*	29	1.9
^{240}Pu	6.54×10^3	40/60*	9.24×10^5	6.8
^{241}Pu	14.4	12/15*	49	4.2
^{242}Pu	3.76×10^5	90/177*	1.85×10^6	0.1
^{241}Am	432	114	1.55×10^3	114
W-Pu	-	$\cong 11$	5×10^4	2.2
R-Pu	-	15	3.2×10^5	10.7
W-U	-	52	1.3	$\cong 0$

*α/δ phases of Pu

In this paper we propose a scheme to optimise the destruction rate of W-Pu and increase the proliferation resistance of spent fuel by increasing the relative amounts of the isotope ^{238}Pu present in the 'ash'. The destruction rate of Pu is maximised by embedding the W-Pu in an inert matrix, thereby avoiding the production of Pu from a fertile matrix. Methods for increasing the amounts of the isotopes of ^{238}Pu considered so far require reprocessing of the spent fuel to obtain ^{237}Np or ^{236}U which can then be added to the fresh fuel [5]. In the present approach, we avoid the reprocessing step by using weapons grade uranium as the additional source of ^{238}Pu through the reaction path shown in Fig.1 (the normal source from ^{239}Pu is also shown). This use of weapons grade U requires an extended burnup to avoid proliferation problems associated with the ^{235}U in the spent fuel. Only after about 2000 days irradiation in

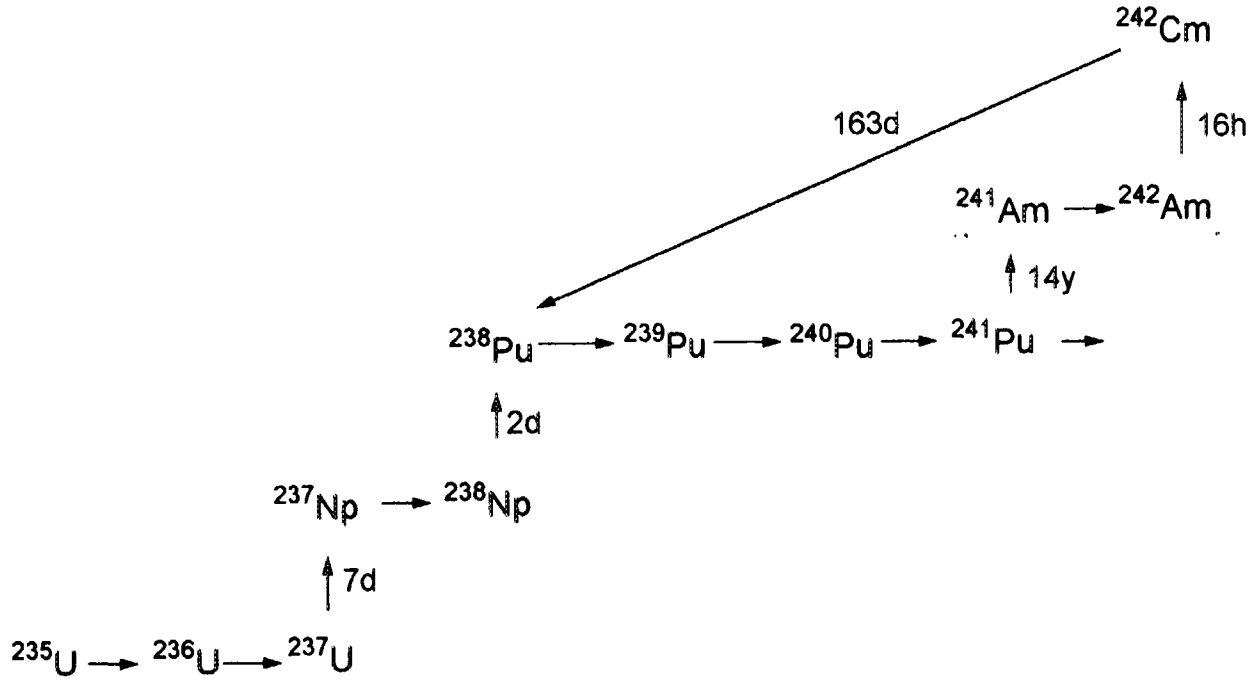

Fig.1. Main pathways for the production of ^{238}Pu from ^{239}Pu and ^{235}U. The sequence of (n,γ) capture processes is interrupted whenever a short lived nuclide is formed.

a PWR neutron spectrum (total flux = $3x10^{14}cm^{-2}s^{-1}$) is the percentage of ^{235}U in the uranium less than 20% i.e. the concentration below which enriched uranium is not considered as a proliferation risk (the bare critical mass of a 20% mixture of ^{235}U in ^{238}U is approximately one tonne [6]). This long irradiation, of course, then leads to a high level of Pu destruction. In the following sections the proliferation and criticality aspects of such an inert matrix containing mixed fissile material are considered.

2. CALCULATIONAL DETAILS

The calculations were made using the point depletion code ORIGEN2 [7] and checked independently with RADONN [8]. The one group cross section library used, PWRUS.lib [7], was that for a standard pressurised water reactor with 3.2% ^{235}U initial fuel enrichment, power level of 37.5 MW/tonne, and burnup of 33 GWd/tonne (achieved after 879 EFPD-effective full power days). To simulate the irradiation of a pin with a different fuel in this reactor, the following procedure was adopted: An initial calculation was made for the standard fuel configuration (i.e. 3.2% ^{235}U, 33 GWd/t) to determine the average neutron flux during irradiation.

Using this average neutron flux and the same one group cross sections (thereby ensuring that the neutron flux and spectrum correspond to a 3.2% ^{235}U enriched fuel with a burnup of 33 GWd/t over 879 EFPD), a new fuel configuration was defined e.g. inert matrix plus fissile material or as MOX, and the calculations were repeated for the irradiation period of 879 EFPD.

A difficulty which arises with the above procedure is that in the initial calculation burnup dependent cross sections are used to account for changes in the neutron energy spectrum brought about by the buildup of additional fissionable isotopes. Depending on the burnup, different cross sections are used especially for the plutonium isotopes. If a different fuel type such as the inert matrix-based fuel is used, the code assumes the core consists entirely of this material. The burnup of this material in the previously defined neutron energy spectrum is clearly different from the standard fuel but it is the inert matrix-based fuel which determines the burnup and therefore which cross sections are to be used.

This problem can be overcome by using burnup independent cross sections. In the present calculations, the burnup independent cross sections have been obtained by linearly averaging the burnup dependent cross sections as shown in Table IV.

Using these averaged cross sections then ensures that the material to be irradiated has no effect on the cross sections used. Clearly, there is no difficulty in extending the calculations to 1757 and 2000 days.

3. PROLIFERATION ASPECTS OF INERT MATRIX-BASED MIXED U-PU FISSILE MATERIAL

We consider the evolution during irradiation of an inert matrix based fuel pin containing 2.5% W-Pu and 2.5% W-U in a PWR. It is assumed, for simplicity, that the inert matrix has the same density as that of the standard fuel. The relevant details of the PWR have been described in the previous section. It is also assumed that the presence of the inert matrix plus fissile material does not disturb the neutron spectrum of the reactor. The results of such an irradiation are shown in Figs. 2 and 3 and summarised in Table V for the plutonium and uranium isotopes respectively. Here the masses of isotopes present per tonne of inert matrix are given at various irradiation times.

As can be seen from Fig. 3 and Table V, an irradiation time of approximately 2.3 cycles (2.3 × 879 days) of the W-U is required to ensure that less than 20% of the total uranium is ^{235}U. This consideration of the proliferation resistance of the W-U thereby fixes the irradiation time for the matrix. During this irradiation period, the plutonium isotope distribution shift almost entirely to ^{242}Pu. The large amounts of ^{238}Pu present originate from the irradiation of the W-U.

Table IV. One group burnup dependent and averaged cross sections for a PWR.

		Cross sections (barns) for fuel burnup (MWd/g-atom heavy metal) of:					Average
Nuclide	Reaction	0.0	2.0	4.0	6.0	8.0	
^{239}Pu	capture	69.09	64.31	59.93	55.85	52.05	60.3
^{239}Pu	fission	121.1	113.7	106.7	100.2	94.07	107.2
^{240}Pu	capture	222.8	176.2	139.4	110.2	87.17	147.2
^{241}Pu	capture	42.02	40.23	38.51	36.87	35.29	38.6
^{241}Pu	fission	125.9	121.4	117	112.8	108.7	117.2
^{242}Pu	capture	33.2	32.4	31.63	30.87	30.13	31.7
^{241}Am	capture	95.7	90.28	85.17	80.35	75.18	85.5

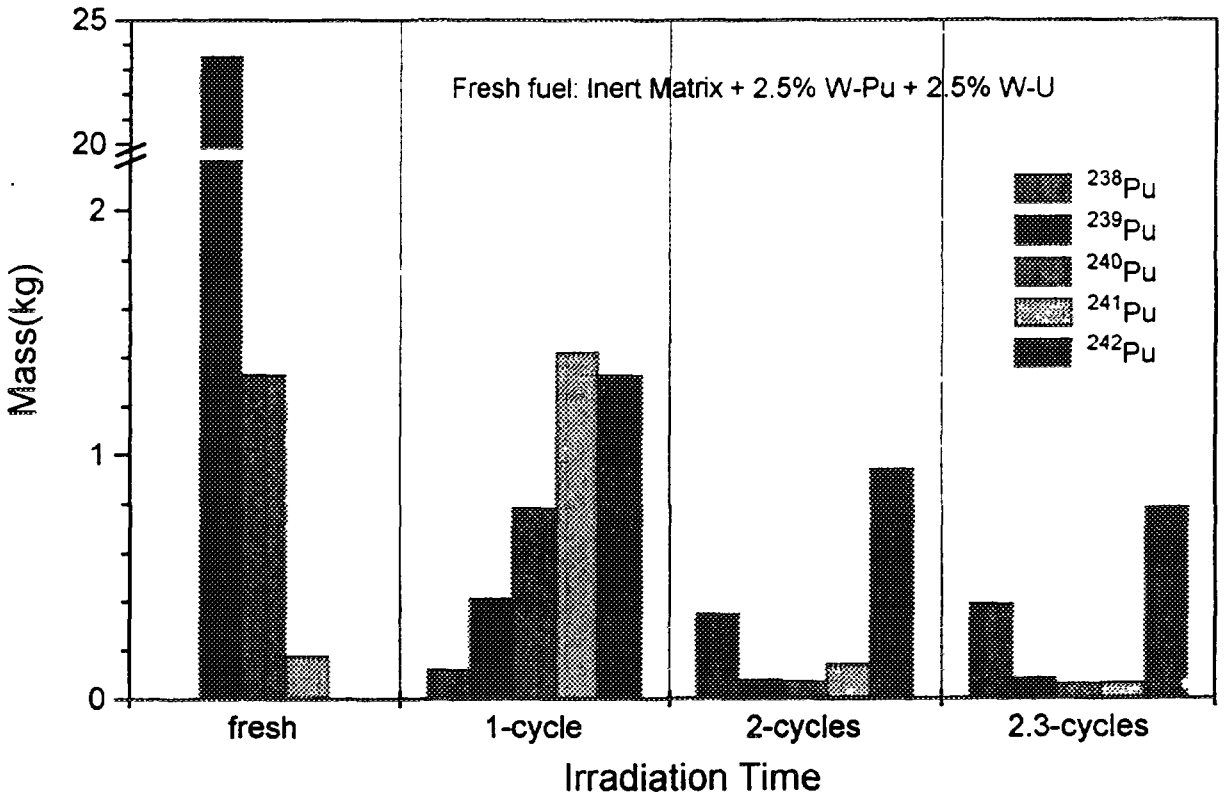

Fig. 2. Pu isotopic masses per tonne of matrix: in fresh fuel, and after 1, 2, and 2.3 cycles in a PWR (1cycle=879days).

Also from Table V, approximately 94% of the total plutonium has been destroyed over 2.3 cycles. What remains is mostly the isotope ^{242}Pu (which from Table III has a critical mass of 100 kg) with approximately 28% ^{238}Pu. If the spent fuel were to be reprocessed to separate out the plutonium, very high heating rates in excess of 150 Watt / kg of Pu metal are to be expected as can be seen in Fig. 4. Since the

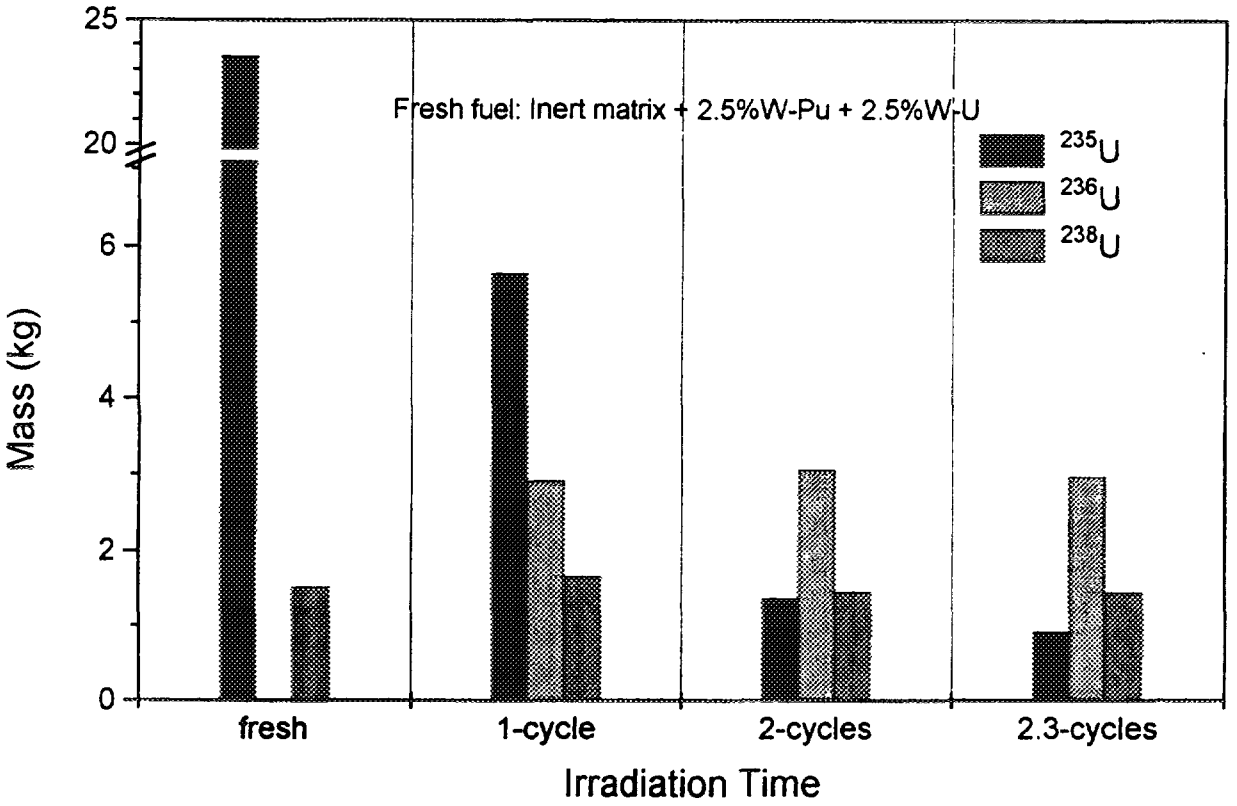

Fig.3. U isotopic masses per tonne of matrix: in fresh fuel, and after 1, 2, and 2.3 cycles in a PWR (1cycle=879days).

Table V. Evolution of an inert matrix containing 2.5% W-Pu+2.5%W-U in the neutron spectrum of a PWR (1cycle = 879 EFPD). Densities of matrix and standard fuel are assumed equal.

	fresh	1 cycle	2 cycles	2.3 cycles
total U	25kg	10.2kg	5.8kg	5.3kg
$^{235}U/U$	94%	55%	23%	17%
total Pu	25kg	4.1kg	1.6kg	1.4kg
$^{238}Pu/Pu$	0	2.9%	22%	28%

critical mass of this mixture is approximately 50 kg [8], heating rates of 7.5 kwatt per device will have to be contended with! Clearly, from this result the amounts of W-U present in the fresh matrix could be considerably reduced.

The neutron emission rates from such chemically separated plutonium are shown in Fig. 5. After 2.3 cycles the neutron emission rate is a factor 32 higher than from W-Pu but only a factor of 2 higher than from R-Pu which is obtained by reprocessing the fuel after 1 cycle.

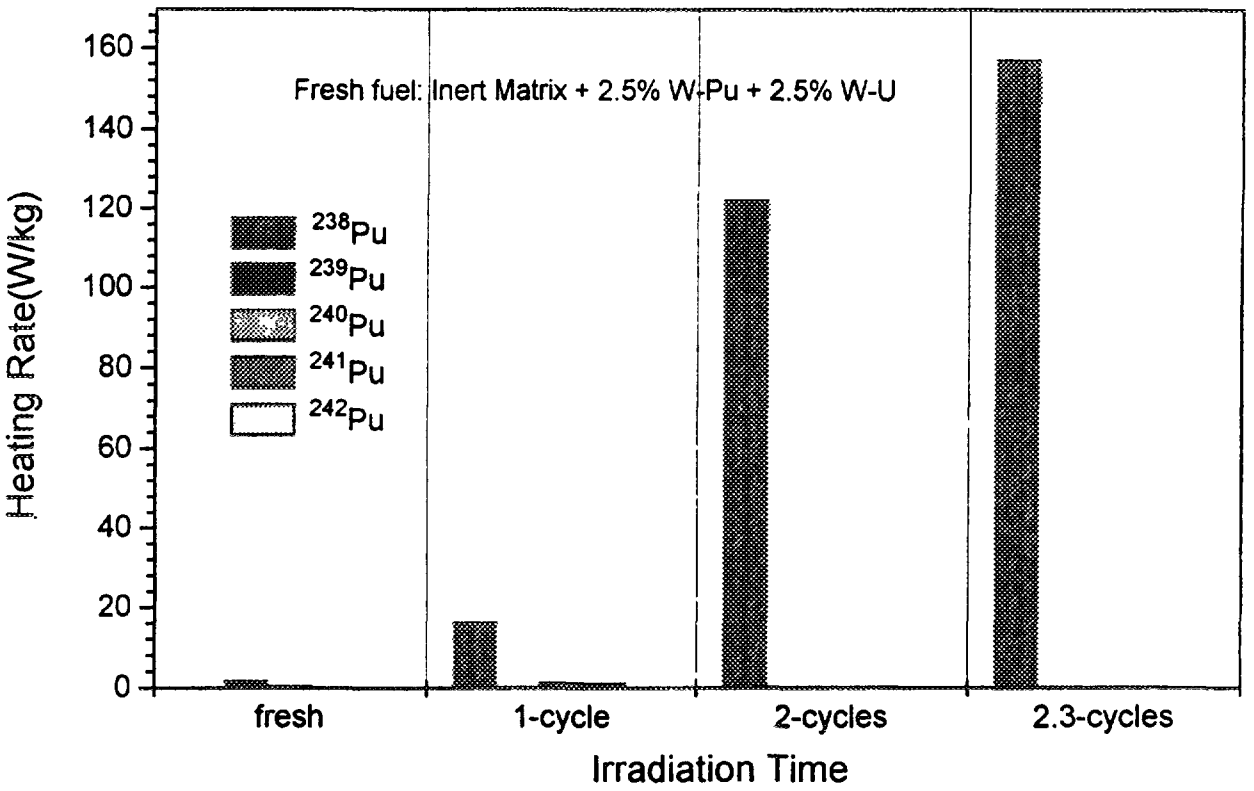

Fig. 4. Isotopic heating rates per unit mass of total Pu: for fresh fuel, and after 1, 2, and 2.3 cycles in a PWR.

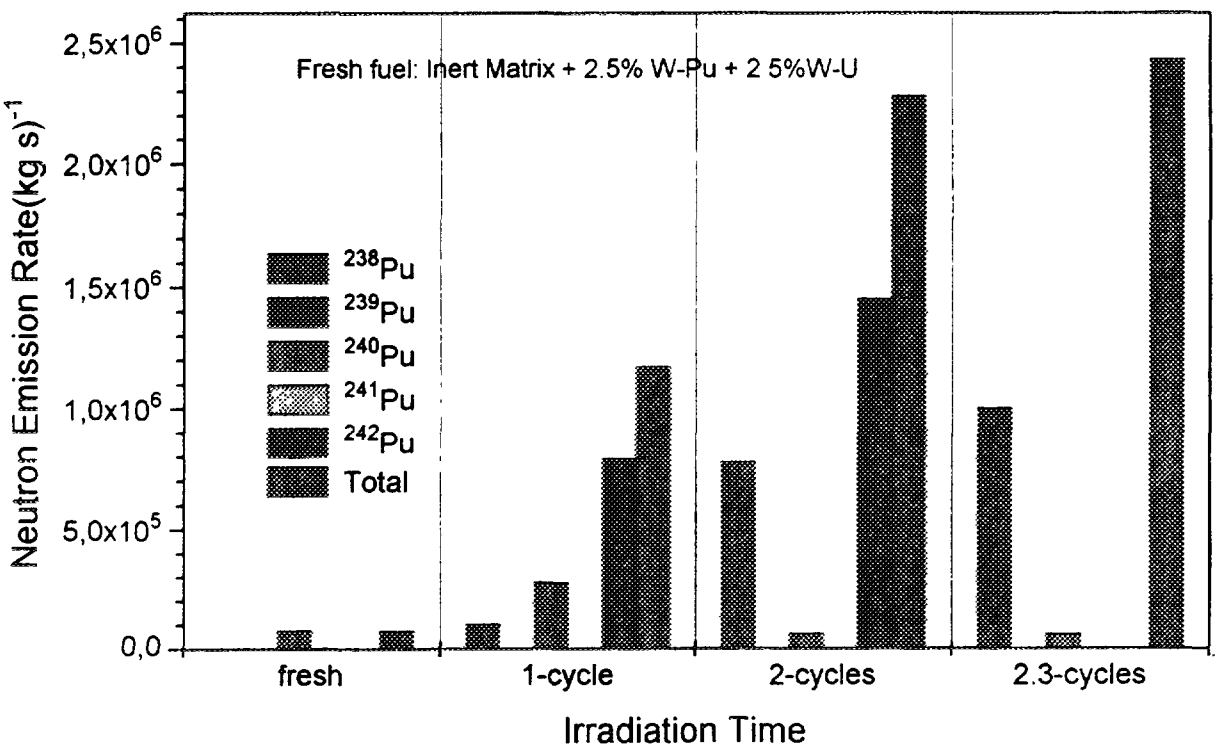

Fig. 5. Isotopic neutron emission rates per unit mass of total Pu: for fresh fuel, and after 1, 2, and 2.3 cycles in a PWR

4. CRITICALITY ASPECTS OF INERT MATRIX-BASED MIXED U-PU FISSILE MATERIAL

In the calculations it was assumed that the presence of inert matrix based fuel pins did not disturb the neutron flux in the reactor. This is only true provided that the variations for the standard and inert matrix based fuels are not too different. In this section we compare the k_∞ variations resulting from the irradiation of standard and inert matrix based fuels.

In Fig. 6 the variation of k_∞ of the standard fuel configuration (described in the previous section) of 3.2% ^{235}U in a natural U matrix is shown over one cycle (i.e. 879 days). Also shown is the average thermal power generated. The variation of k_∞ of an inert matrix containing 2.5% W-Pu and 2.5% W-U in a neutron flux of the LWR is shown in Fig 7. The irradiation time extends to 2000 days corresponding to 2.3 cycles of the LWR. Clearly, large variations in k_∞ can be seen. Indeed after about 900 days irradiation the configuration inert matrix plus fissile material becomes subcritical. It should be noted here that although $k_\infty < 1$ this material is still producing net energy with a multiplication factor $M \propto 1/(1-k_\infty)$.

How can such an irradiation be accomplished within a LWR with a standard fuel life of 879 days? One possible scheme is to consider the core composed of an inner core and an outer blanket with the following properties:

- the inner (critical) core contains standard and inert matrix fuel pins with $k_\infty > 1$.
- an outer (sub-critical) blanket of irradiated inert matrix fuel pins with $k_\infty < 1$.

In steady operation of the reactor, after each 293 days (one third of a standard cycle):

- one third of the standard pins (i.e. those which have been in the reactor for 879 days) are removed from the reactor core and replaced by fresh pins,
- one third on the inert matrix fuel pins irradiated in the central core for 879 days are removed to the blanket where they remain for another 1.3 cycle irradiation. These inert matrix pins are replaced by fresh inert matrix pins,

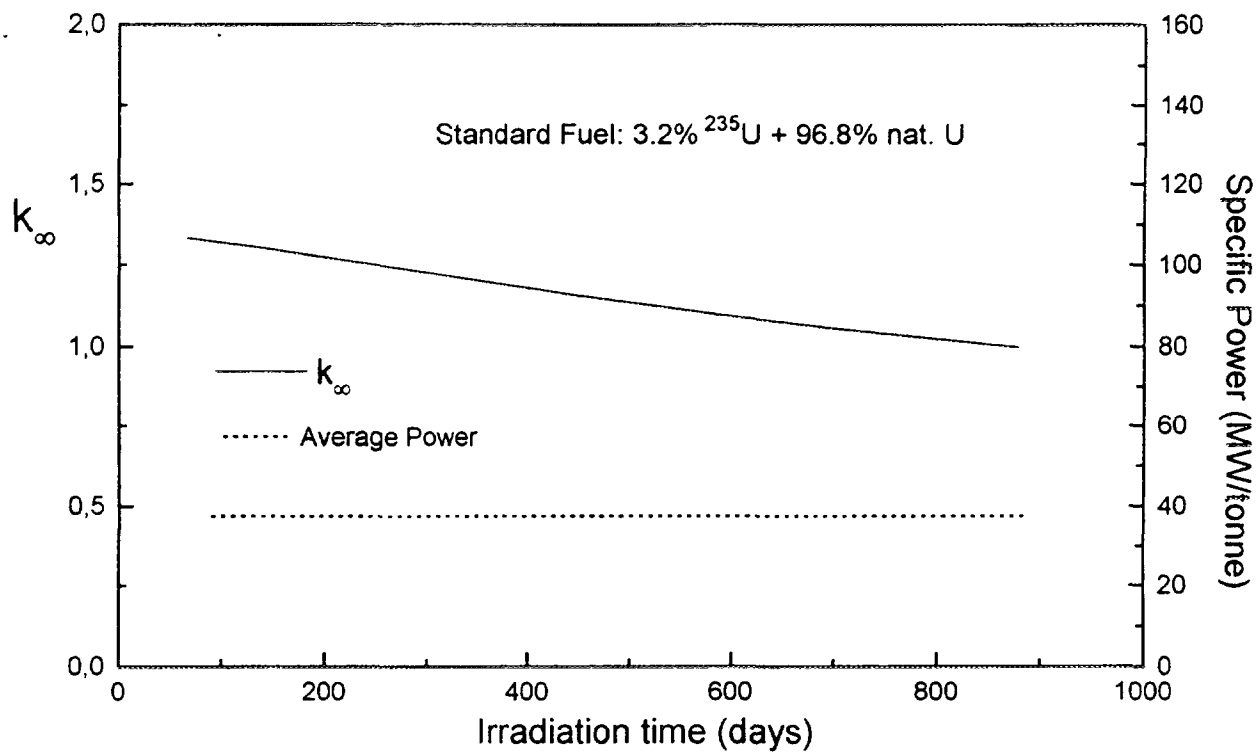

Fig. 6. Variation of k_∞ and average power with time for standard PWR fuel.

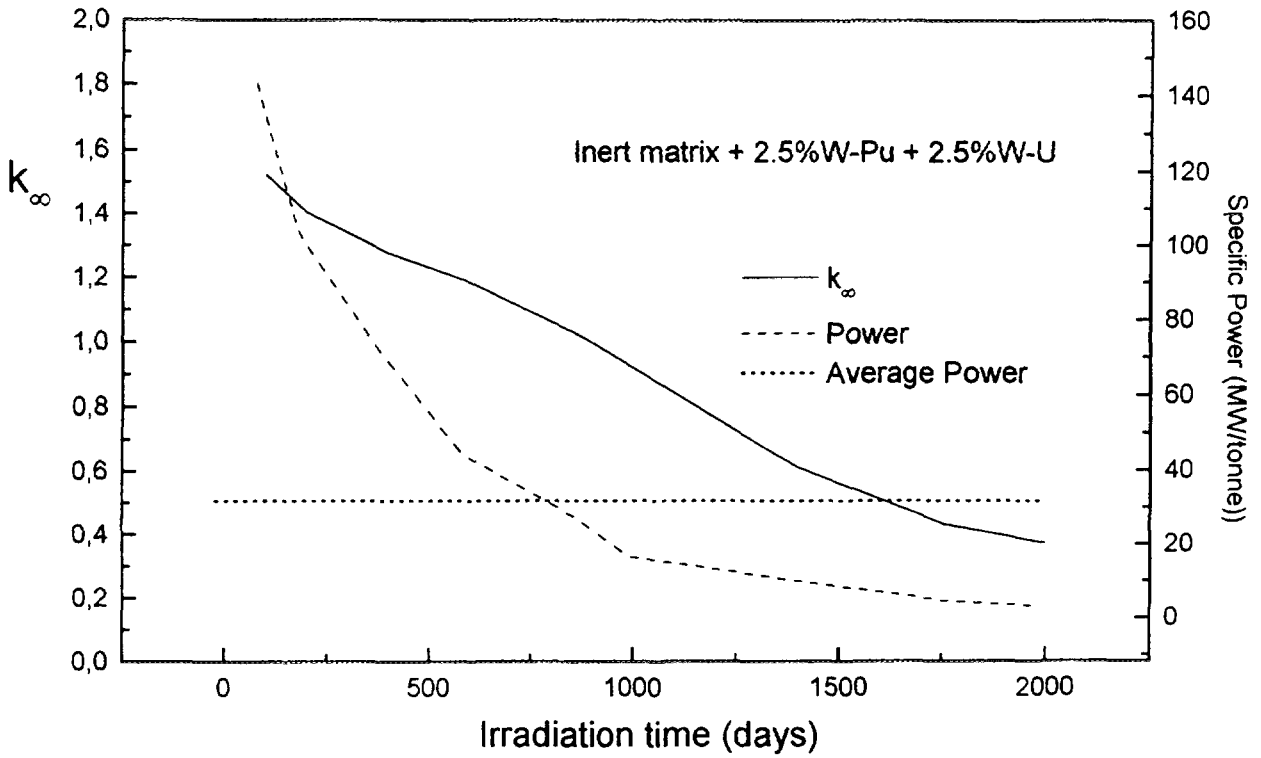

Fig. 7. Variation of k_∞ and power with time for the inert matrix based fuel in a PWR neutron spectrum.

- one quarter of the inert matrix pins in the outer blanket of the reactor are removed (these pins have been in the reactor for a period of 7 × 293 = 2050 days) from the reactor.

Such a scheme will ensure that the standard pins are in the reactor for 878 days whereas the inert matrix pins are in the reactor for the required 2050 days. It is not a requirement that the outer blanket should contain the sub-critical inert matrix based pins. Depending on the neutron distribution, this array of sub-critical pins could be placed in an annular cylinder in the core region or dispersed through the core.

The above heterogeneous core layout cannot be investigated with a zero dimensional code such as ORIGEN2. However, an approximate treatment of such a heterogeneous core can be made by considering a homogeneous core containing fuel at different stages of irradiation. Hence a standard core can be considered as containing fuel - one third of which is fresh, one third has been irradiated for 293 days and one third irradiated for 586 days. The variation of k_∞ over one third of a cycle i.e. 293 days is shown in Fig. 8. Similarly, one can consider a core consisting of inert matrix based fuel. Since the fuel here is in the reactor for seven periods of 293 days (≅2000 days), the homogeneous material contains a mixture - one seventh of which is fresh, one seventh irradiated to 293 days, one seventh irradiated to 586 days, etc. Notice that this core is being irradiated with the spectrum of the standard PWR. Provided that the variation of k_∞ with time is not too different from that of the standard PWR, this should be a reasonable approximation. The extent to which this is the case may be seen in Fig. 8. Here it can be seen that by varying the amounts of fissile material i.e. W-Pu and W-U, the ratio W-Pu:W-U, and through the use of a burnable neutron absorber [9], the variation of k_∞ can be approximated to that of the standard reactor.

5. SOME CONSIDERATIONS ON INERT MATRICES

In the previous sections the inert matrix has not been defined. In the calculations a given amount of weapons grade material is contained in a volume of inert matrix. If the density of the inert matrix were

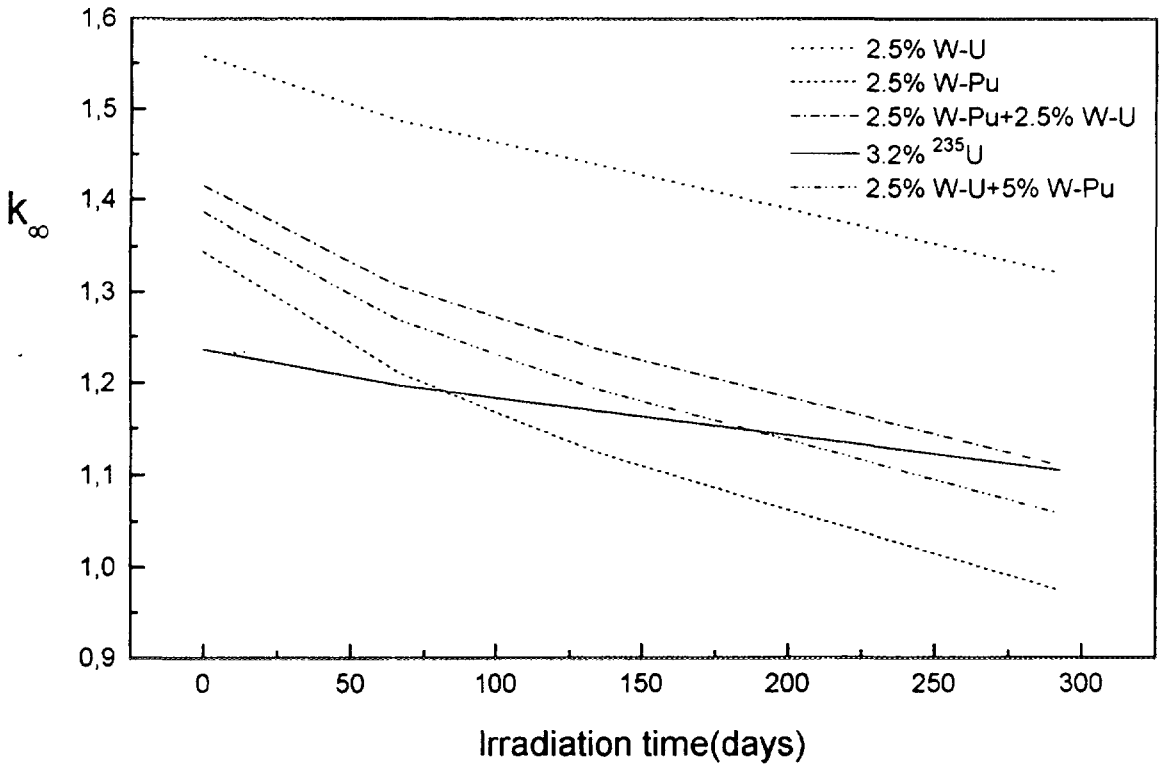

Fig. 8. Variation of k_∞ with time for different fuels and mixing ratios of W-Pu and W-U.

the same as that of the UO_2 fuel, then approximately 50 kg of W-Pu per tonne of matrix (i.e. 5% assuming equal densities) is required for equivalent power production.

The inert matrix is a material which replaces conventional UO_2. It should not form actinides by neutron capture, but otherwise have properties similar to or superior to those of UO_2. To avoid actinide formation, the atomic number of all components should be significantly less than 92. To have good reactor properties, the melting point T_m should be high (greater than 2000C; for UO_2 T_m = 2880C), the thermal conductivity and mechanical properties should be equivalent to those of UO_2, and the matrix should be compatible with the cladding and coolant. Finally, it should not have a high neutron capture cross section in order to obtain good neutron economy.

Two classes of ceramics can be selected which meet these criteria [10,11]. One class contains Al_2O_3, spinel ($MgAl_2O_4$), or MgO which have no solid solubility for actinides. In this case, a two phase heterogeneous fuel will result with UO_2 or PuO_2 particles in the inert matrix. The second class contains CeO_2, and zircon ($ZrSiO_4$), and shows solid solubilities high enough to allow fabrication of a homogeneous fuel. A major research activity is being pursued at ITU [12] to investigate fabrication routes and properties of the above candidate ceramics in order to define the most suitable matrix.

6. DECAY OF ^{238}Pu

Because the half life of ^{238}Pu is 87.75 y, the proliferation resistance of the spent inert matrix-based fuel will decrease with time. After approximately 200 years the level of ^{238}Pu in the Pu is less than 5% as can be seen in Fig. 9. If at this time the spent fuel is still considered a proliferation risk, one need only re-irradiate the material again without reprocessing. The result of doing this every 200 years up to 800 years is shown in Fig. 9. The neutron flux used here is that of the standard PWR. Finally, in Fig. 10 we show the variation of the masses of the Pu isotopes resulting from a series of irradiation and decay steps

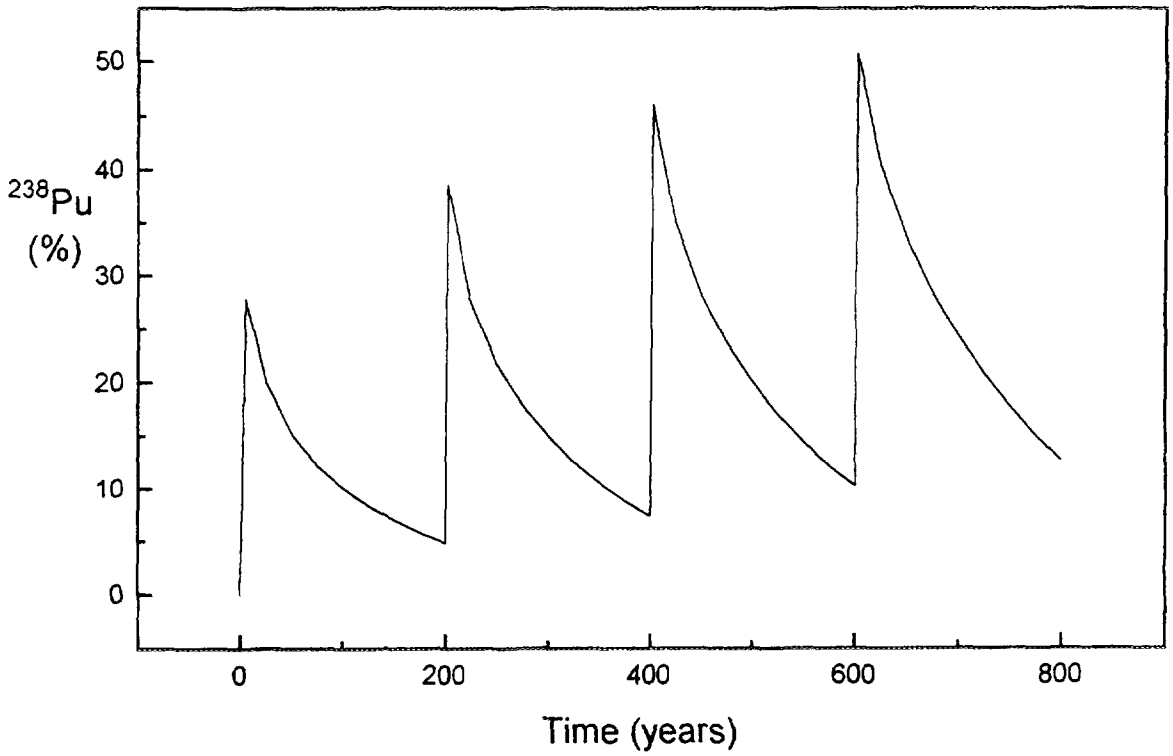

Fig. 9. The percentage of ^{238}Pu in total Pu resulting from irradiation and decay of an inert matrix containing 2.5%W-Pu+2.5%W-U. Initially the fresh matrix is irradiated for 2000 days. At 200, 400, and 600 years, the matrix is re-irradiated for 1000 days.

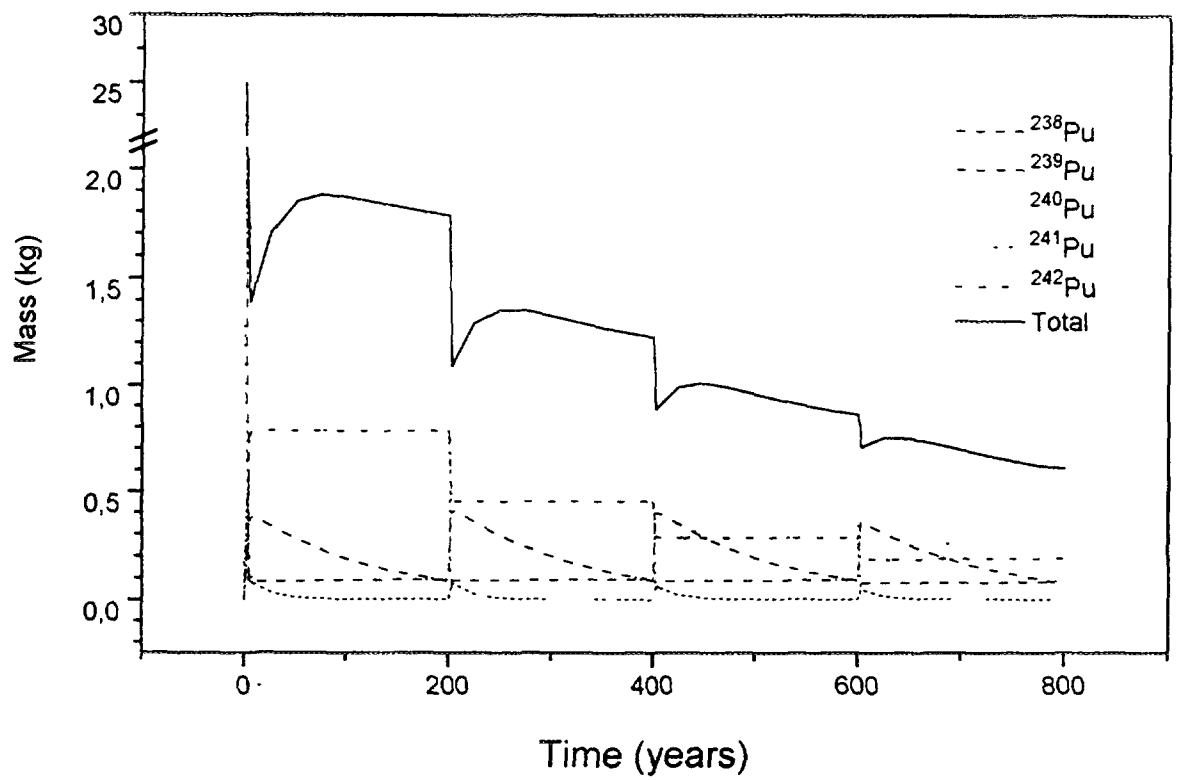

Fig. 10. The masses of Pu isotopes resulting from the irradiation and decay of an inert matrix containing 2.5%W-Pu+2.5%W-U. Initially the fresh matrix is irradiated for 2000 days. At 200, 400, and 600 years, the matrix is re-irradiated for 1000 days

over a period of 800 years - starting with the fresh W-Pu in the inert matrix. The fresh matrix is irradiated for 2000 days. At 200, 400, and 600 years the matrix is re-irradiated for 1000 days. From Fig. 10 it can be seen that following an irradiation step, in which the total Pu decreases rapidly, the total Pu increases. This is due to the increasing amounts of ^{240}Pu which arise from ^{244}Cm formed during the irradiation step which then decays with a half-life of about 18 years.

7. CONCLUSIONS

An analysis of a novel scheme for W-Pu disposition by burning the plutonium in LWRs has been presented. The main characteristics of this scheme are that it provides a high destruction level of the W-Pu and a high proliferation resistance of the spent fuel ashes in a once through cycle. The high destruction level of the W-Pu is guaranteed by embedding the W-Pu in an inert matrix and, in addition, by irradiation in a standard PWR for 2000 days. After this time essentially only ^{242}Pu is present from the W-Pu irradiation. Proliferation resistance of the spent fuel is considerably increased by the use of W-U in the fresh fuel. The presence of this material leads to an additional source of ^{238}Pu which, after irradiation for 2000 days, forms approximately 30% of the total plutonium. In contrast to alternative methods for increasing the amounts of ^{238}Pu in the spent fuel, which are based on the use of ^{237}Np or ^{236}U obtained by reprocessing, the present method requires no reprocessing of the inert matrix-based fuel. The requirement of an irradiation time of approximately 2000 days in the reactor ensures that less that 20% of the total uranium remaining is ^{235}U, thereby overcoming a potential proliferation problem with the irradiated uranium (notice that the 20% level applies to separated uranium - in the present case, since this is mixed with alpha active waste, the 20% level could be significantly increased implying an irradiation time less than 2000 days). The materials problems of inert matrices and clad for such irradiations has not been considered but are under investigation [12].

The compatibility, with regard to neutronics and reactor operation, of mixing standard fuel pins with lifetime of 879 days with inert matrix based pins with lifetime of 2000 days in a standard PWR reactor is considered and a scheme for fuel pin exchange is proposed. An analysis of the criticality aspects of this scheme is made by assuming that the heterogeneous system can be approximated by a homogenous core consisting of fuel at different stages of irradiation. This analysis shows that the standard PWR variation in criticality, based on the variation of k_∞, can be closely approximated by varying the total amounts of fissile material in the inert matrix and by varying the mixing ratio of W-Pu/W-U. Hence, as far as neutronics and criticality are concerned there should be little difficulty in the use of inert matrix-based fuel pins in a standard PWR. The limitations of the use of such pins will be determined by other factors such as the requirement of a negative temperature coefficient, peak power limitations, fuel material behaviour, cladding resistance, and fuel pellet interactions.

Finally, since ^{238}Pu has a half-life of approximately 88 years, the proliferation risk of the spent matrix based fuel will decrease with time. Over a period of 200 years the level of ^{238}Pu in the plutonium will drop below 5% - the minimum value required for proliferation resistance. The advantage of the present scheme is that the high level of ^{238}Pu can be restored by re-irradiating the spent matrix for a further period of 1000 days. The process of re-irradiation and decay can be repeated, *without reprocessing*, for as long as the spent matrix is considered a proliferation risk.

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PLUTONIUM DISPOSITIONING IN CANDU

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Abstract

Recently, the U.S. Department of Energy (DOE) sponsored Atomic Energy of Canada Limited (AECL) to evaluate salient technical, strategic, schedule, and cost-related parameters of using CANDU[1] reactors for dispositioning of weapons-grade plutonium in the form of Mixed OXide (MOX) fuel. A study team, consisting of key staff from the CANDU reactor designers and researchers (AECL), operators (Ontario Hydro) and fue suppliers, analyzed all significant factors involved in such application, with the objective of identifying an arrangement that would permit the burning of MOX in CANDU at the earliest date. One of Ontario Hydro's multi-unit stations, Bruce A nuclear generating station (4x769 MW(e)), was chosen as the reference for the study. The assessment showed that no significant modifications of reactor or process systems are necessary to operate with a full MOX core. Plant modifications would be limited to fuel handling and modifications necessary to accommodate enhanced security and safeguards requirements. No safety limitations were identified. An important task of the study was to define the optimum design parameters of MOX fuel to achieve the target disposition rates -- two tonnes of plutonium per year in the reference case, and four tonnes per year for an alternative case -- without altering the design base of operating and safety parameters of the reactor system, and without requiring excessive fuel supply. The reference MOX fuel design employed the standard 37-element CANDU geometry bundle. This fuel would operate within the same burnup and power rating envelope as standard CANDU natural-uranium fuel, and its nuclear parameters would allow the reactor to operate within its existing licensing envelope. The 43-element CANFLEX fuel bundle was chosen for the alternative case. This fuel bundle, which is currently being qualified for commercial use, has two sizes of elements, and operates at a lower linear power rating, thus permitting higher plutonium concentrations and higher burnups. Use of this design would reduce the quantity of MOX fuel bundles required by almost half, a significant economic advantage. Two Bruce A reactors would be used for the reference case of dispositioning of two tonnes of plutonium per year, and four for the alternative case of dispositioning of four tonnes per year.

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[1] CANDU: CANada Deuterium Uranium; registered trademark

INTRODUCTION: OVERVIEW OF PLUTONIUM DISPOSITIONING IN CANDU

The U.S. Government is currently considering about a dozen options for dispositioning of weapons plutonium. The U.S. National Academy of Sciences, in its 1994 January report, "Management and Disposition of Excess Weapons Plutonium", urged the U.S. and Russian Governments to act expeditiously to demilitarize excess fissile material from dismantled nuclear weapons, calling the continued availability of such materials, even when placed in safe storage, a "clear and present danger".

As part of this assessment, the U.S. DOE sponsored AECL to lead a study on dispositioning weapons-grade plutonium using CANDU technology. To perform this study, AECL Technologies Inc., the U.S. corporation of AECL, assembled a team to analyze significant technical, strategic, schedule, and economic aspects of plutonium dispositioning in CANDU. The team consisted of AECL's reactor design and research staff, Ontario Hydro (which owns and operates twenty CANDU reactors), the U.S. DOE Hanford site contractor (which manages an existing facility that could be converted to fabricate CANDU MOX fuel), Zircatec Precision Industries (a Canadian CANDU fuel supplier that fabricates about half of the natural uranium fuel bundles used in Canada), MOX fuel fabrication experts from Babcock and Wilcox, and technical experts from Gamma Engineering Corporation. In addition, input was provided by the IAEA, the Atomic Energy Control Board (the Canadian nuclear regulator), and DOE's Los Alamos and Lawrence Livermore National Labs.

The focus of the study was on utilizing the ex-weapons plutonium as MOX fuel in CANDU. The U.S. DOE specified a plutonium disposition rate of two tonnes per year in the reference case, and four tonnes per year in an alternative study case. These options would render the plutonium inaccessible to diversion through the characteristics of spent MOX fuel (such as high radiation fields), and at the same time generate electricity. A longer-term option considered was plutonium annihilation in CANDU, mixing the plutonium in a non-fertile matrix material (hence, destroying the ex-weapons plutonium without producing new plutonium).

The study concluded that the main objective, a plutonium disposition rate of two tonnes per year, could be achieved by burning the plutonium as MOX fuel in two Bruce A reactors. The Bruce reactor site contains eight reactors, four Bruce A reactors, and four Bruce B reactors, each about 769 MW(e). The Bruce generating station, located on Lake Huron, about 300 km northeast of Detroit, is particularly suited for this mission, because of its base load operating mode, its proximity to the U.S. border, and its existing safeguards and security infrastructure. The Bruce A units have further neutronic advantages for accommodating plutonium, which will be discussed later in the paper.

A primary objective in the reference case was a MOX fuel design that would allow the reactor to operate within its current licensing envelope. The standard 37-element bundle design was chosen for the reference case, with depleted uranium as the matrix material. The outer two rings of elements contained plutonium, while the central seven elements in the bundle contained a burnable neutron absorber (dysprosium). The neutron absorber allowed a greater amount of plutonium to be loaded into the bundle than would otherwise have been possible, by suppressing the extra reactivity, and resulted in the coolant void reactivity coefficient being negative. The alternative case, of dispositioning four tonnes of plutonium per year, clearly could be achieved by burning the MOX reference fuel in four Bruce A reactors rather than two. However, the economics favour using the same size of fuel-fabrication facility as in the reference case, and not increasing significantly the MOX fuel-fabrication rate. This was achieved by using the higher-burnup 43-element CANFLEX design [1] for the alternative case, and increasing the plutonium content of the bundle, and hence the fuel burnup.

The study concluded that the reference MOX fuel design could be used without any changes to the reactor core. The main engineering change required to the plant would be to enhance the physical security for the MOX fuel; specifically, a new, secure building would be required for the storage of the fresh MOX fuel, and the route to the new fuel loading room would require hardening.

The philosophy in MOX fuel fabrication was to manufacture the MOX fuel close to the source of the plutonium, and to transport finished MOX bundles to the Bruce site. The study found a significant economic advantage and shorter implementation schedule by using an existing facility and infrastructure, rather than constructing a new facility. The simplicity of the CANDU fuel-bundle design facilitates MOX fuel fabrication. The fuel-fabrication requirements in both the reference and advanced MOX fuel cases could be met by modifying the existing Fuel and Materials Examination Facility (FMEF) at the U.S. DOE Hanford reservation (with a CANDU MOX fuel fabrication capacity of about 170 metric tonnes of heavy metal per year). A lead time of about four years would be required for conversion, licensing, and testing of the facility. A subsequent study determined that the existing, unused Barnwell Nuclear Fuel Plant, which is located adjacent to DOE's Savannah River facility, would also be suitable.

One month's supply of fuel in the reference case will be about 754 bundles, for two Bruce A reactors. It is proposed that seven CANDU MOX bundles (each with its own packaging/shielding material) would be packaged in a standard, stainless-steel, 55-gallon (US) drum. The drums would be loaded into a DOE Safe Secure Transport (SST) vehicle, which has the ability to carry 48 drums on pallets. Three such SSTs would be loaded and travel in a convoy to the Bruce site. Hence, one month's fuel supply (for two Bruce A reactors) can be moved in one convoy. The fabrication, transport, and utilization at the Bruce site of the CANDU MOX fuel would comply with all national and international safeguards and security regulations. The study assumed that the spent MOX fuel would be stored at the Bruce site in wet and/or dry interim storage, and would then be transported to permanent disposal at either a U.S. or a Canadian geological repository.

The focus of the remainder of this paper is on the fuel design and performance, and the reactor physics results of the study.

MOX Fuel Design

The high neutron economy of the CANDU reactor and fuel design, required for using natural-uranium fuel, facilitates the use of other fuels [2]. While this paper focuses on the use of military plutonium as MOX fuel in CANDU, the CANDU reactor is an ideal machine for deriving the maximum energy potential from spent PWR fuel [3,4,5].

The simple CANDU fuel bundle design not only simplifies MOX fuel fabrication, but provides a large degree of flexibility in terms of the fuel composition across the bundle. Some considerations in optimizing the reference MOX fuel design for plutonium dispositioning were as follows:

- Maximize the amount of plutonium in the bundle (to minimize the number of bundles required), without needing to hold down additional excess reactivity in the core. A neutron absorber was added to the bundle, to accommodate a larger amount of plutonium, and to obviate the need to add additional dissolved poison in the moderator. The use of depleted uranium as the matrix material also necessitated a higher plutonium content in the bundle.

- Achieve acceptable reactivity coefficients. The addition of a burnable poison to the central elements made the coolant void reactivity negative. This offsets the effects of both a smaller delayed neutron fraction and neutron lifetime with MOX fuel. Accident consequences are similar or are more benign than with natural-uranium fuel.

- Minimize the peak linear element ratings over the fuel burnup. Enrichment grading was chosen for the outer two rings of fuel containing plutonium, to reduce ratings. The choice of enrichments also was near-optimal, from the thermalhydraulics consideration of critical heat flux.

- Maintain the existing fuel power/burnup envelopes for the reference case. Average discharge burnup in the reference MOX fuel case was only slightly higher than natural uranium, and peak

element burnup was about the same as for natural uranium. Also, the fuel-management strategy (regular bi-directional two-bundle shift) helped reduce peak bundle powers, and the refuelling ripple.

- Minimize the implementation time. The reference MOX fuel design has a high degree of "provenness", employing the standard 37-element bundle and a power/burnup envelope within that of natural uranium. Hence, the time required to verify the fuel design and performance for MOX fuel, and to licence the reactor, will be minimized.

The reference fuel design has depleted uranium throughout (0.2% U-235), with 5% dysprosium in the central 7 elements (the central element, and the next ring of 6 elements); 2.0% plutonium in the third ring of 12 elements, and 1.2% plutonium in the outer ring of 18 elements. The bundle average burnup of the reference MOX fuel is 9700 MWd/te heavy element (HE), compared to 8300 MWd/te HE for natural uranium fuel in Bruce A. Peak element burnup is about the same as for natural uranium (about 16 000 MWd/te HE). The fresh reference MOX fuel design contains 232 g plutonium per bundle, of which 94% is fissile.

The advanced MOX fuel design utilizes the 43-element CANFLEX geometry, which features two element diameters, arranged in rings of 1, 7, 14 and 21 elements. The central 8 elements are larger than the outer 35 elements. The greater subdivision in the bundle, along with two element sizes, reduces the peak element rating by about 20%, compared to the 37-element bundle operating at the same bundle power. This lower element rating facilitates the achievement of extended burnup in CANDU, by lowering the fuel temperature and fission-gas release. The core-average burnup of the advanced MOX design was chosen to be 17 100 MWd/te HE, which results in a peak burnup of under 30 000 MWd/te HE. These are burnups for which we have some experience. The advanced MOX bundle contains 374 g plutonium in the fresh fuel. As in the reference bundle, the plutonium is confined to the outer two rings of fuel: 3.5% plutonium in ring 3, and 2.1% in ring 4, mixed with depleted uranium. The central 8 elements contain 6% dysprosium mixed with depleted uranium. There is some minor optimization of the internal element design (pellet size and shape, and clearances).

In both the reference and advanced MOX fuel designs, coolant void reactivity is negative, about -4.7 mk and -1.7 mk, respectively, compared to about + 11 mk for natural uranium. This number refers to the change in reactivity that would accompany a hypothetical, instantaneous voiding of all the coolant in the reactor core.

Each Bruce A reactor would consume about one tonne of plutonium per year, in both the reference and alternative cases. In the reference 37-element MOX fuel design, the plutonium content in the spent fuel is 154 g, while in the advanced MOX fuel the plutonium content in the spent fuel is 254 g. In both cases, the initial plutonium content is reduced by about one third in the spent fuel. (Keep in mind that the objective of this strategy is not to *destroy* the plutonium, but to convert it to a form that has a high degree of diversion resistance through the characteristics of spent fuel, while producing electricity.)

CANDU Reactor Physics With MOX Fuel

The use of MOX fuel is facilitated in the Bruce A reactor through two features of the core design. First, Bruce A has no adjuster rods. Adjuster rods are used in some CANDU reactors, primarily to provide a certain xenon override capability following a reactor shutdown. The adjuster rods also flatten the flux in the center of the core. With enriched fuel, a suitable axial power profile can be obtained through use of a simple axial fuel-management scheme, so the adjuster rods are not needed for shaping the axial power distribution. Second, the Bruce A station plans to change the fuelling direction to that of the coolant flow. This will increase the margin to dryout with the MOX fuel.

The on-power refuelling of CANDU not only enables the reactor to be operated with only a small amount of excess reactivity in the core, but also provides a great deal of flexibility in fuel management, because

of the ability to shape the power distributions through the core, both axially and radially. With both the reference and advanced MOX fuel designs, a very simple, bi-directional (adjacent channels are refuelled in the opposite direction), two-bundle shift fuelling scheme in the direction of coolant flow results in an excellent axial power distribution. The power peaks around axial bundle position 4, and decreases along the length of the channel. Hence, the bundles at the downstream end of the channel have the lowest power, which increases the margin to dryout. This fuelling scheme causes only relatively fresh fuel to experience a power boost as a result of refuelling. This simple fuel-management strategy results in good axial flattening of the axial power distribution, with the peak bundle power being about 20% lower than with natural-uranium fuel.

Radially, the distribution of burnup through the core was chosen to give a similar channel power distribution to natural uranium.

The core design with MOX fuel was based on the lattice code WIMS-AECL [6], with the two-dimensional finite-difference reactor code RFSP [7]. A 100-day time-dependent refuelling simulation was performed for the reference MOX core, in which the refuelling of individual channels was modelled. The maximum element, bundle and channel powers, power/burnup envelopes, power-boost envelopes, and refuelling ripples were all below the corresponding values experienced for Bruce A with natural uranium fuel. Table 1 compares the characteristics of the natural uranium, reference MOX, and advanced MOX cores. The burnup with the advanced MOX fuel design of course extends past that normally experienced in Bruce A. However, the power/burnup envelope for the advanced MOX fuel is within that for experimental fuel irradiations in the NRU research reactor at AECL's Chalk River Laboratories.

The reactivity worths of the reactivity devices (liquid zone controllers, mechanical control absorbers, and shutoff rods) are lower with MOX fuel than with natural uranium. However, the worths are adequate for reactivity control and shutdown. The reactor control system was modelled for MOX fuel, and no hardware changes are required (a modest change to the primary control feedback gain may be required.)

CANDU MOX Fuel Performance

Figure 1 shows a typical snapshot of peak linear element ratings and corresponding element burnup for the reference MOX fuel, for an arbitrary point in time from the refuelling simulation. Each point in the plot corresponds to the peak element rating and corresponding element burnup for a bundle in the core. Superimposed on the snapshot is the natural-uranium high-power envelope, for Bruce A. The power envelope for the reference MOX fuel is considerably lower than for natural uranium, and is well below known failure thresholds.

Further confirmation of fuel performance was provided by modelling the fuel behaviour for the high-power envelope, for both the reference and advanced MOX fuel designs, and for natural uranium. The ELESTRES [8] code was used for this modelling, with a focus on fission-gas release, internal pressure, and ridge strains. In ELESTRES, a single element is modelled by accounting for the radial and axial variations in stresses and displacements. Predictions of total fission gas released to the "free inventory", maximum internal gas pressure, and sheath strains for both the reference and advanced MOX fuel designs were well below the values corresponding to the natural-uranium high-power envelope. This is due to the lower peak element ratings for the MOX fuel designs, which reduce fission-gas release, and the optimized internal design of the MOX fuel elements, which reduces internal pressure and sheath strains.

The thermalhydraulic performance of the MOX fuel was assessed using the ASSERT [9] subchannel code, and the NUCIRC [10,11] steady-state-system thermalhydraulics code. ASSERT provided critical heat flux (CHF) data for the MOX bundles, while NUCIRC provided the critical channel power (CCP), the channel power at which CHF first occurs on any fuel element in the reactor. The ASSERT calculations (at constant flow) indicated that the steep radial power profile through the MOX bundles reduced CHF, while the axial power profile, skewed towards the inlet end, increased CHF. The net result for the reference MOX fuel was a slight increase in both the CHF, as well as in the pressure drop along the channel. To

Snapshot of Intermediate- and Outer-Element Powers and Burnups for Reference MOX Fuel in Bruce A

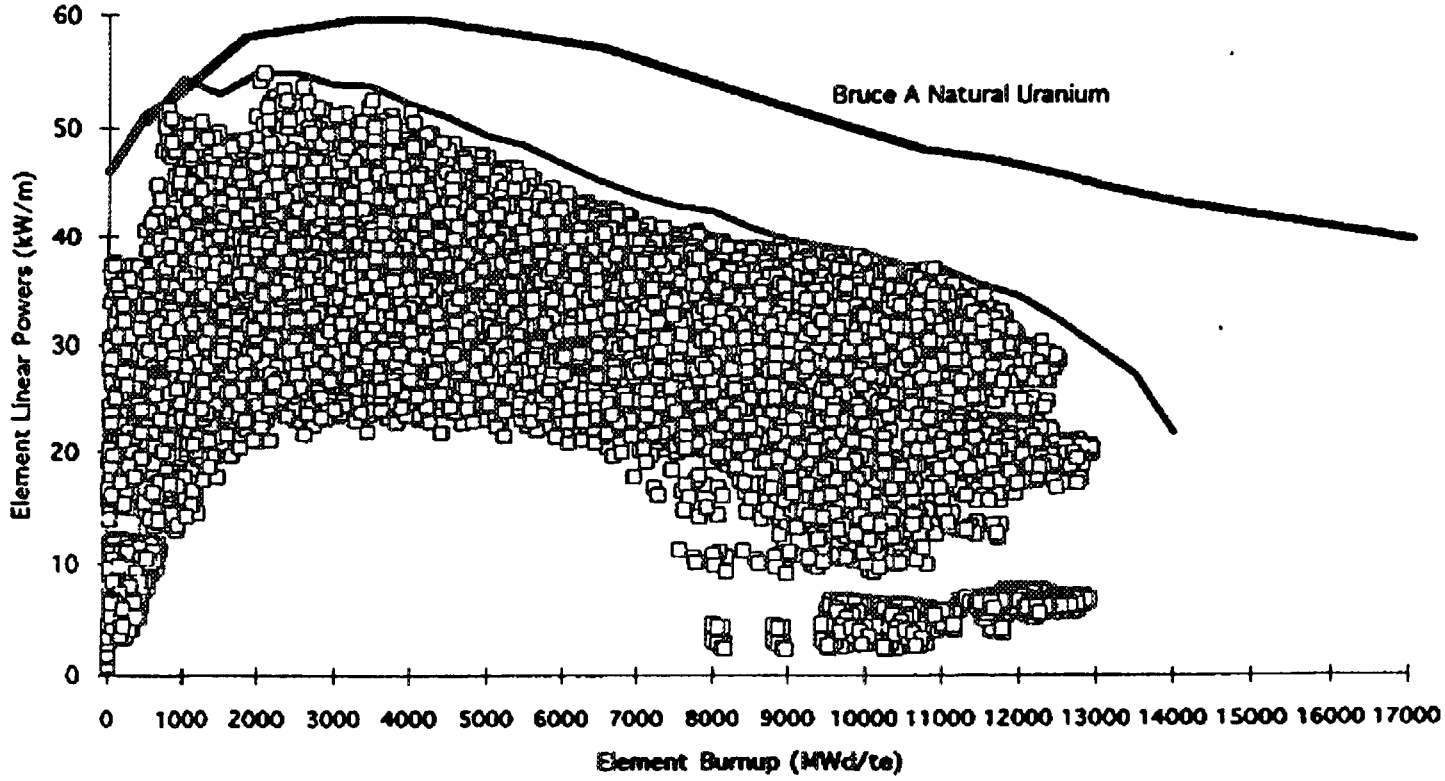

FIGURE 1.

calculate the effect on CCP, the effect of the increase in both CHF and pressure drop on the dryout power at constant header-to-header pressure drop was determined using NUCIRC. The same dryout power was predicted for the reference MOX and natural-uranium cases, within the uncertainty of the calculation. For the CANFLEX bundle with MOX fuel, ASSERT predicted slightly lower CHF and pressure drop (because of the larger flow area in the CANFLEX bundle compared to the 37-element bundle). NUCIRC predicted a slightly greater (2%) CCP for the CANFLEX MOX fuel compared to the 37-element natural-uranium reference case. It is noted that this calculation did not include the CHF-enhancement features of the CANFLEX bundle, which are expected to increase CCP by 6-8% for natural uranium. Thus, it is expected that the advanced MOX bundle will have a CCP several percent higher than the existing 37-element natural-uranium bundle.

Safety and Licensing with MOX Fuel

Table 1 includes a comparison of the neutron kinetic parameters of MOX and natural-uranium fuel. With both a smaller delayed neutron fraction and prompt neutron lifetime, the response of MOX fuel to a reactivity change is faster than with natural-uranium fuel. This faster response is compensated by the negative coolant void reactivity designed in the MOX fuel bundle.

A systematic investigation of all the design-basis accidents was made for the MOX fuel, with emphasis on those accidents that rely on a neutronic trip. For some design-basis accidents, typically loss-of-coolant-accidents (LOCA) and loss-of-flow events, neutronic trips would not occur, due to the negative coolant void reactivity in the MOX core. However, it was found that existing process trip parameters will provide effective protection for these events.

In the particular case of a large-break LOCA, with natural-uranium fuel the reactor trips quickly (in less than 0.5 s) with one of the two, independent shutdown systems. With MOX fuel, both shutdown systems will trip the reactor on either heat-transport-system low pressure or low flow. The total energy deposited in the fuel five seconds after LOCA initiation will be lower for the MOX fuel than for natural-uranium fuel, indicating that fuel heatup will be reduced and that existing safety design objectives would be met.

The response to other design-basis accidents was either similar or better than for natural-uranium fuel.

Transition to MOX Fuel

The on-power refuelling of CANDU would enable the reference MOX fuel to be introduced to the natural-uranium core during the normal course of refuelling. The similarity of burnups between natural uranium and the reference MOX fuel, and the two-bundle shift fuelling scheme with the MOX fuel, would reduce the reactivity perturbations during refuelling. Natural uranium would be replaced by MOX, two bundles at a time. The fuelling rate is 15.5 MOX fuel bundles per full-power day (FPD), and there are 6240 fuel bundles in the core. Hence, it would take at least 400 FPDs to displace all the natural-uranium fuel bundles with MOX. (The fact that channels are refuelled at different rates means that it will take longer to convert to a full MOX core; it is expected to take at least 600 FPDs to reach the equilibrium MOX core condition.) The absence of adjuster rods in Bruce A would also be an advantage during the transition. The same procedure could be used to replace the reference MOX fuel with the CANFLEX MOX fuel bundles at a later stage, if desired. This approach would enable the earliest start to the transition, and would derive the maximum energy from the MOX fuel.

Another strategy for the transition would be to start up after a major scheduled outage with a full MOX core.

The transition from natural uranium to MOX fuel has not been modelled during this study, and would be optimized as part of the implementation program. On-power refuelling again provides flexibility in shaping the axial power profile during the transition, should that be required.

Plutonium Annihilation In CANDU

A longer-term option for the near complete destruction or annihilation of the plutonium is to burn the plutonium in an inert matrix (rather than in a fertile uranium matrix) in CANDU [12-14]. The absence of uranium-238 eliminates the source of further creation of plutonium and over 40% of the neutron absorption in the lattice, resulting in a remarkable improvement in neutron economy. The absence of the neutron absorption reduces the fissile requirement of the plutonium annihilator relative to the natural-uranium-fuelled core. A lower fissile inventory requires a correspondingly higher operating neutron flux level, to produce the rated power, and makes CANDU superior to other reactor types (including fast breeder reactors) in the annihilation process. This superiority is evident from the high (>80%) annihilation rate that is achievable per pass through the reactor. By using the on-power fuelling machines to "shuffle" the fuel through one or two additional passes, essentially all of the plutonium may be annihilated. While this option would require substantially more development than the MOX option, it resolves the question of ultimate disposal without requiring highly advanced technology or reprocessing.

AECL is currently investigating the suitability of various candidate inert matrix materials, with emphasis on SiC.

Status of Technology

There is now significant favourable world-wide experience with the fabrication and irradiation performance of MOX fuel. AECL has over 25 years of experience with MOX fuel. Irradiation testing in the NRU research reactor and post irradiation examinations are still being conducted as part of AECL's advanced fuel-cycle program. Rehabilitation of the Recycle Fuel Fabrication Laboratory (RFFL), a series of glove boxes for the remote fabrication of alpha-active fuel, is currently underway at Chalk River Laboratories. A companion paper in this conference summarizes AECL's experience and current programs with MOX fuel [15].

The CANFLEX (CANDU Flexible Fuelling) program, upon which the advanced MOX fuel design is based, is nearing completion [1]. This program was started by AECL in 1986, and since 1991, the Korean Atomic Energy Research Institute (KAERI) has been a partner in the program. The program is aimed at demonstrating the CANFLEX bundle to the point that would enable a power reactor demonstration in the next couple of years. The major milestones over the next year are:

- fabrication of 50 natural-uranium CANFLEX bundles for flow, endurance and handling tests, and for reactor physics measurements,
- ZED-2 reactor physics measurements with 35 CANFLEX bundles with natural-uranium fuel,
- completion of licensing-rigour CHF tests in Freon, to demonstrate the improvement in CCP over the 37-element bundle,
- completion of fuel-handling tests and fuelling-machine-compatibility tests,
- initiation of flow and endurance tests, and
- continuation of NRU irradiation of CANFLEX bundles to burnups beyond natural uranium.

Hence, the timing of the CANFLEX program is consistent with its availability for plutonium dispositioning.

Another advanced fuel program underway in AECL that would provide support to the plutonium dispositioning program is low void reactivity fuel (LVRF) [16]. This fuel design is similar to the MOX fuel designs used in the plutonium dispositioning study, but employs enriched uranium rather than MOX fuel in the outer two rings of elements. By varying the level of burnable poison in the center of the bundle, and the enrichment in the outer two rings, the level of void reactivity and fuel discharge burnup can be tailored to meet customer requirements. The concept was conceived for those jurisdictions in which reduced or negative void reactivity is required. A short-term demonstration program is currently underway to establish the technical feasibility of the LVRF design, in both the 37-element and CANFLEX

geometries. The program includes NRU prototype irradiations, reactor physics testing in the ZED-2 reactor, and measurements of CHF in Freon. Prototype elements containing dysprosium have already been irradiated and examined, and the expected good fuel performance has so far been confirmed. This program provides a solid technology base for the MOX fuel designs for plutonium dispositioning.

Deployment Strategies

The CANFLEX design would require fabrication of roughly half the number of MOX fuel bundles to achieve a certain plutonium disposition rate, compared to the 37-element design. Hence, with a given size MOX fuel fabrication plant, CANFLEX could be used to either double the amount of plutonium that could be dispositioned in a given time, or to halve the time required to dispose of a given amount of plutonium.

One of the attractive features of the CANDU plutonium dispositioning option is the symmetry of a CANDU reactor in Canada burning military plutonium from both the U.S. and from Russia. Conversion of the Russian military plutonium to CANDU MOX fuel could take place in Russia, and would involve considerable Russian nuclear technology, resources and labour. Shipment of CANDU MOX fuel containing only about 2% plutonium to Canada would be better, from a safeguards and security viewpoint, than would shipment of relatively pure plutonium. Alternatively, a CANDU reactor in Russia or Eastern Europe could burn the MOX fuel fabricated in Russia from weapons plutonium. This option would provide energy value from the plutonium, an important objective of Russian policy.

Fabrication of CANDU MOX fuel from military plutonium is the subject of a similar joint study being planned by the Russian and Canadian Governments.

Finally, the plutonium annihilation option in CANDU offers the possibility of a longer-term solution to the ultimate destruction of plutonium, in parallel with an immediate short-term solution to dispositioning of military plutonium.

Table 1: Comparison of Core Characteristics with Natural Uranium and MOX Fuel

	Natural Uranium	MOX, 37-element Reference	MOX, CANFLEX
Average burnup (MWd/te HE)	8300	9700	17 100
Maximum burnup (MWd/te HE)	15 000	15 500	28 000
Bundles / full power day / reactor	18	15.5	9
Bundles per channel refuelled	2, 4 or 8	2	2
Maximum channel power (kW)	7200	7000	7000
Maximum bundle power (kW)	960	780	800
Full core void reactivity (mk)	+11	-4.7	-1.7
Fuel temperature coefficient (micro-k/degree C)	-6.0	-3.0	-2.0
Total delayed neutron fraction	0.00582	0.00383	0.00369
Prompt neutron lifetime (s)	0.0009	0.0005	0.00046

Summary

This study performed for the U.S. DOE has identified practical and safe options for the dispositioning of military plutonium in existing CANDU reactors. By careful fuel design, the fuel and nuclear characteristics will be within existing envelopes for fuel performance, safety and licensing. Utilization of existing fuel fabrication and transportation facilities and methods has resulted in a low-cost, low-risk method for long-term plutonium dispositioning. The integrated system can be ready to begin plutonium consumption in four years. No changes are required to the existing reactor system, other than for provision of safe and secure storage of new fuel. An annihilation option that uses the unique features of the CANDU system to achieve high levels of destruction of plutonium offers an attractive option for ultimate disposition without requiring reprocessing of spent fuel or advanced technology.

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SESSION 5
FUEL PERFORMANCE

PRACTICES AND TRENDS IN MOX FUEL LICENSING IN FRANCE

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Abstract

The French programme aiming at recycling plutonium in pressurized water reactors (PWRs) was initiated in 1985 by Electricite de France (EDF) in close cooperation with the Commission for atomic energy (CEA), Cogema and Fragema. At the request of the French safety authority (DSIN), the programme proposed by EDF was submitted in 1986 to a preliminary safety examination, concerning the different stages of the plutonium and uranium mixed oxides (MOX) fuel cycle: manufacturing, transport, use in reactors, reprocessing and wastes management. The first load of MOX fuel was introduced in the reactor of Saint-Laurent B1 in 1987. So far, seven 900 MWe PWRs have already been loaded with MOX fuel and operated under specific conditions, dedicated to ensure the safety of their operation.

1. CURRENT PROGRAMME OF PLUTONIUM RECYCLING IN THE 900 MWE PWRS

At the present time, the seven reactors, which have already been loaded with MOX fuel, are the following: Saint-Laurent B1 and B2, Gravelines 3 and 4, Dampierre 1 and 2 and Blayais 2.

The MOX fuel loaded in these reactors consists of a mix of plutonium and uranium oxides, at the rate of 5.3 percent of Pu02 for 94.7 percent of U02 in assembly average. Actually, the MOX fuel assembly is made of fuel rods whose plutonium concentration varies from the center of the assembly to the periphery (3 zones from 6.75 to 3.35 percent of Pu02). Thus, this assembly is the neutronic equivalent of the standard 3.25 percent U235 enriched assembly, in terms of power and irradiation time. The structure of the MOX fuel assemblies is also similar to the one of the standard Fragema U02 fuel assemblies.

At the present time, MOX fuel assemblies are manufactured partly by Cogema in the CFCa plant (Cadarache, France) and partly by Belgonucleaire (Dessel, Belgium). In August 1994, the safety authority authorized Cogema to put in operation the Melox plant located in Marcoule (France). This new plant is dedicated to manufacture MOX fuel rods and assemblies. At present, the qualification of the manufacturing process is in progress.

In the core of the 900 MWe PWRs loaded with MOX fuel, there are 48 MOX fuel assemblies, coming from three reloads of 16 assemblies each. Each MOX fuel assembly is irradiated during 3 annual cycles. The remainder of the core consists in 109 U02 fuel assemblies : 3.7 percent U235 enriched assemblies for reactors under "hybrid" refueling or 3.25 percent U235 enriched assemblies under "third-core" refueling. Thus, the plutonium recycling rate of so-loaded reactors, i.e. the ratio between the number of MOX fuel assemblies and the total number of assemblies in the core, is 30 percent.

2. SAFETY REQUIREMENTS RELATED TO THE USE OF MOX FUEL IN PWRS

A preliminary safety examination of the programme proposed by EDF was carried out in 1986. The French safety authority, assisted by the Institute for nuclear protection and safety (IPSN) within the

CEA and the standing group for nuclear reactors ("Groupe permanent reacteurs" or GPR), authorized in 1987 the recycling of plutonium in the 900 MWe PWRs under the following conditions which were judged necessary to ensure the safety of the handling and the storage of MOX fuel and the safety of the operation of reactors loaded with such fuel.

Fuel handling and storage

Fresh fuel assemblies containing plutonium have a higher dose rate as the one of standard fuel assemblies, due to neutrons and gamma rays. This entails special radiological protection measures during handling. In-plant handling requires the use of a dedicated examination ring.

For radiological protection reasons as well, it is better not to store MOX fuel assemblies, as plutonium progressively turns into americium. Experience shows that consequences on the composition of MOX reloads are acceptable. However, the number of MOX loaded reactors in operation exceeds the strictly necessary one, according to the capacity of production. Used MOX fuel requires a longer cooling-down period than U02 fuel, before it may be transported.

Reactor operation

At the request of the French safety authority, EDF examined the consequences of the presence of MOX fuel on the behaviour of the core in normal operation, as well as in accidental situations. Adjustments and limitations were required, in order to ensure that the safety criteria, which were applied to U02 fuel, would be fulfilled with MOX fuel. These requirements were grounded on the neutronic properties of MOX fuel and the available knowledge on its behaviour in reactor :

The higher neutron absorption of MOX fuel requires a greater neutron flux, in order to generate the same energy quantity; thus, in the MOX fuel assembly, the concentration of plutonium decreases from the center to the periphery (6.75 percent to 3.35 percent), in order to avoid the local power peaks which would, otherwise, appear at the interfaces between MOX fuel assemblies and U02 fuel assemblies.

The higher temperature coefficients (moderator and Doppler), in absolute value, induce a lower worth of the neutron absorbers; in other respects, the lower proportion of delayed neutrons has some effects on the reactor kinetics, in particular during accidents resulting in a reactivity excursion in the core; an increase of the control rod number (4 additional control rods) and a slight increase of the boron concentration make up for these characteristics.

On the basis of the neutronic analyses and experiments carried out in the past, the proportion of MOX fuel in the core has been limited to 30 percent.

Further limitations were required at the beginning of the programme: restriction to operation at constant power and limitation of the irradiation to 3 annual cycles (around 36000 MWday/t), in bundle average; these wise provisions were motivated by the insufficient guarantees about the reliability of MOX fuel behaviour under load-follow conditions and at high burn-up levels.

3. LICENSING ISSUES RELATED TO THE USE OF MOX FUEL IN PWRS

Licensing procedures

In France, the use of MOX fuel in the 900 MWe PWRs is submitted to special licensing procedures. MOX fuel can be used in reactor only under the following conditions:

- The construction license decree, signed by the Prime Minister and countersigned by the ministers in charge of the environment and industry must make provision for the use of MOX fuel. This decree defines the perimeter of the nuclear plant and the requirements which must be met by the operator,

- The use of MOX fuel is subject to the approval of the ministers in charge of industry and environment, after advice of the minister in charge of health. The DSIN, acting on the authority of the ministers in charge of environment and industry, authorizes the delivery and the storage within the perimeter of the concerned nuclear installation of each reload of MOX fuel assemblies, its introduction in the reactor as well as the operation of the so-loaded reactor.

In other respects, on the occasion of these authorizations issued for each MOX fuel reload, the operator is asked to bring to the notice of the DSIN and the Office for protection against ionizing radiations (OPRI) which assists the ministry in charge of health, a quarterly evaluation including, among others results, those about the production of wastes and the activity levels of alpha emitters in the reactor coolant.

At the present time, sixteen 900 MWe PWRs have a construction license decree which provides for the use of MOX fuel: Blayais 1 and 2, Dampierre 1 to 4, Gravelines 1 to 4, Saint-Laurent B1 and B2 and Tricastin 1 to 4.

Examination of the licensing requests

The main bodies involved in the examination of the licensing requests of the French operator, aiming at using MOX fuel in one of its 900 MWe PWRs, or more generally, aiming at modifying the conditions of such use, are the following :

- The DSIN examines the licensing requests of EDF, with respect to technical safety. It is assisted by the IPSN which carries out the technical safety analyses of the provisions made by the operator.

- The service for the radiological protection ("Bureau de la radioprotection") within the ministry in charge of health examines these requests, with respect to radiological protection. It is assisted by the OPRI.

Information

Since the beginning of the programme, the French safety authority has made available information to the public on safety problems related to the recycling of plutonium in the 900 PWRs. The high council for nuclear safety and information (CSSIN) has played a major role in the process of informing the public; this council issued recommendations on the subject in 1989.

4. SHORT TERM AND MID TERM PROSPECTS

Experience of plutonium recycling

Since 1987, experience has been gained far more than 20 reactor.years : around 350 MOX fuel assemblies have been loaded, of which around 150 have been unloaded. The results of this experience are now available.

The behaviour of MOX fuel irradiated in reactor seems similar to the one of U02 fuel. Up to now, burn-up reached by MOX assemblies has averaged 38000 Mwday/t. The fission gas release within the rod is greater with MOX fuel than with U02 fuel but internal pressure is kept within the specified limits, thanks to the physical characteristics of the MOX pellet.
Technical specifications for reactor coolant activity are the same as those applied to U02 assemblies. So far, only one MOX rod showed a slight leaktightness fault below the threshold for unloading set by the French operator. Having been reloaded and closely observed, its behaviour appeared identical to the one of U02 assemblies having a similar defect. So far, no alpha contamination has been observed in the reactors using MOX fuel.

Reinforced by this experience, the French operator intends to pursue and extend its recycling policy. Thus, EDF has demanded the authorization of the DSIN on several purposes. The requests, which have been recently examined by the safety authority, deal mainly with the extension of the use of MOX fuel to further reactors and modifications of the refueling and operation conditions of the reactors loaded with such fuel.

Starting of Melox production and extension of the use of MOX fuel to further reactors

Melox plant shall be put in industrial operation in 1996. The production capacity will be 115 tons per year (weight of MOX fuel contained in the fuel elements). The plant will be able to supply MOX assemblies for 20 reloads of 900 MWe reactors per year, thus balancing the flow of plutonium produced by reprocessing and the one entering the production of MOX fuel assemblies.

EDF intends to increase the plutonium recycling capacity of its reactors, in accordance with the evolution of MOX assemblies production capacity:

- EDF intends to burn MOX fuel progressively in its sixteen reactors whose construction license decree provides for the use of MOX fuel ; the approval of the DSIN will be required for each reactor concerned ; to date, EDF plans to extend the use of MOX fuel to nine reactors in 1996, fourteen reactors in 1997 and sixteen reactors in 1998.

- EDF intends to prepare progressively the licensing files for further reactors ; besides the sixteen reactors already licensed to receive MOX fuel, twelve additional 900 MWe reactors are technically designed to do so; as the use of MOX fuel is not mentioned in the construction license decree of these reactors, the licensing procedure will require to resume the initial decree and to obtain a ministerial approval; in particular, a public inquiry will have to be carried out, as part of the licensing procedure; the reactors of Chinon B are the first to be examined through this procedure.

Use of a new MOX fuel assembly design

The French safety authority is at present examining the request of EDF concerning the use a new MOX fuel assembly design. The acquired experience has led to advances in the design of fuel assemblies. For MOX fuel, the resulting assembly is the AFA 2G MOX fuel assembly designed by Fragema, which includes the improvements applied to the current AFA 2G U02 fuel assembly: mainly an optimized Zircaloy 4 cladding with enhanced corrosion resistance, mixing grids with higher thermohydraulic performance and a device aiming at retaining debris.

"Hybrid" refueling

"Hybrid" refueling consists in differentiating U02 assemblies from MOX ones, in order to optimize the use of U02 fuel : 3.7 percent U235 enriched U02 assemblies are irradiated for four annual cycles whereas MOX fuel assemblies are irradiated for three annual cycles. Having been experimented on two reactors (Dampierre 2 et Gravelines 4), the extension of this type of refueling to the other reactors loaded with MOX assemblies was authorized in August 1994. This fuel management replaces the former "third-core" refueling under which all the fuel assemblies remain three annual cycles in the core.

Operation under load follow conditions

The possibility to remove the initial limitation to operation at constant power has been recently examined. At the request of the French safety authority, EDF has set up an experimental programme, in order to assess the consequences of load follow operation on the behaviour of MOX fuel, with respect to pellet-cladding mechanical interaction loading. According to the results of the power ramp tests which have been carried out in experimental loops, this operation mode does not seem to raise any particular technical problem linked to the presence of MOX fuel. Its behaviour seems similar to the one of U02 fuel. In other

respects, two reactors (Saint Laurent B1 and B2) have been operated under load follow conditions for a probationary period of two annual cycles. The operation of these reactors has not raised any difficulty for the operators. On the basis of the experimental knowledge as well as the operating experience, the French safety authority has recently authorized EDF to generalize the load-follow operation mode to all its MOX loaded reactors (approval in June, 1995).

5. LONG-TERM PROSPECTS

Concerning the burn-up limit applied to MOX fuel, the DSIN judges that it is not able at present to authorize an increase of this limit. This position applies to MOX fuel as well as standard U02 fuel. In order to get the approval for higher limits, EDF will have to assess the behaviour of highly irradiated fuel, under incidental and accidental conditions, more particularly under design-basis accident conditions. Indeed, these conditions have been judged critical to assess the requests of the operator.

In this respect, the CABRI-REP programme has been set up by EDF since 1992, in order to better understand the effects of irradiation on the fuel behaviour during a reactivity initiated accident. The tests, which have been so far carried out on U02 fuel, will be also performed on MOX fuel (2 tests scheduled in 1996 on MOX fuel irradiated during 2 and 3 cycles). Parallel to the R&D programme, EDF was authorized in 1993 to irradiate during 4 annual cycles (up to 46000 MWday/t) some MOX fuel assemblies in the reactor of Gravelines 4, in order to investigate on highly irradiated MOX fuel. In order to ensure the safety during the operation cycle, the corresponding assemblies were not situated in the core at places where the energy deposition in case of a control rod ejection would be maximal. The experimental investigations will provide one part of the safety demonstration, the other part being grounded on theoretical analyses and code simulations.

With the technical support of the IPSN, the French safety authority carries out a thorough analysis of the experimental and operating data related to the behaviour of MOX fuel under normal and accidental conditions. So far, the reliability of the use of MOX fuel in PWRs, under the present refueling and operation conditions, has been confirmed by the experimental results as well as the operating experience.

The French safety authority is open-minded on possible modifications of these conditions, insofar as they are justified through a satisfactory safety demonstration. As it was said previously, the operation under load follow conditions has been recently authorized, on such basis. In the same way, the technical elements, which will be provided by EDF on the behaviour of highly irradiated MOX fuel, will be thoroughly examined by the DSIN, in order to assess the reliability of the use of MOX fuel at high burn-up levels.

VALIDATION OF MOX FUEL THROUGH RECENT BELGONUCLEAIRE INTERNATIONAL PROGRAMMES

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Abstract

The paper reviews the present experience of BELGONUCLEAIRE in promoting and managing international programmes dedicated to improvement and updating of MOX fuel data bases on what concerns core physics and rod behaviour with a view of assist all MOX fuel designers and users in their validation and modelisation work.

All these programmes were completed or will be completed with the support of numerous international organizations deeply concerned by MOX recycling strategies.

1. INTRODUCTION

As MOX fuel manufacturer, BELGONUCLEAIRE is traditionally concerned by the validation, by the performance and by 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 and D work mainly devoted to pending validation and licensing questions of plutonium recycling in commercial reactors.

Such work is since more than 15 years proposed through a set of international programmes, the goal of 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) :

- The first set of programmes which are generally organized jointly by BELGONUCLEAIRE and SCK•CEN are mainly devoted to neutronic validation for reactor physics parameters performed in the clean conditions of the VENUS critical facility located in Mol (Belgium).

Table 1 **MOX International Programmes**

MOX Fuel Behaviour	MOX Fuel Core Physics Data
PRIMO (PWR) CALLISTO (PWR) DOMO (BWR) FIGARO (PWR) NOK-M109 (PWR) NOK-M305/M308 (PWR)*	VIP-PWR VIP-BWR VIPO ARIANE VIPEX*

(*) under preparation.

Such programmes are focused on the experimental determination of MOX rods fission rate distribution, reactivity effects and spectral indices measured in various critical mock up configurations simulating recent designs of PWR on BWR assemblies.

- The second set of programmes concerns complete investigations with respect to MOX fuel behaviour under normal conditions which are obtained through large scale irradiations in various reactors like BR3, DODEWAARD and BEZNAU-1 reactors and under off normal conditions which are simulated in BR2, OSIRIS and HALDEN testing reactors.

  Such programmes are focused on the determination of the MOX fuel rods characteristics for what concerns fission gas release, fuel microstructure, pellet cladding interaction, isotopic vectors, etc., measured after extended irradiation conditions.

This paper summarizes the most significant and recent international programmes which are supported by numerous organizations being interested in collecting recent MOX data.

Emphasis is made on Core physics experimental work already performed through the VIP-PWR, VIP-BWR and VIPO programmes and on MOX rod behaviour data already obtained through the PRIMO and the DOMO programmes.

Targets, parameters as well as some selection of significant validation data for high burnup MOX fuel are summarized (Section 2) to show the large spectrum of investigation already completed.

Furthermore, a description of the new international programmes such as ARIANE, VIPEX, FIGARO and NOK-M109 and NOK-M305/M308 which have started recently or are in their implementation phase will show that major pending questions of interest regarding MOX fuel data base improvements are going to be covered in the very near future, such as:

- Questions on the source term accuracy for high burnup MOX fuel (ARIANE)
- Reactivity parameter evaluation in clean critical MOX configurations (VIPEX).
- Modelization of fission gas release through sophisticated refabrication and instrumented techniques applied to high burnup MOX fuel (FIGARO)
- PCMI and fuel structure analysis of irradiated MOX fuel close to 50 GWd/t (NOK-M109)
- Transient conditions applied to MOX irradiated fuel segments (NOK-M305/M308).

Finally, it will be recalled that such R and D activities call upon the competence and the experience not only of well trained staff in the MOX business but also of numerous European laboratories making use of adequate techniques ensuring the highest scientific level of these international programmes.

2. MOX PROGRAMME EXPERIENCE

2.1. Primo

The PRIMO Programme, which ended last year, covered the irradiation of 15 MOX fuel rods at different burnup stages up to 60 GWd/tM in the BR3 reactor. Two fuel fabrication processes, three cladding types and five fuel rod designs were included in the study.

The power histories were representative of those of large PWR power plants. The programme has demonstrated the ability of MOX fuel rods to sustain irradiation conditions comparable to UO_2 fuel and has allowed the benchmarking of the thermal mechanical codes in case of MOX use. Power ramps have been performed in the BR2 and OSIRIS reactors on selected rods to verify mechanical performance.

Among the numerous obtained results, one has to point out :

- the difference in local plutonium distribution in MOX pellet fabricated according to the Reference or MIMAS process (Figure 1) ;
- the consequences of local Pu concentrations in pellet on fission product build-up and fuel restructuring (Figure 2) ; such concentrations have no influence on the pellet overall behaviour;
- a fission gas release (FGR) from irradiated fuel mainly controlled by the rod power history; neither the fuel type (Reference or MIMAS fabrication process), nor the rod design (fuel cladding gap, plenum volume) have a significant effect on FGR (Figure 3);
- the radial distribution of several fission products and actinide isotopes (Figure 4) providing useful results for neutronic code validation and input data for thermal--mechanical calculations of rod behaviour.

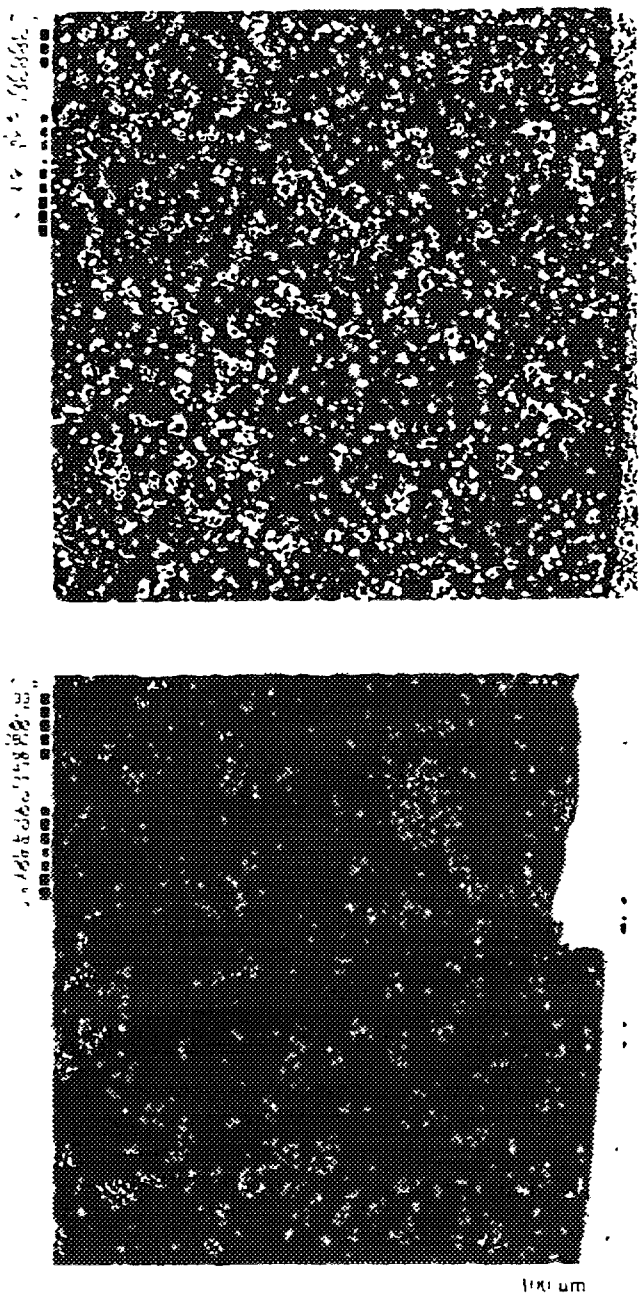

FIG. 1. PRIMO - Plutonium concentration map at periphery of irradiated MOX pellet fabricated following reference (a) and early MIMAS (b) process

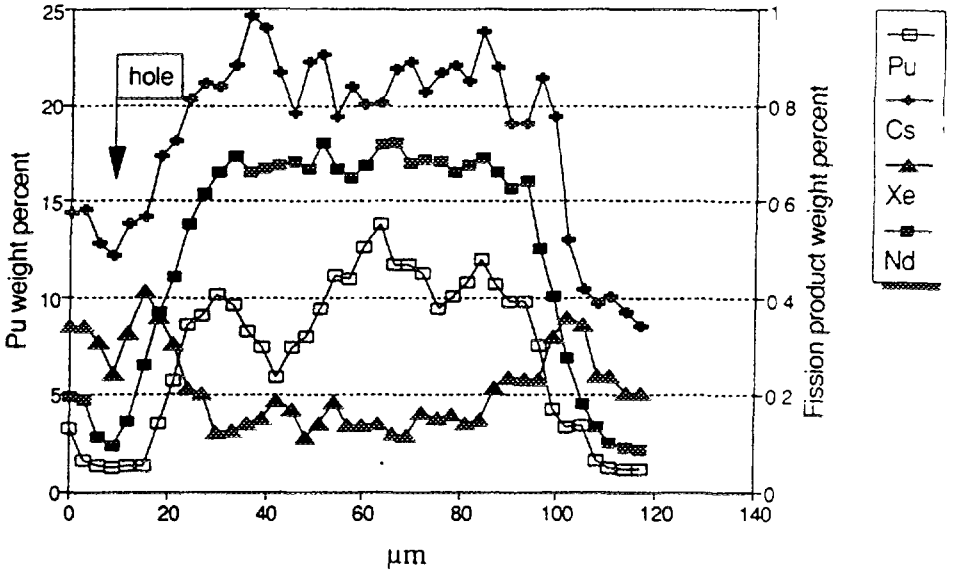

FIG. 2. *PRIMO – Inventory of Pu and fission products in Pu rich zone at periphery of irradiated MOX pellet*

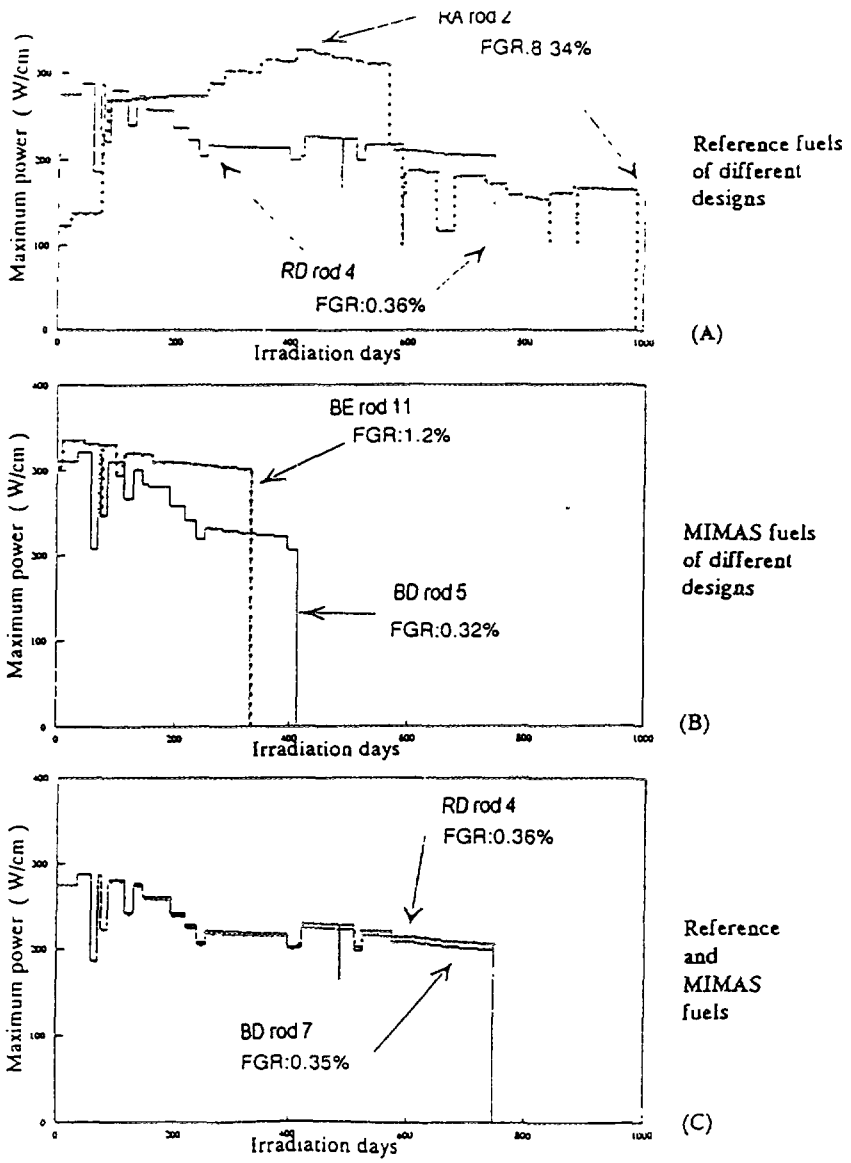

FIG. 3. *PRIMO – Power history and fission gas release*

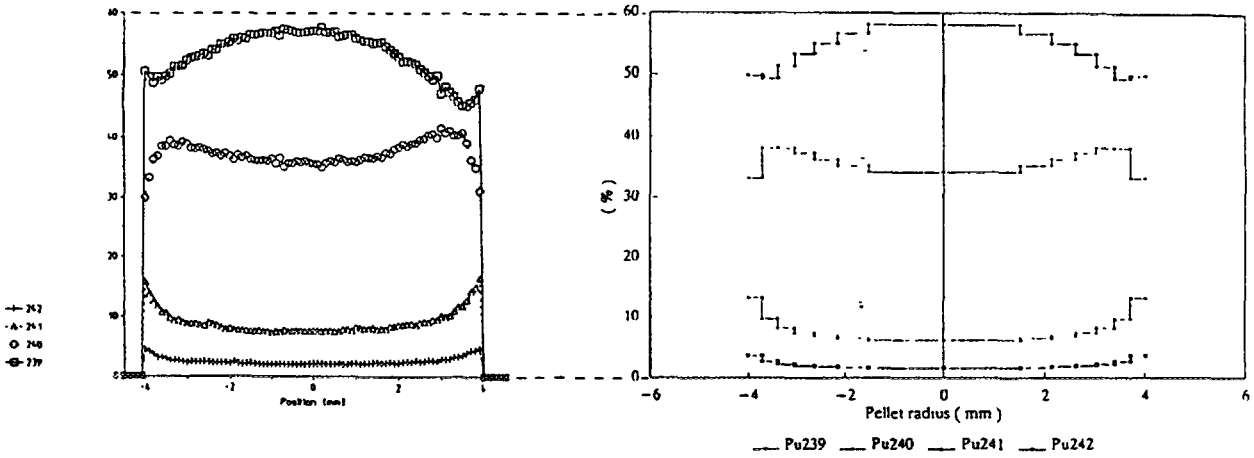

FIG. 4. *PRIMO - Comparison between experimental and calculated local plutonium isotopic composition in MOX fuel irradiated at high burnup*

2.2. Domo

Once again for MOX, but in a BWR commercial reactor environment, we have initiated in 1985 the DOMO Programme (DODEWAARD MOX Programme), which is practically the only programme on MOX in BWR conditions. The main objective of the DOMO Programme is to accumulate thermal--mechanical fuel behaviour data at high burnup (60 Gd/tM target peak pellet) on MOX and UO_2 fuel segments of different fabrication types irradiated under typical BWR conditions. In this programme 10 MOX fuel rods (of four segments each) and 5 UO_2 rods (of four segments each) were irradiated in the DODEWAARD reactor in The Netherlands. After irradiation, power ramps were performed in BR2 on selected segments. The DOMO Programme is due to be completed in 1996.

Segment puncture (Figure 5) after 2 and 4 irradiation cycles confirm the relative insensibility of FGR in regard of MOX fabrication process (PNC or BN) ; the lower FGR for UO_2 in the present experiment is due to its lower rating compared to UO_2.

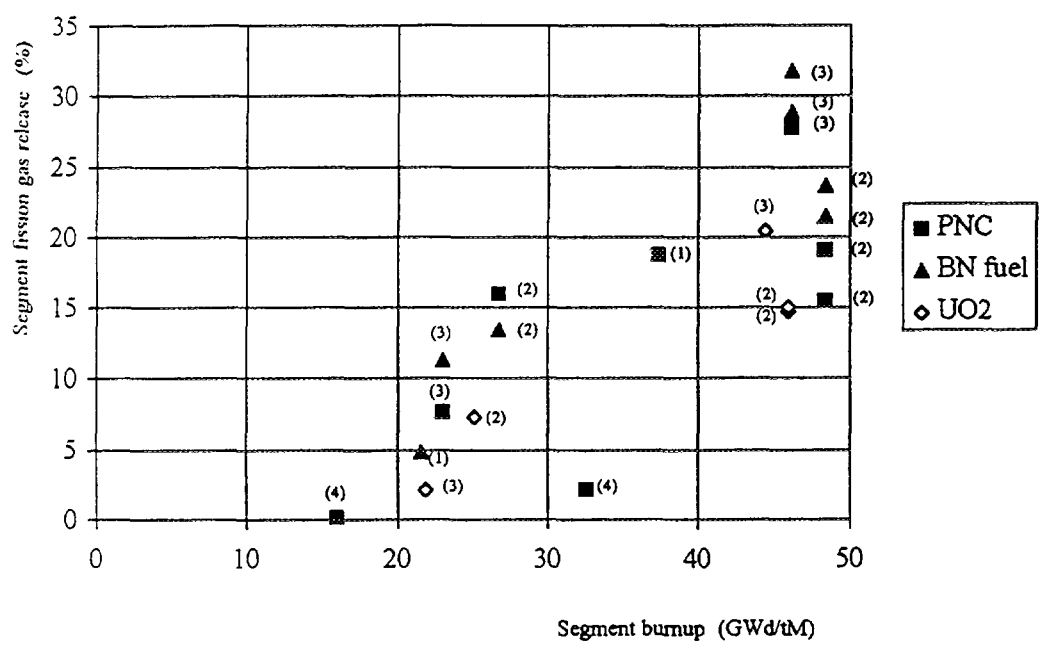

FIG. 5. *DOMO - Evolution of fission gas release versus burnup for PNC and BN MOX fuel, and UO_2 fuel*

2.3. Callisto

With the permanent shut--down of BR3 in 1987, an important irradiation tool was lost and fuel vendors and Japanese utilities expressed their need to continue experimental irradiations. Decisions were immediately taken at BELGONUCLEAIRE and SCK•CEN to design and construct, in close collaboration, a PWR loop to be integrated within the BR2 material testing reactor. This installation offers a comprehensive range of experimental conditions representative of a PWR, as well as a versatile and multipurpose water test facility for testing fuel rod clusters in off normal conditions.

This new loop called CALLISTO (CApabiLity of LWR Irradiation in Steady and Transient Operations) was put in operation in mid 1992 and is today a major asset of the light water reactor fuel development programme at SCK•CEN. In the present configuration, the CALLISTO pressurized water loop has 3 in--pile sections containing up to 9 one meter long fuel rods each and can be considered as a small scale experimental PWR reactor. Individual irradiation programmes containing UO_2 and MOX fuel rods of different fabrication types have been performed with burnups up to about 25--30 GWd/tM. Ramp tests and extensive post-irradiation work are underway to verify the mechanical performance and behaviour of those fuels.

2.4. Figaro

This new programme on MOX fuel has been recently established.

From the experience accumulated so far in Europe on irradiated MOX fuel, and despite its excellent overall behaviour, it appears that data are today still missing to properly benchmark thermal and fission gas release models, mainly at high burnup. Appropriate data are necessary, as it is also the case for UO_2, to reduce the uncertainties in FGR predictions, these uncertainties leading to excessive penalties in fuel operation when approaching the limits of rod design criteria. Moreover, the particular neutronic behaviour and the slightly lower thermal conductivity of MOX fuel compared to UO_2 are leading -- for the same linear power -- to different central fuel temperatures and thus to different FGR. Finally, the fuel structure (e.g. grain size) is a parameter needing further investigation in relation to fuel behaviour.

Giving initial answers to such questions is the goal of this new programme launched in 1994, called FIGARO (Fission Gas Release of MOX). To reach this goal, two MOX fuel rods fabricated by BELGONUCLEAIRE and irradiated among reload fuel at low power in BEZNAU-1 (peak pellet burnup $\approx$ 50 GWd/tM) have been extracted for further examinations. They have been cut and prepared as instrumented segments (central thermocouple and pressure gauge) to be irradiated at variable linear power in the HALDEN reactor (Figure 6).

The objectives of this irradiation are to determine at a burnup of about 50 GWd/tM the way of fuel temperature changes with power and to simultaneously determine the kinetics of fission gas release versus temperature for MOX fuels having different grain sizes.

2.5. NOK - M109

Taking the opportunity of the extraction of the 2 FIGARO rods from a BEZNAU-1 MOX assembly M109 irradiated during five cycles, 8 additional MOX rods have been extracted as well. These 8 rods are now going to be analysed specially on what concern puncture and gas analyses. These experiments will be completed by gamma-scanning measurements and by visual, ECT, profilometry, ceramography measurements on two of these rods.

2.6. NOK - M305/M308

The main objective of this programme is to submit to post-irradiation examination and to ramp testing MIMAS MOX fuel segments (53 cm each) presently irradiated in BEZNAU-1 plant in two assemblies (M305/M308). The target peak pellet burnups are 49 to 55 GWd/tM.

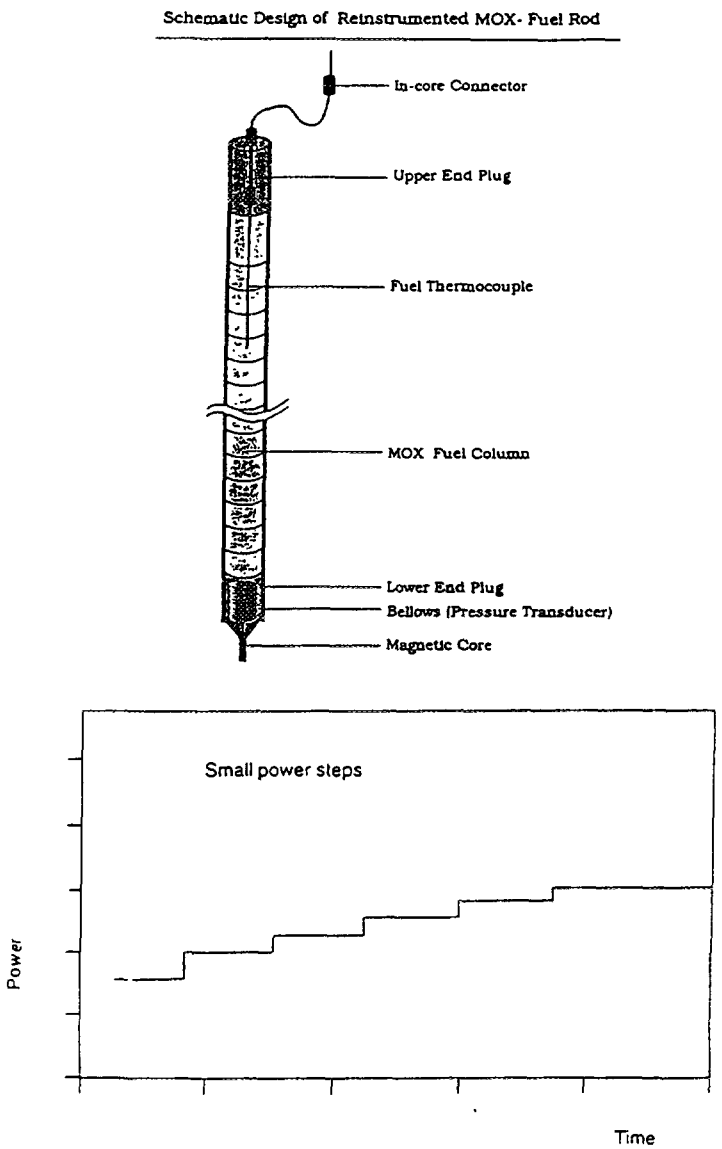

FIG. 6. FIGARO - Design of instrumented pre-irradiated MOX fuel segment and staircase power history

Another objective is a comparison with an alternative MOX fabrication process. These alternative MOX segments have been fabricated by PSI from the gelation process. The segments fabricated in the frame of this programme have been assembled together to constitute four segmented rods. The fuel rods specifications were fully compatible with the standard MOX reload fuel specifications in order to induce no penalty to the BEZNAU-1 plant operation. Some segmented fuel rods will be extracted after 5 or 6 cycles of operation (depending on the plant management's decision) for obtaining data at various burnups.

2.7. The VIP Programmes

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.

These programmes were divided into two programmes :

- VIP-PWR

mock-up 1 : All MOX 17×17 subassembly
mock-up 2 : MOX-Gd 17×17 subassembly.

- VIP-BWR

mock-up 1 : All UO2 : 8×8 subassembly
mock-up 2 : All MOX : 8×8 subassembly
mock-up 3 : Island MOX : 8×8 subassembly.

A comparison of some experimental and calculational data are provided under figures 7 and 8.

2.8. The VIPO Programme

Since 1993, the void coefficient in LWR's was investigated. Calculations had 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.

Figure 9 compares fission rate distribution in one of the configurations.

2.9. VIPEX Programme

As a complement of the VIP Programme the VIPEX Programme, which is now under discussion and should start before the end of 1995, will complete basic core physics data by some specific investigation of MOX data deduced from critical MOX mock-up measurements in the VENUS facility.

Parameter going to be investigated are the Am241 effect, the Beta effective, the flux tilt in a corner MOX rod, the effect of simulated water density by introduction of microrods in the MOX lattice, the control rod worth in MOX conditions and a preliminary investigation on an overmoderated MOX assembly.

2.10. Ariane

Recently the ARIANE Programme which is now underway is going to investigate the MOX irradiated fuel source terms and will serve to validate licensing computer code of the ORIGEN type.

Accent will be focused on accuracy in determining actinides, minor actinides and fission products analysed in three laboratories (SCK•CEN, TUI and PSI) and extracted after extended irradiation in commercial reactors, such as BEZNAU-1 (MOX samples), GÖSGEN (UO_2 samples) and DODEWAARD (MOX and UO_2 samples).

3. INTERNATIONAL SUPPORT

In order to complete this survey of MOX validation programme managed by BELGONUCLEAIRE, Table 2 gives a summary of the permanent and international support which has permitted to extend and to update the MOX data base of numerous organizations.

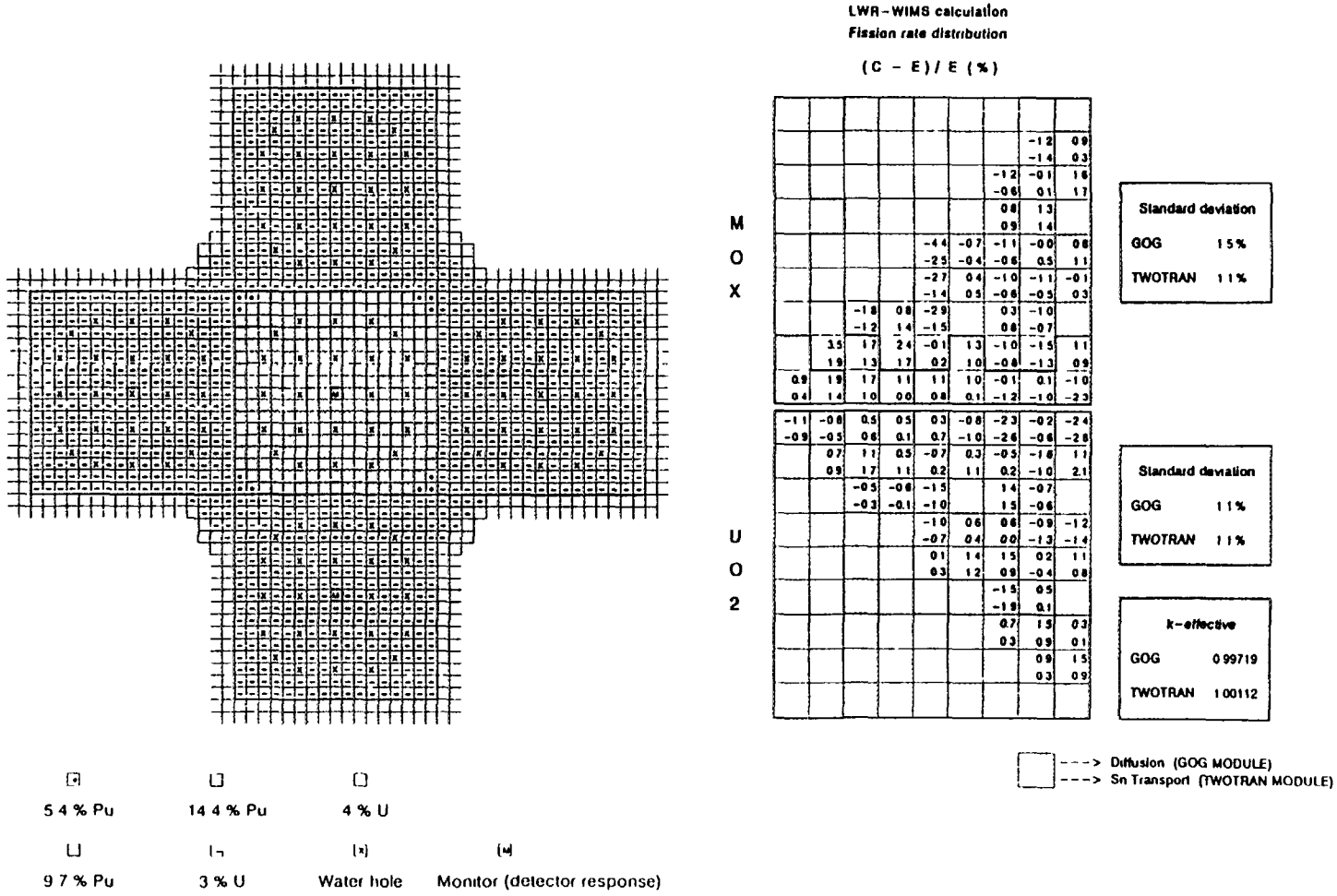

FIG. 7. VIP - BWR All - MOX Configuration

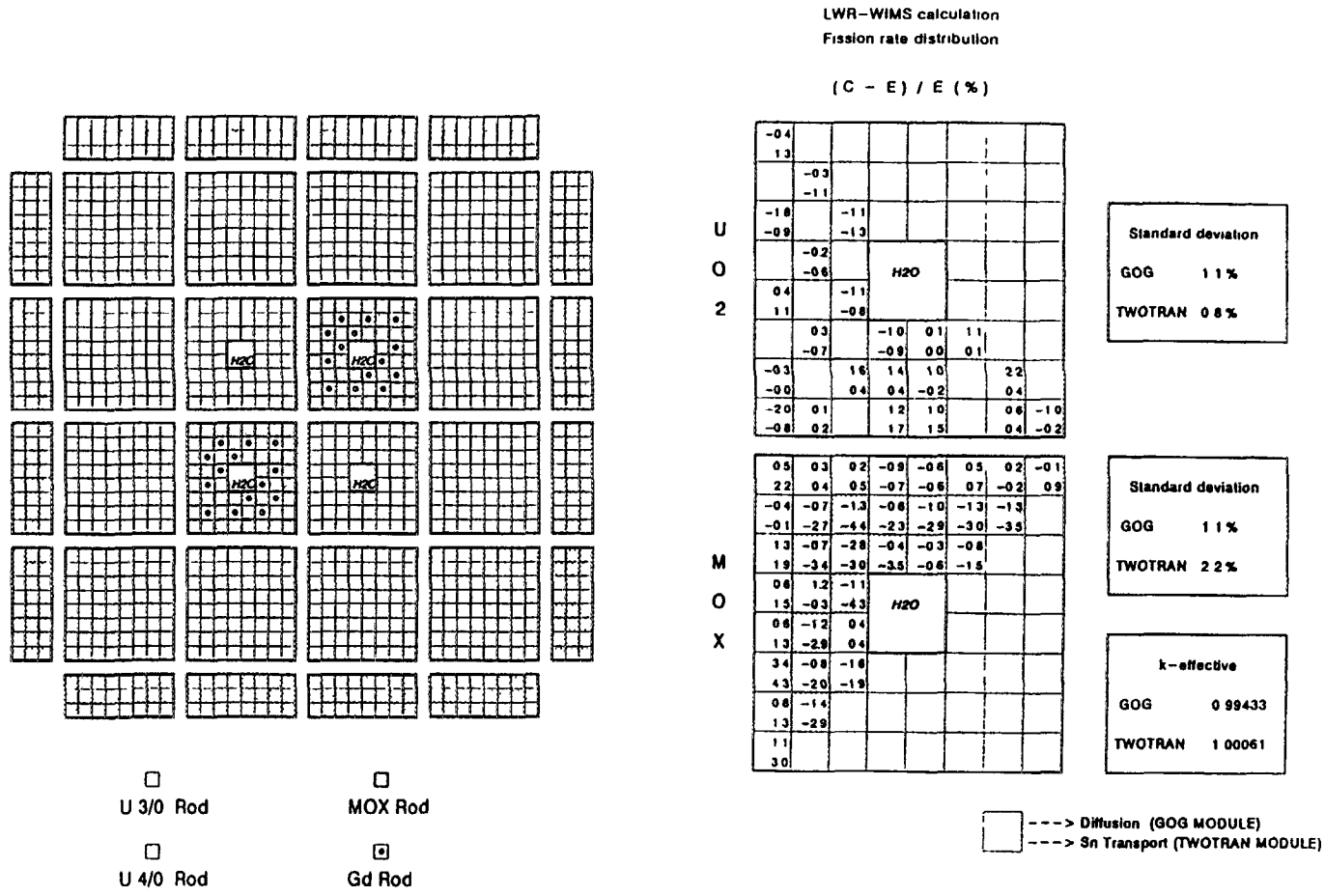

FIG. 8. VIP - PWR Configuration

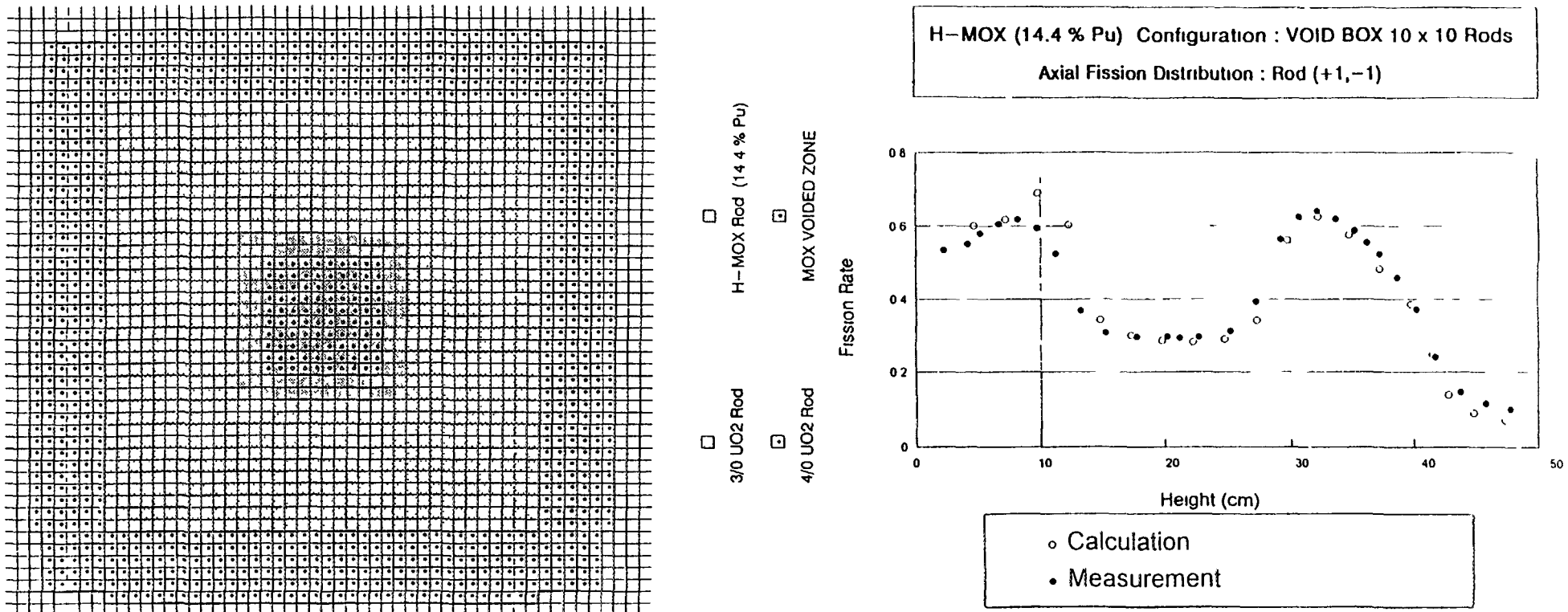

FIG. 9. *VIPO with 10 x 10 void box*

Table 2. Support of MOX International Programme Managed by BELGONUCLEAIRE

Programme	Participants	Facilities
PRIMO	BN, SCK•CEN, CEA-Saclay, FRAMATOME, EDF, COMMOX, AEA Technology, BNFL, NUCLEAR ELECTRIC, PSI, KKG, NOK, NFI, MAPI, PNC, Japanese PWR Utilities	BR3, BR2, OSIRIS, hot cells of SCK•CEN, AEA, CEA
DOMO	BN, SCK•CEN, GKN, TOSHIBA, HITACHI, PNC, NFD, Japanese BWR Utilities, PSI	DODEWAARD, BR2, hot cells of SCK•CEN, PSI
CALLISTO	BN, SCK•CEN, MHI, MAPI, NFI, Nuclear Development Corporation (NDC), BNFL	BR2, hot cells of SCK•CEN
FIGARO	BN, NOK, PSI, MHI, EDF, FRAMATOME (COGEMA), TRACTEBEL (ELECTRABEL), KAERI	BEZNAU-1 HALDEN hot cells of PSI, KJELLER
NOK M109	BN, MHI, NOK, BNFL, (SIEMENS)	BEZNAU-1, hot cells of PSI
NOK M305/308	New programme under discussion	BEZNAU-1, hot cells of PSI
VIP-PWR	BN, SCK•CEN, MHI (NFI, NEL, Japanese Utilities), BNFL	VENUS
VIP-BWR	BN, SCK•CEN, TOSHIBA, HITACHI (BWR Japanese Utilities)	VENUS
VIPO	BN, SCK•CEN, EDF, NFI (Japanese PWR Utilities), BNFL, MHI	VENUS
ARIANE	JRI, TOSHIBA, HITACHI, TRACTEBEL (ELECTRABEL), NOK, GKN, SCK•CEN, BN, SIEMENS, GNS, MHI	BEZNAU-1 GÖSGEN DODEWAARD SCK•CEN, hot cells of PSI, TUI
VIPEX	New Programme under discussion	VENUS

5. CONCLUSIONS

For more than 15 years up to now, BELGONUCLEAIRE as MOX manufacturer is continuously promoting the investigation of MOX fuel through international programmes to obtain better knowledge, to improve necessary models, to update data for design and licensing of MOX fuel in all operating conditions and for extended burn-up levels.

The authors would like to take this opportunity to express their thanks to all organizations which have and wish to continue to support technically and financially this activity.

PLUTONIUM RECYCLING IN FRENCH POWER PLANTS: MOX FUEL IRRADIATION EXPERIENCE AND BEHAVIOUR

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Abstract

Plutonium recycling in PWR's started in France in 1987 with the first reload containing MOX fuel in the Saint-Laurent B1 reactor. By the end of 1994, more than 400 MOX fuel assemblies had been delivered by Fragema to 7 different EDF 900 MW power plants. As the number of PWR units recycling Plutonium is increasing, MOX fuel is required to assume the same operational flexibility as UO2 fuel. So, MOX fuel must follow UO2 fuel developments (1/4 core fuel management, load follow) in terms of discharge burnup and maneuverability. The acquisition of new irradiation data of MOX fuel representative of the current product and operation mode allows to optimize the fuel design leading to improved performance. The development programme set up by French partners CEA, EDF and FRAMATOME, analyzes MOX fuel behaviour through: analytical experiments in experimental reactors conducted under normal and transient operation and, surveillance programmes consisting of the irradiation in commercial reactors and the subsequent examination of MOX fuel assemblies and fuel rods. The paper describes our ongoing programme and discusses the main results obtained so far with a particular focus on the surveillance programme that has been set up on the first MOX reload. Several characterized MOX fuel rods have been withdrawn after each of the three irradiation cycles and have been examined in hot cells. Those data are related to a burnup range up to about 43 Gwd/tM. Power ramp tests performed with fuel segments of those rods showed that MOX fuel behaves particularly well from the pellet-cladding interaction standpoint. Recent data related to high burnup fuel rods (4 irradiation cycles, 52 GWd/tM) and to MOX fuel irradiated under load follow conditions are also presented. These examinations show that from both the waterside corrosion and rod dimension aspects, the MOX assemblies behaved similarly to UO2 fuel. However, the rod puncture data indicate a somewhat higher fractional gas release than UO2 rods: this behaviour is first and mainly explained by the linear heat rates of the MOX which are higher than those of UO2 rods at the same burnups, and to a lesser extent by the more heterogeneous microstructure of the MOX fuel. Fuel restructuring during normal or transient conditions as well as the behaviour of fission products have been carefully examined through micrographs and electron probe micro-analysis providing useful information for modelling purposes.

INTRODUCTION

In the early eighties, it was decided in France to implement a closed nuclear fuel cycle policy by recycling the valuable materials contained in the spent fuel. Such a policy was also adopted by other European countries and by Japan.

By the end of 1994, 460 MOX fuel assemblies had been supplied by Fragema to 7 French 900 MWe PWR's and abroad. Table 1 shows the deliveries and the irradiation experience in the French EDF

reactors. 196 assemblies have already been normally discharged after three cycles, with average burnups of 36 to 38.5 Gwd/tM. Four more have completed successfully a fourth cycle up to 44 Gwd/tM. Two of the French reactors loaded with MOX have been operated under load follow conditions.

Reliability of MOX fuel assemblies has proved to be at least as good as for UO2 fuel ($1.5\ 10^{-5}$ leaking rod, per cycle) since only one MOX assembly has shown a leak, which was slight enough to allow reloading according to EDF criteria [1].

Upgrading of MOX product performance is going ahead according to a progressive and pragmatic approach involving the experience feedback and the joint R & D programme of Framatome, EDF and CEA in order to confirm the good behaviour of the current fuel products and to support the licensing of extended burnup and maneuverability.

FUEL DESIGN AND FABRICATION

Currently, the recycle rate is 30 % MOX assemblies per reload and the type of management used is the OUT-IN-IN third-core strategy (UO2 core management mode at the time of plutonium recycle licensing) on an annual cycle basis. However, new MOX assemblies are not loaded immediately near the baffle in order to minimize their contribution to the vessel fluence.

A typical mixed core reload comprises 16 MOX assemblies and 36 UO2 assemblies. The reload assemblies are all of the current 17 x 17 AFA design with a UO2 fuel enrichment of 3.25 % (equilibrium enrichment of third-core UO2 management). The UO2 matrix for the MOX fuel is made of depleted uranium. The average plutonium concentration of the MOX assemblies is adjusted from the isotopic compositions of the plutonium and of the uranium matrix to obtain cycle lengths equivalent to those obtained with an all-uranium core also reloaded on a three-batch basis. The total plutonium average contents for each MOX production run lies within a 4.5 % - 5.3 % range. This content range is due to the wide isotopic diversity of the plutonium. In addition, the same reload may contain MOX from different production runs [2,3].

The neutronic design of the MOX assembly has to be optimized in order to reduce the power peaking at the UO2 and MOX interface and to obtain the lowest possible shape factor. That is achieved by zoning the assembly using three different plutonium contents in a concentric distribution.

Two processes are currently used for the MOX fuel fabrication:

- the MIMAS (Micronized Masterblend) process adopted by the Belgonucléaire/Dessel plant (35 t/year capacity), consists in the micronizing of UO2 and PuO2 powders to form a master blend with a plutonium content in the range of 20-30 %; this primary blend is then mechanically mixed with a free-flowing UO2 powder (ex. AUC or ex. ADU) to obtain the specified plutonium content

- the COCA (Cobroyage Cadarache) process implemented in the CFCa/Cadarache plant (15 t/year capacity), consists in a single step (ball-milling) mixing of the UO2 (ex. ADU) and PuO2 powders.

  The new MELOX plant (Marcoule) will use the MIMAS process with a UO2 powder obtained via the ADU route. The production is scheduled to start this year, to reach a capacity of 100 tons per year in 1997, with a further increase by the end of the century.

R & D PROGRAMME

It is known that not only the presence of plutonium, but also the fuel materials and the manufacture process used could have an impact on the mixed oxide fuel behaviour compared to the standard Fragema UO2 fuel.

The design codes and models take into account the MOX specific characteristics, nevertheless progressive adjustments are made, based on the development programme set up by the French partners CEA, EDF and FRAMATOME.

It consists of :

(1) analytical experiments in experimental reactors conducted under normal and transient operation in order to assess the kinetics of fission gas release, the fuel temperature evolution, the in-pile densification, the behaviour of pre-irradiated fuel rods under power ramp test in order to determine the failure limit due to PCI, the mechanical behaviour of MOX pellets and the release of fission products from a leaking rod.

(2) global experiments consisting of the irradiation and the examination of MOX fuel assemblies in commercial reactors.

This programme is overviewed hereafter, presenting the main results obtained so far, with a particular focus on the surveillance programmes.

Up to now, most of the data are obtained through the examination of the 3 cycle rods of the first reload in the St Laurent B1 reactor : several characterized MOX fuel rods have been withdrawn after each of the three irradiation cycles and sixteen have been examined in hot cells. These data are related to a burnup range up to about 43 GWd/tM and to the three different plutonium contents of the current MOX assembly.

The examinations of three-cycle fuel rods irradiated under load follow conditions during the last cycle in the St Laurent B2 reactor are almost completed.

On-site measurements have been performed on MOX fuel rods irradiated four cycle in the Gravelines 4 reactor up to a rod burnup of 52 GWd/tM and hot cells examinations are being carried out.

This programme involves also examinations of different fuel types : one, for instance, aimed at providing experience feed-back on the behaviour of MOX pellets fabricated according to the process (MIMAS) implemented in the Melox plant, with a matrix using UO2 powder obtained via the ADU route.

ANALYTICAL EXPERIMENTS

- Physical properties : out-of-pile measurements of lattice parameters, heat capacity and thermal diffusivity as a function of Pu content, Pu homogeneity, stoichiometry of MOX representative of the current PWR fuel fabrication (a significant amount of data on the high Pu content FBR fuel still exists) have been recently carried out in the CEA laboratories of Cadarache. The study of the lattice parameters of the fuel after homogenization confirmed, for example, that the stoichiometric mixed oxide follows the solid solution law (Figure 1). Also, the thermal conductivity of the low Pu content PWR fuel doesn't show significant deviation compared to UO2 fuel (Figure 2).

- Oxide densification : the DENSIMOX experiment was designed to determine the in-pile densification kinetics of MOX fuel from various manufacturing routes in relation with out-of-pile sintering tests. Pellet stack length was monitored through accurate neutron radiographies up to maximum densification. The results concerning two MOX fuels (ADU and AUC powder) are now acquired [4].

- Fuel centerline temperature and fission gas release kinetics : the GRIMOX instrumented in-pile experiment was designed to monitor fuel centerline temperature and fission gas release kinetics during irradiation. The irradiation device also contained UO2 pellets allowing "in-situ"

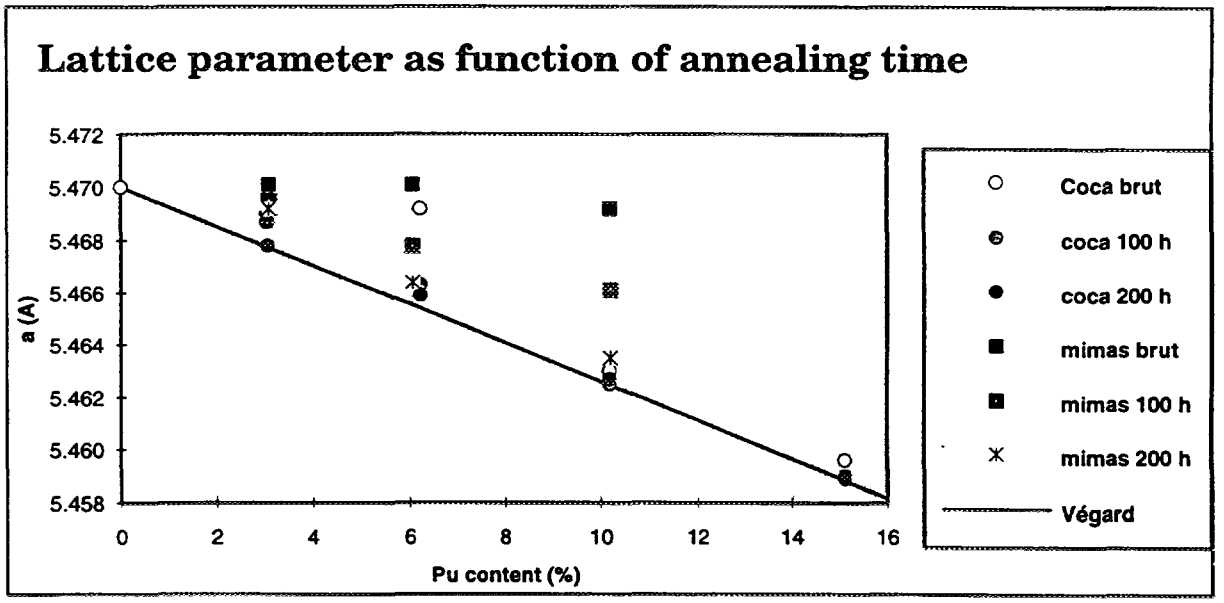


FIG. 1. Lattice parameter as function of annealing time

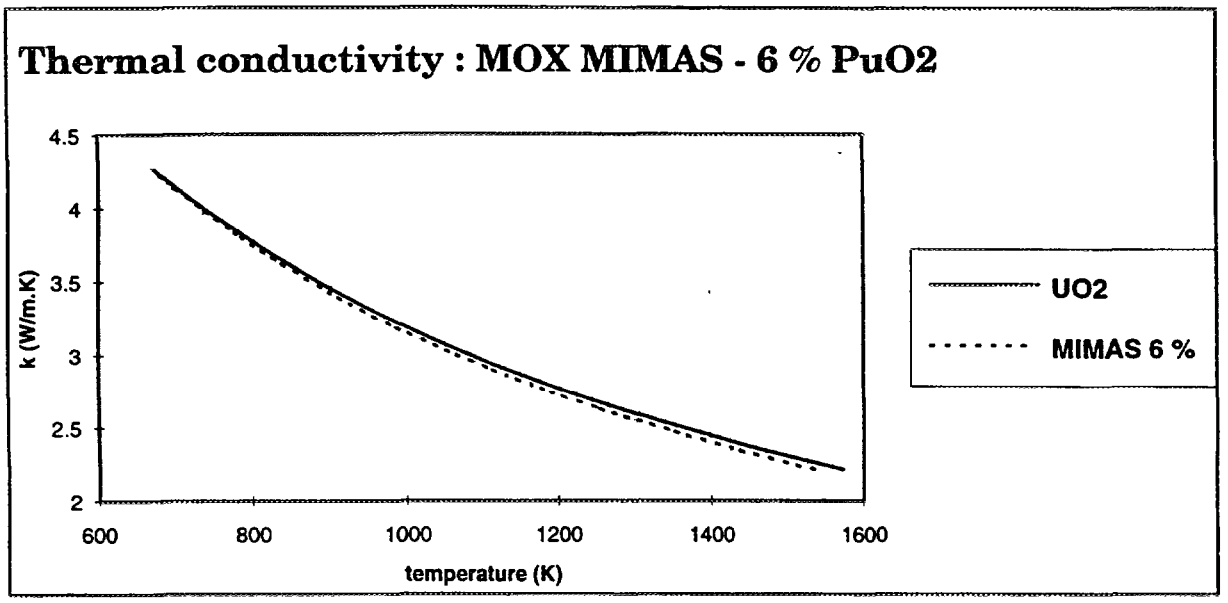


FIG. 2. Thermal conductivity: MOX MIMAS - 6% PuO_2

comparison of changes in parameters. The experiment completed six irradiation cycles in the SILOE experimental reactor (~ 5000 MWd/tM) monitoring moderate as well as high linear heat rates. The results showed that, in the investigated linear heat rate range the MOX temperature was slightly higher than that of the UO2 fuel (Figure 3).

- Pellet-cladding interaction : power ramp tests have been run on 2 and 3 cycle refabricated fuel rods irradiated in the SLB1 reactor. Linear power levels of 480 W/cm have been reached without failure, proving conclusively that MOX fuel behaves particularly well from the pellet-cladding interaction, and therefore from the power plant maneuverability standpoint. Such a favorable

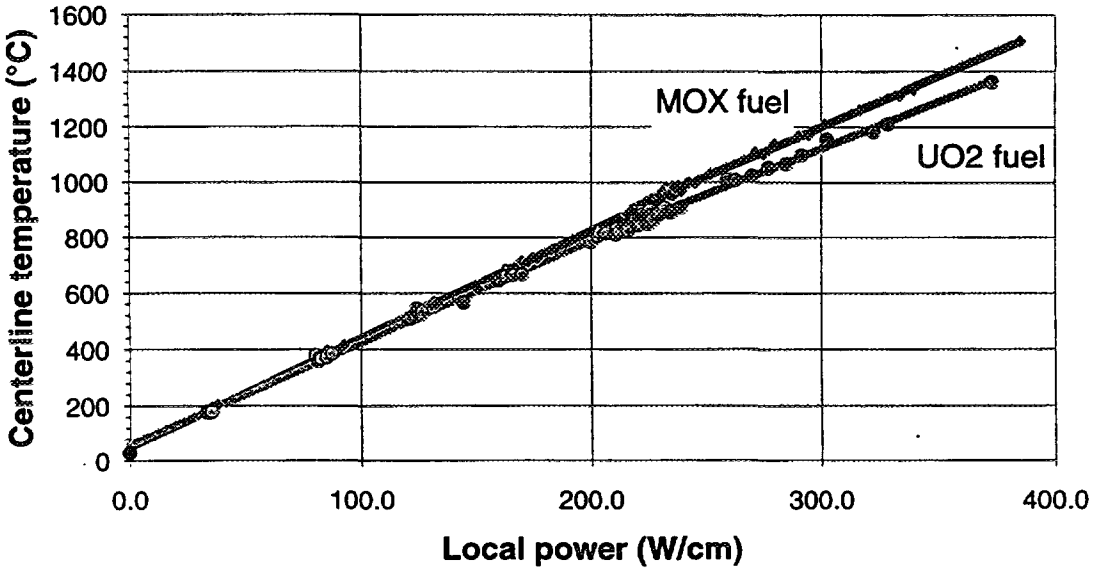


FIG. 3. GRIMOX O2 : centerline temperature versus local power

behaviour is attributed to the higher MOX pellet creep with respect to the UO2 pellet during power transients. This behaviour is in accordance with other published data. Nevertheless, new analytical experiments are being carried out in order to get a better evaluation of this benefit : first, out-of-pile creep tests and also, continuous in-pile measurements of the deformations of MOX and UO2 pellets (of the same geometry) using the DECOR experimental device [5].

- Fission products release : the EDITHMOX experiment consisted of the irradiation of a leaking fresh MOX fuel rod in an experimental loop. The measured gaseous and airborne fission products release rates were comparable with the findings of the uranium oxide experiments. Metallographic examinations showed an appreciable evolution of microstructure with gas precipitation bubbles due to fuel oxidation under water vapor conditions [6].

THE SURVEILLANCE PROGRAMMES

The overall dimensions and the cladding characteristics of the examined fuel rods were similar to those of the standard AFA UO_2 17x17 fuel rod. However, so far as higher fission gas release was expected in design calculations, the helium fill pressure has been lowered for the MOX fuel rods. The total plutonium content of the pellets ranged from 2.88 % (low content) to 5.57 % (high content).

Two characterized MOX fuel assemblies were loaded in 1987 in the 900 MW St Laurent B1 reactor and irradiated during three cycles. A total of sixteen fuel rods have been extracted after one, two or three irradiation cycles for hot-cell examination. Moreover, poolside examinations were also performed in order to verify the geometric behaviour of the assembly components.

The highest burnup rod reached 43 GWd/tM at a nearly constant linear power of about 220 W/cm.

The cladding of the examined fuel rods showed a similar dimensional and corrosion behaviour compared to those of UO_2 rods [7]. Pellet-clad interaction was observed at the end of the second irradiation cycle and a maximum ridge height of 20 μm was measured at the end of three cycles. After three cycles the maximum corrosion layer of 43 μm lies among the low values of the standard UO_2 data

range. Nevertheless, higher fuel densification than standard FRAGEMA UO_2 fuel was observed, in agreement with the expected behaviour of the ex-AUC pellets. Fuel swelling, however follows the same trend as standard UO_2.

The rod puncture data shown in Figure 4 indicates 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 %.

This behaviour mainly results from the quite high power levels of some MOX rods (ranging from about 150 W/cm to 220 W/cm) during the last irradiation cycle, as can be seen on Figure 5.

The data obtained from the St Laurent B2 (SLB2) surveillance programme are coherent with the SLB1 results and don't show an effect of the load follow operation mode during the third irradiation cycle.

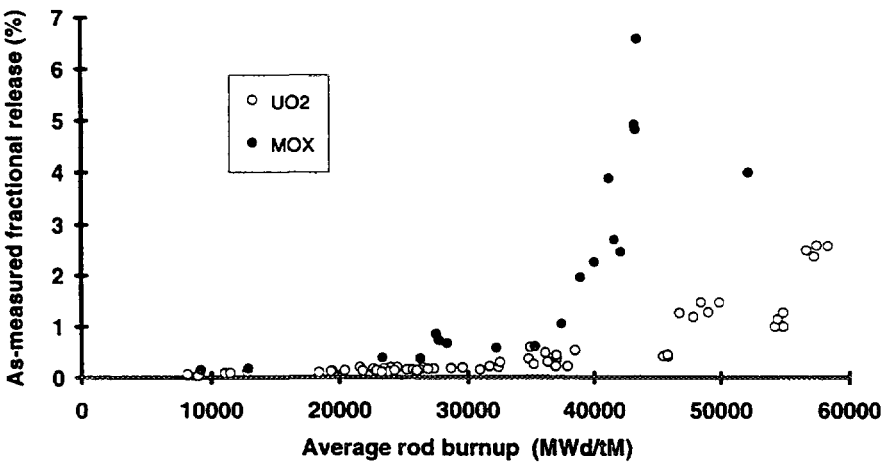


FIG. 4. Rod puncture data

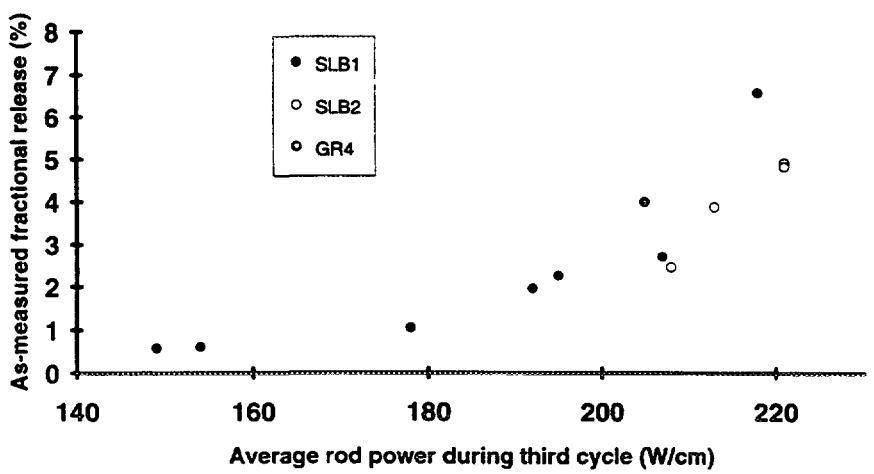


FIG. 5. MOX fractional gas release as function of linear heat rate during the third irradiation cycle

Only a part of this higher fractional release is attributed to the specific MOX fuel properties :

- somewhat higher fuel temperature during irradiation (a little bit lower thermal conductivity than UO_2 fuel and a different radial power profile),
- somewhat higher initial open porosity compared to the standard Fragema UO_2 fuel,
- plutonium agglomerates (up to 30 % Pu rich zones) resulting from the MIMAS fabrication process in which the local burnup is very high (more than 100 GWd/tM) producing high local fission gas concentration. Some fission gas atoms can be dissolved in the UO2 matrix and, to a lesser extent, migrate to the rod void volume through cracks or porosities.

Fuel microstructure as well as the behaviour of fission products were carefully examined through micrographs and electron probe micro-analysis (EPMA) providing useful information for modelling purposes [8].

The typical microstructure of the 3 cycle MOX fuel is shown in Figure 6.

The specific features relevant only to the MOX fuel behaviour and to fission gas release are described hereafter.

At the pellet edge, the Pu agglomerates are well characterized by a dense pore population resulting from fission gas generation and bubble coalescence. Fine metallic precipitates are also observable.

Towards the pellet center, the fine porosities coalesce to yield larger pores in the plutonium rich zones ; the surrounding UO_2 matrix preserving its as-fabricated microstructure.

At the pellet center, pore coalescence in the Pu agglomerates leads to the formation of big cavities surrounded by large metallic precipitates (4-5 μm). Grain boundary porosity is also observed in the UO_2 matrix of the fuel which yielded significant fractional gas release.

It is evident that this fuel restructuring is correlated to the burnup and the linear power the fuel experienced, nevertheless it is difficult to find a correlation with the measured fractional release of the fuel rods. In this respect, EPMA provided interesting information on the fission gas release mechanism in MOX fuel by means of the analysis of 1, 2 and 3 cycle fuel irradiated at various power levels.

In the low temperature regions, high xenon concentration peaks are measured in, or in the close vicinity of the plutonium rich zones, whereas in the high temperature zones or in the high burnup fuel, xenon is no longer detected in the Pu agglomerates and exhibits a quite flat distribution in the surrounding UO_2 matrix. The fission gases which are present in the UQ matrix due to the migration from the Pu agglomerates, recoil or in-situ formation follow the expected mechanism of the thermally-activated fission gas release. This is illustrated in Figure 7 which compares the diametral profile of Xenon concentration in 1, 2 and 3 cycle fuel.

In one-cycle rod, xenon is mainly localized in the plutonium agglomerates, very little is found in the UO_2 matrix. With increasing burnup, the amount of xenon in the matrix increases significantly and for the high rated three-cycle fuel, release of xenon at pellet center is clearly observed.

EPMA data showed also that no measurable plutonium diffusion in the UO_2 matrix occurred in this fuel operated under normal condition. Conversely, UO2 and PuO2 homogenization has been observed at the pellet centerline of the ramped fuel rods.

Globally this programme as well as previously published data show that MOX fuel, in spite of its heterogeneous microstructure, has a similar fission gas release mechanism in the thermal regime as UO_2 fuel. Nevertheless, it seems that a small fraction of the fission gases coalesced in bubbles in the plutonium rich zones can leave the fuel for the rod void volume via cracks and open porosities of the pellet. That

could explain the slightly higher fission gas release observed for the MOX fuel which has experienced a significant local burnup in MOX agglomerates (typically after two irradiation cycles).

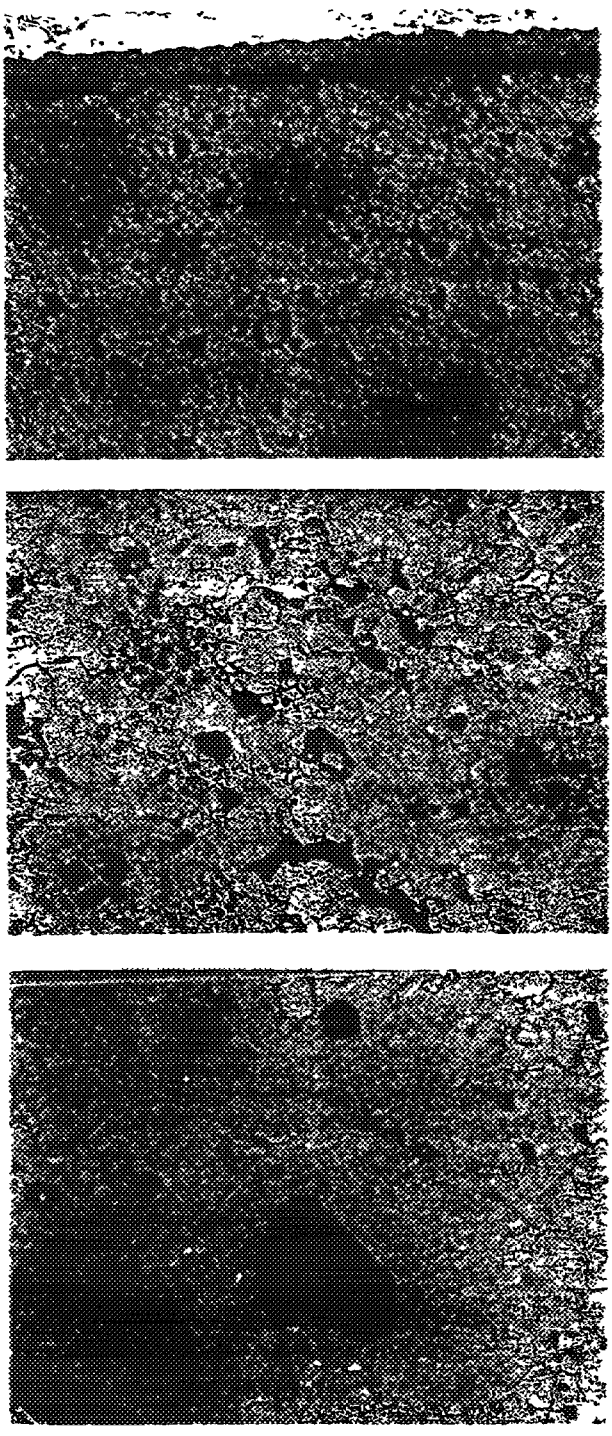
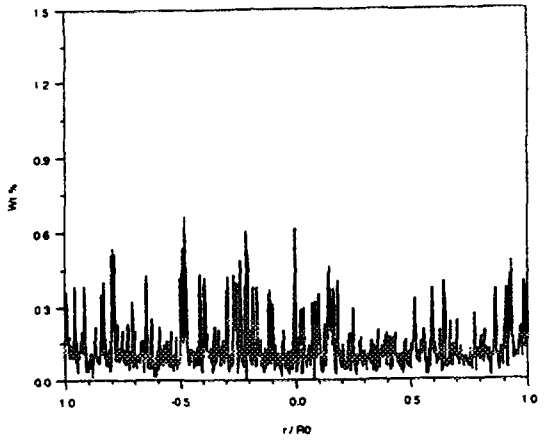
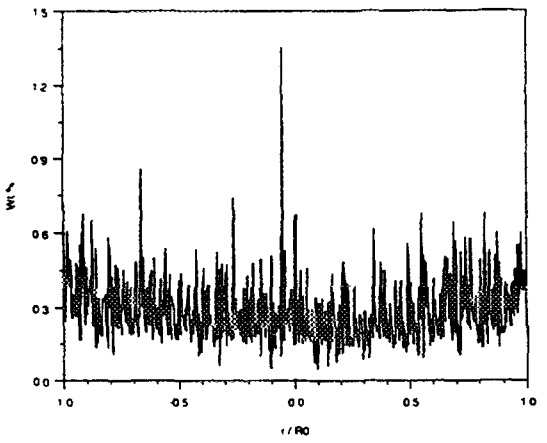


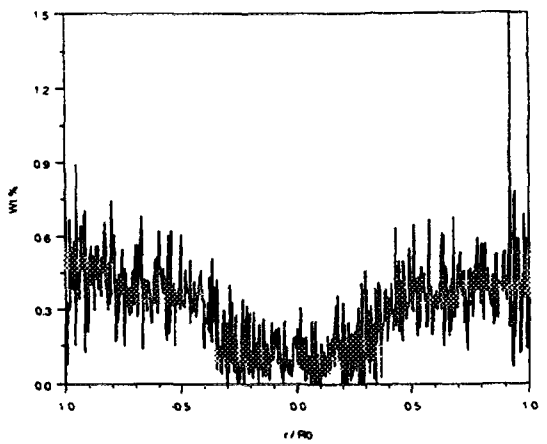
FIG. 6. Microstructure of a 3 cycle MOX fuel (local burnup : 46 Gwd/tM)



1-Cycle fuel



2-Cycle fuel



3-Cycle fuel

FIG. 7. EPMA diametral scan of the Xe element across 1, 2 and 3 cycle fuel pellets

The first data obtained from the High Burnup Programme (GRA4) showed no burnup enhancement of the fractional release. The fission gas release after 4 cycles is similar to that measured after 3 cycles due to the lower heat rate experienced by the rods during the last irradiation cycle.

Modelling of these phenomena in fuel rod design codes leads to calculations with satisfactory predictive quality.

However, investigations are still underway to take the most advantage of the large amount of data obtained in these programmes.

Table 1 : MOX fuel assembly deliveries and irradiation experience in FRANCE up to end 94

YEAR	87	88	89	90	91	92	93	94
Number of assemblies delivered	16	32	48	56	64	64	56	88
Number of reactors with Mox fuel	1	2	4	5	5	5	6	7
Irradiation Experience (cumulated number of assemblies x cycles)	0	16	64	128	256	448	624	820

CONCLUSION AND PROSPECTS

These results obtained so far confirm the good behaviour (dimensions, corrosion, internal pressure, PCI) of MOX fuel up to the typical burnup reached with 1/3 annual core management.

- Nevertheless, with the applied fuel managements, the fission gas release measured in MOX fuel was larger than in UO_2 fuel. This is mainly due to higher power histories, particularly a higher power level during the last irradiation cycle.

However, some fission gas release data obtained at higher burnup by reloading MOX fuel assemblies for a fourth cycle don't show any unexpected burnup enhancement.

The results of analytical experiments provided useful information for the confirmation or the validation of the main models simulating the behaviour of the fuel rod.

The acquired experience feed-back, together with the very satisfactory operating results, has led to advances in MOX assembly design [9]. The resulting product - AFA 2G MOX - presents an optimized zoning and includes the improvements made on the current AFA 2G UO2 product : mainly an optimized Zircaloy-4 cladding with an enhanced corrosion resistance, mixing grids with higher performance and an anti-debris device. AFA 2G MOX is designed for an assembly burn-up of 43-45 GWd/t and for an intensive grid-follow operation in order to satisfy the needs of EDF reactors.

AFA 2G MOX batches will be delivered this year to a 900 MWe EDF unit. They will be used with the newly implemented "hybrid" fuel management scheme where 3.7 % enriched UO2 assemblies are irradiated for four annual cycles and MOX assemblies remain three cycles.

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MOX FUEL IRRADIATION BEHAVIOUR: RESULTS FROM X-RAY MICROBEAM ANALYSIS

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Abstract

The behaviour of plutonium, xenon and caesium were investigated in two sections of irradiated MOX fuel produced by the OCOM process. In one fuel (OCOM30), the MOX agglomerates contained 18 wt% fissile plutonium, and had a low volume fraction of 0.17; in the other (OCOM15) the agglomerates contained 9 wt% fissile plutonium, and had a high volume fraction of 0.34. Both fuels had been irradiated under normal power reactor conditions to a burn-up of approximately 44 GWd/t. The main aim of the work was to establish whether the above differences in compostion affected the percentage fission gas released by the fuels. Since U/Pu interdiffusion did not occurred during the irradiation, both fuels remained inhomogeneous on the microscopic scale. However, the concentration of plutonium in the MOX agglomerates decreased by about 50% as a result of fission, whereas the plutonium content of the UO_2 matrix increased by about a factor of four to approximately 2 wt% due to neutron capture by ^{238}U. The agglomerates in the OCOM15 fuel generally exhibited a finer structure due to the lower burn-up. More than 80% of the fission gas had been released from the oxide lattice of the MOX agglomerates in both fuels. However, a very high fraction of this gas precipitated and remained in the pore structure of the agglomerates. Consequently, puncturing revealed that for both fuels the percentage of gas released to the rod free volume increased from less than 0.5% at 10 GWd/t to a maximum of 3.5% at 45 GWd/t. The conclusion is that the percentage of gas released by MOX fuel is largely unaffected of the level of inhomogeneity of the fuel. In both fuels caesium showed near complete retention in both the MOX agglomerates and the UO_2 matrix.

1. INTRODUCTION

The AUPuC and OCOM manufacturing processes pioneered by Siemens-KWU produces MOX fuel that is highly soluble even in the as-fabricated (unirradiated) condition [1,2]. The manufacturing process adopted depends on the form in which the plutonium is supplied, i.e., whether Pu-nitrate from reprocessing tanks or PuO_2 powder. Fabrication of the fuel involves mechanically blending a master-mix rich in PuO2 with natural UO_2 powder and plutonium containing scrap from MOX fuel pellet production. The product contains about 5 wt% Pu and consists of master-mix particles (MOX agglomerates) up to 200 μm in size, irregularly dispersed in a UO_2 matrix. Thus, compared with UO_2 fuel, the fuel has a duplex structure and exhibits areas of high and low burn-up.

This paper looks at the effect of the inhomogeneity of OCOM fuel on the percentage of fission gas released under steady-state irradiation conditions. The investigations reported are a direct extension of previously published work [3], which in essence considered the influence of the fuel temperature on the redistribution of plutonium and the behaviour of xenon and caesium. Electron probe microanalysis

Table 1 Fuel pellet and pin segment design characteristics

Design Parameter	OCOM15	OCOM30
Fissile Pu [a] (wt%)	3.06	3.06
Total Pu (wt%)	4.43	4.47
PuO_2 in agglomerates (wt%)	15	30
Vol% of agglomerates	26	13
U enrichment (% ^{235}U)	0.72	0.72
Grain size [b] (μm)	5-6	5-6
Stoichiometry (O/M)	1.994	1.992
Fuel density (%TD)	95	95
Pellet diameter (mm)	9.13	9.13
Diameterical gap (mm)	0.17	0.17
Fill gas [c]	He (2.25)	He (2 25)
Cladding material	Zircaloy-4	Zircaloy-4

a $^{239}Pu+^{241}Pu$.
b Linear intercept.
c Figure in parenthesis is the pressure in Mpa.

(EPMA) results for two OCOM fuels are compared. In one fuel (OCOM30), the MOX agglomerates contained a high concentration of fissile plutonium, but had a low volume fraction; in the other (OCOM15), the agglomerates contained a low concentration of fissile plutonium, but had a high volume fraction. In these fuels, EPMA was used to measure the concentrations of xenon, caesium, neodymium and plutonium in the MOX agglomerates and their radial distributions in the UO_2 matrix. The agglomerates analysed were located at different radial positions in the fuel, including the pellet rim, and varied in size from 40 to 160 μm. To obtain a comprehensive picture of the behaviour of the above elements, point analysis and area analysis, which included both agglomerates and the UO_2 matrix, were performed.

It was found that although the two OCOM fuels differed markedly in their makeup, no significant difference in the level of gas release could be detected.

2. FUEL CHARACTERISATION AND IRRADIATION HISTORY

The design characteristics of the two OCOM fuels are given in table 1. Both fuels contained about 4.5 wt% plutonium. Roughly 5-10% of the fuel material was recycled from MOX fuel pellet production which was contained in the UO_2 matrix. The master-mix (MOX) agglomerates constituted 34 vol% of the OCOM15 fuel and 17 vol% of the OCOM30 fuel.

The fuels were irradiated under normal PWR conditions in the KWO reactor at Obrigheim in Germany The irradiation spanned four reactor cycles and lasted 1260 EFPD. During the irradiation the average linear power was 19.4 kWm^{-1} for the OCOM15 segment and 20.4 kWm^{-1} for the OCOM30 segment. The fuel cross-sections on which EPMA was carried out had burn-ups of 43.6 GWd/t (OCOM15) and 44.5 GWd/t (OCOM30). It is apparent from the small gap width and from the low power rating that the fuel temperature during the irradiation was relatively low.

3. METHODS

EPMA was carried out at an electron acceleration potential of 25 keV and a beam current of 250 nA. The matrix correction was carried out using the QUAD2 program of Farthing et al. [4].

Xenon was analysed using the approach pioneered at the Institute for Transuranium Elements [5]. This gives the concentration of xenon in solution and trapped in intragranular gas bubbles smaller than 0.1 μm. The confidence interval on the measured Xe concentrations at a significance level of 99% is about 5% relative on a concentration of 0.5 wt% and 10-20% relative at 0.05 wt%. This uncertainty is due solely to the statistics of X-ray counting. Similar uncertainty levels are expected in the cases of Cs and Nd.

Plutonium was analysed using the M X-ray line. The measured intensity of this line was corrected for X-ray contributions from the U M_2 line as described elsewhere [6]. The confidence interval on a measured Pu concentration of 2 wt% is around 10% relative at a significance level of 99%. This uncertainty is due to the statistics of X-ray counting and increases to 20% relative after correction for the use of a compound standard and after accounting for the X-ray contributions from the U M_2 line.

3.1. Analysis of the UO_2 matrix

The radial distributions of Pu, Nd, Xe and Cs in the UO_2 matrix of the OCOM30 fuel were obtained by point analysis at radial intervals of 50 to 250 μm. At each location six determinations for Xe, Cs and Nd, and four determinations for U and Pu were made. These were up to 10 μm apart and were placed to avoid grain boundaries, pores and cracks.

In the OCOM30 fuel the concentrations of Pu and Nd in single grains of the UO_2 matrix were measured at three radial positions; r/r_o= 0.82, 0.36 and 0.14. At each location six grains were analysed. In the OCOM15 fuel the concentrations of Pu, Nd, Xe and Cs were measured in the UO_2 matrix. In this case, however, the determinations were restricted to the cold outer region of the fuel at r/r_o= 0.98, 0.90, 0.80 and 0.70.

The specimen current image (absorbed electron current) was used to obtain information about the fuel microstructure at the locations selected for analysis, and to position the electron beam.

3.2. Analysis of the MOX agglomerates

The MOX agglomerates in both fuels were analysed in detail. In the OCOM15 fuel, agglomerates at the pellet rim, at r/r_o= 0.85 and 0.35 were analysed. The agglomerates analysed in the OCOM30 fuel were situated at the pellet rim, at r/r_o= 0.86, 0.76, 0.58 and 0.28. At each radial position except 0.76 and 0.28, one large and one small agglomerate was selected for analysis. In addition, in both fuels an agglomerate located directly at the pellet rim and open to the fuel gap was analysed. For the purpose of comparison, these agglomerates were analysed together with one or more of similar dimensions located just below the fuel surface.

The concentrations of Pu, Nd, Xe and Cs in the MOX agglomerates were determined by point analysis at intervals of 5-10 μm along a line traversing the agglomerate. The specimen current image was used to obtain information about the distribution and morphology of the pores and gas bubbles in the agglomerates selected for analysis. These features appear white in the photomicrographs shown in Fig.1.

3.3. Integral Analysis of the Fuel Cross-section

The integral concentrations of Pu and Nd in the OCOM15 fuel and of Pu, Nd, Xe and Cs in the OCOM30 fuel were measured using area analysis along the fuel radius. A raster measuring 300x300 μm was employed and the distance between the mid-planes of adjacent rasters was also 300 μm. In this way an unbroken radial strip of fuel containing both matrix and MOX agglomerates was analysed. At each of the sixteen steps along the radius three determinations were made.

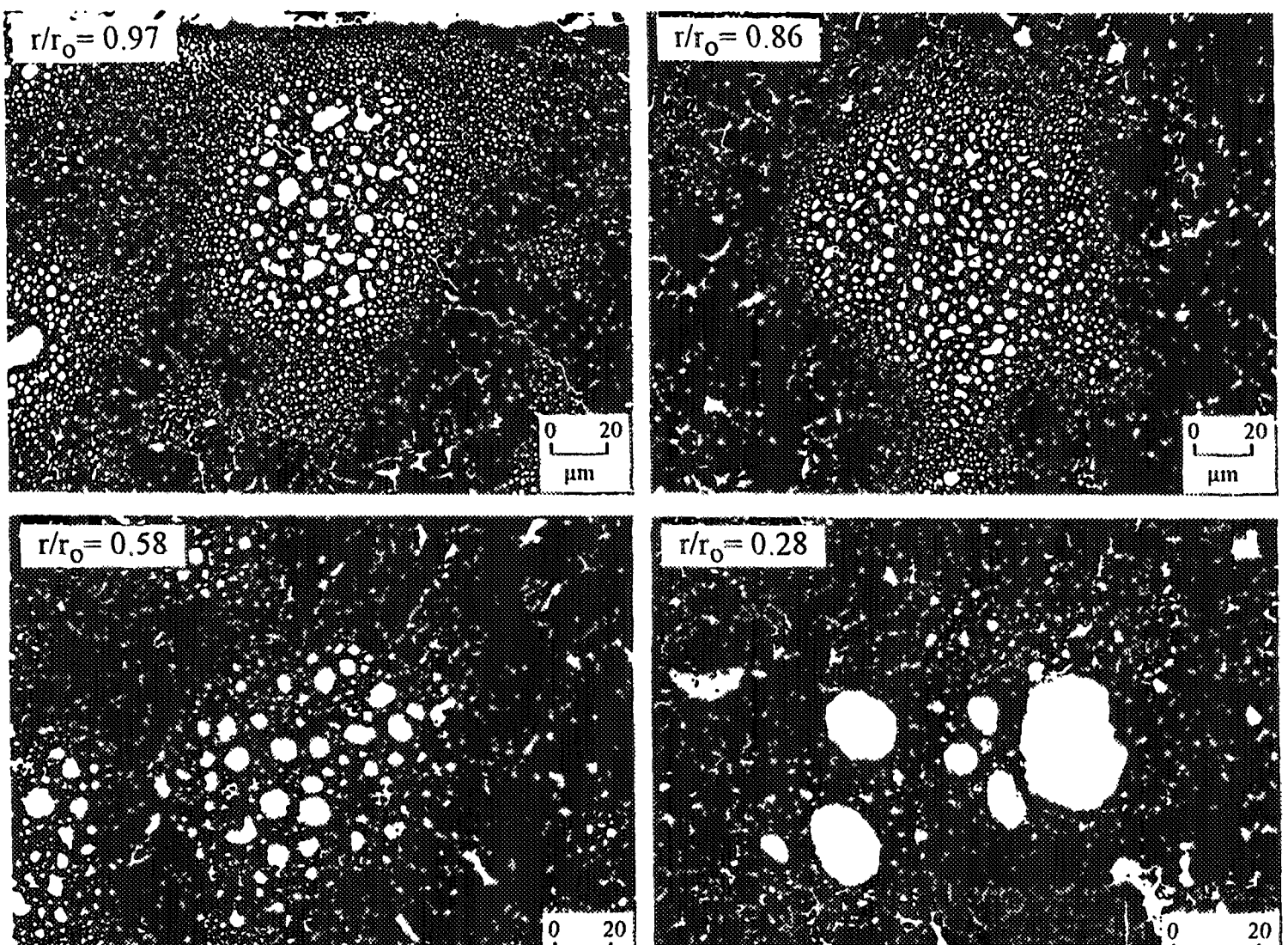

Fig.1. Electron absorption micrographs showing the appearance of MOX agglomerates at four radial positions in the OCOM30 fuel. Note that the porosity is coarser in the agglomerate at the fuel surface than in the one at r/r_o= 0.86.

4. RESULTS

4.1 Microstructure of the MOX agglomerates

Figure 1 shows the appearance of MOX agglomerates at four radial positions in the OCOM30 fuel. It is seen that the Pu-rich MOX agglomerates were extremely porous after irradiation. In the cold outer region of the fuel the agglomerates were characterised by dense clusters of small pores a few microns in size. With increase in fuel temperature, however, the number of pores decreased and the pore size increased until in the central region of the fuel the MOX agglomerates were distinguished by just a few coarse pores. Remarkably, in the outer region of the fuel, agglomerates near the fuel surface contained larger pores then agglomerates at greater depths. The increase in pore size is attributed to the higher local burn-up at the fuel surface, which is a consequence of the radial depression of the thermal neutron flux.

In Fig.2, the microstructures of MOX agglomerates at similar radial positions in the cold outer regions of the OCOM15 and OCOM30 fuels are compared. It is seen that the porosity is coarser in the OCOM30 agglomerates. The difference in pore size undoubtly reflects the large difference in burn-up between the MOX agglomerates of the two fuel (see next section).

4.2 Burn-up in the MOX agglomerates and the UO_2 matrix

The local concentration of neodymium was taken as a measure of the local burn-up in the MOX agglomerates and the UO_2 matrix. For this purpose, 0.106 wt% neodymium was assumed to correspond to 10 GWd/t. As seen from Table 2, neodymium concentrations of 0.46 and 0.47 wt% were measured in the OCOM15 and OCOM30 cross-sections, respectively. These concentrations correspond to 43.4 and 44.3 GWd/t which are very close to the reported burn-up values of 43.6 and 44.5 GWd/t.

The burn-up in the MOX agglomerates was considerably higher in the OCOM30 fuel. Figure 3 shows the burn-up in a number of agglomerates at different radial positions in the OCOM15 and OCOM30 fuels. The burn-up reported is based on the highest neodymium concentration measured in each case. It is seen that at the fuel surface the burn-up in the OCOM30 agglomerates was about 270 GWd/t whereas in the OCOM15 agglomerates it was about 160 GWd/t. Similarly, in the central region of the fuel, the burn-up in the OCOM30 agglomerates was around 190 GWd/t, but did not exceed 110 GWd/t in the OCOM15 agglomerates. It is evident from these figures that the MOX agglomerates in the outer region of the fuel had accumulated more burn-up than those in the central region of the fuel.

As in conventional UO_2 fuel, the radial burn-up profile in the UO_2 matrix of the MOX fuel was quite flat in the body of the fuel, but increased sharply near the fuel surface. The radial distribution of neodymium in the UO_2 matrix of the OCOM30 fuel, which represents the radial burn-up profile, can be seen in Fig.4. Over most of the fuel radius, the concentration of neodymium is slightly less than 0.3 wt%, which corresponds to about 28 GWd/t, but at $r/r_o \approx 0.90$ it increases abruptly from this level to 0.66 wt% at the pellet rim. This concentration is eqivalent to a burn-up of 62 GWd/t. The averge concentration of neodymium in the UO_2 matrix was 0.33 wt%, giving a burn-up for the cross-section of 31.1 GWd/t.

Although, the neodymium profile in Fig.4, indicates that the burn-up in the UO_2 matrix of the OCOM30 fuel was rather uniform, the analysis of single grains revealed that in truth the burn-up varied widely from grain to grain. Figure 5 shows the burn-up in single grains at $r/r_o = 0.82$, at $r/r_o = 0.36$ and $r/r_o = 0.14$ in the OCOM30 fuel. At each of these radial positions, the burn-up in six grains is seen. The burn-up in the grains was found to vary between 13 and 88 GWd/t. The large variation in burn-up is due to the different concentrations of fissile plutonium in the grains, which represent different material; i.e., UO_2, MOX fuel scap and the MOX agglomerates (see section 4.4). It is assumed that 13 GWd/t is representative of the burn-up in areas of the UO_2 matrix that were unspoilt by plutonium from agglomerate particles or plutonium from MOX scrap. The neodymium profile in Fig.4 indicates a uniform burn-up distribution, because the data points used to draw concentration profile are the average of six determinations made on different grains.

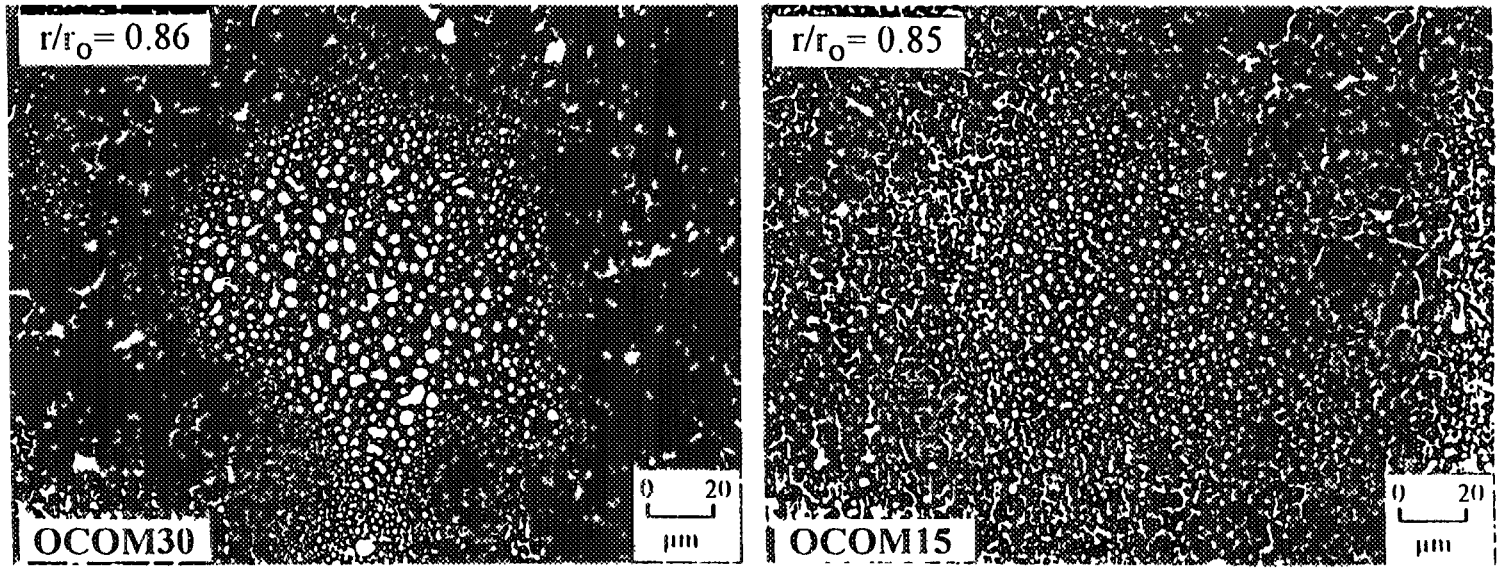

Fig.2. Electron absorption micrographs showing the appearance of MOX agglomerates at similar radial positions in the cold outer regions of the OCOM15 and OCOM30 fuels. The porosity is coarser in the OCOM30 agglomerate.

Table 2 Integral concentrations of Pu, Nd, Xe and Cs in the fuel cross-section and in the UO_2 matrix

Fuel	Concentration, wt%			
	Plutonium	Neodymium	Xenon	Caesium
		MOX Agglomerates + UO_2 Matix		
OCOM30	3.6	0.47	0.52	0.53
OCOM15	3.2	0.46	...	...
		UO_2 Matrix		
OCOM30	1.9	0.33	0.52	0.28
OCOM15	...	...	...	...

4.3 Plutonium burn-out in the MOX agglomerates

In Fig. 6 the concentration of Pu in the MOX agglomerates in the two fuels is plotted against the neodymium concentration (i.e., the burn-up). Included in the plot are data from MOX agglomerates of different size (40 to 180 μm) and from agglomerates at different radial positions, including the fuel rim. The plutonium concentration in the OCOM30 agglomerates had decreased from 26.5 to approximately 13 wt% at a burn-up above 140 GWd/t, while in the OCOM15 agglomerates it had decreased from 13.2 to roughly 6.5 wt% at a burn-up higher than 55 GWd/t. Thus, in both fuels approximately 50% of the plutonium in the agglomerates had been burnt during the irradiation.

4.4 Plutonium in the UO_2 matrix

The radial distribution of Pu in the UO_2 matrix of the OCOM30 fuel can be seen in Fig.4. Like the neodymium concentration profile, the plutonium concentration profile exhibits a steep rise close the pellet surface due to enhanced neutron capture. Volume integration of this profile gives an average plutonium concentration of 1.9 wt%. Thus, it seems that neutron capture during the irradiation caused a four fold increase in the concentration of plutonium in the UO_2 matrix.

The plutonium in the UO_2 matrix was far less homogeneously distributed than one would be led to believe from the plutonium concentration profile in Fig.4. Results from analysis of single grains in the OCOM15 and OCOM30 fuels are shown in Fig.7. It is seen that the measured plutonium concentration ranged from 1.2 to 14.3 wt%. It is also seen that the plutonium concentrations fall in to three distinct categories. Those resulting from MOX agglomerate particles, those resulting from the addition of MOX fuel scrap, and those resulting from neutron capture by ^{238}U during the irradiation.

Presumably, agglomerate particles were created when the master-mix was mechanically blended with the UO_2 power during fabrication of the fuel. The presence of this plutonium-rich material, resulted in grains with plutonium concentrations as high as 14.5 wt%. Grains with plutonium concentrations in the range 1 to 6 wt% and neodymium concentrations above 0.3 wt% are assumed to be grains of MOX fuel scrap. While grains with plutonium concentrations of 3 wt% or less and neodymium concentrations below 0.3 wt%, are assumed to have started life as pure UO_2 and their plutonium is assumed to result solely from

neutron capture. Incidentially, matrix grains rich in plutonium contained clusters of small fission gas bubbles which made them easy to identify.

4.5 Fission gas release

The EPMA data for fission gas release from the OCOM30 fuel is summarised in Table 3. As can be seen, the integral released from the fuel cross-section was approximately 12%. In the case of the MOX agglomerates, however, about 90% of the fission gas had been released from the mixed oxide lattice. It is clear from Fig.8 that an extremely high percentage of gas had been released from $(U,Pu)O_2$ lattice of

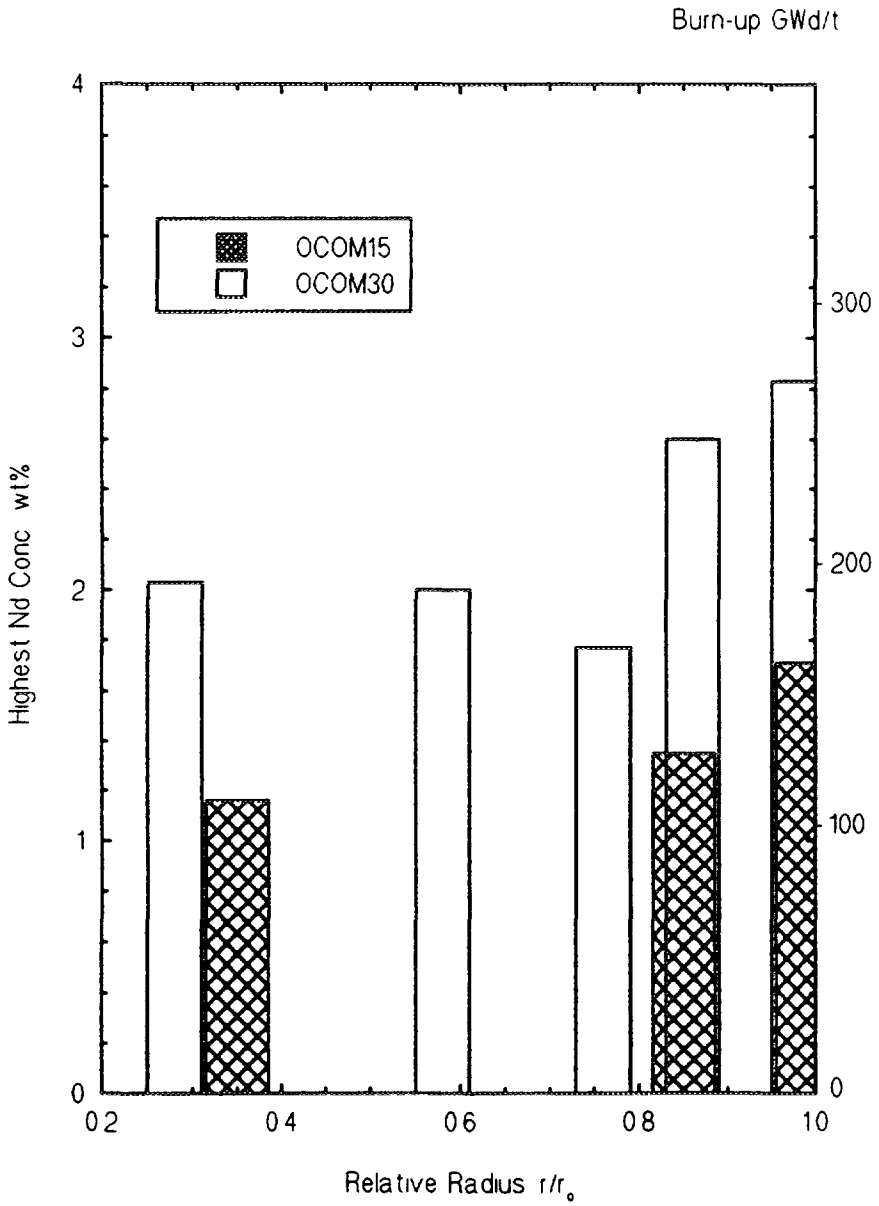

Fig.3. Burn-up in the MOX agglomerates in the OCOM15 and OCOM30 fuels as derived from their neodymium content. A much higher burn-up was reached in the OCOM30 agglomerates.

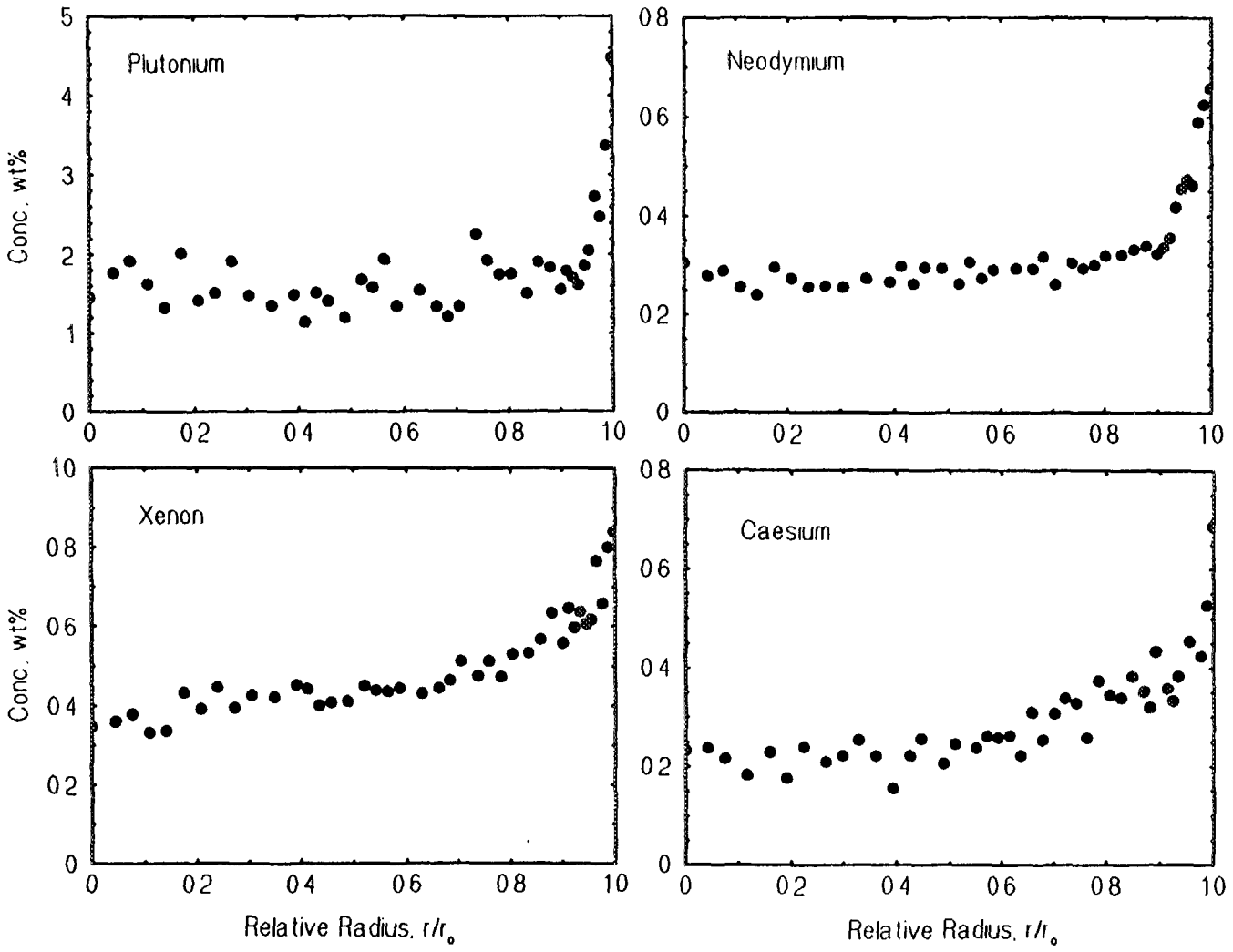

Fig.4. Radial distributions of Pu, Nd, Xe and Cs in the UO_2 matrix of the OCOM30 fuel.

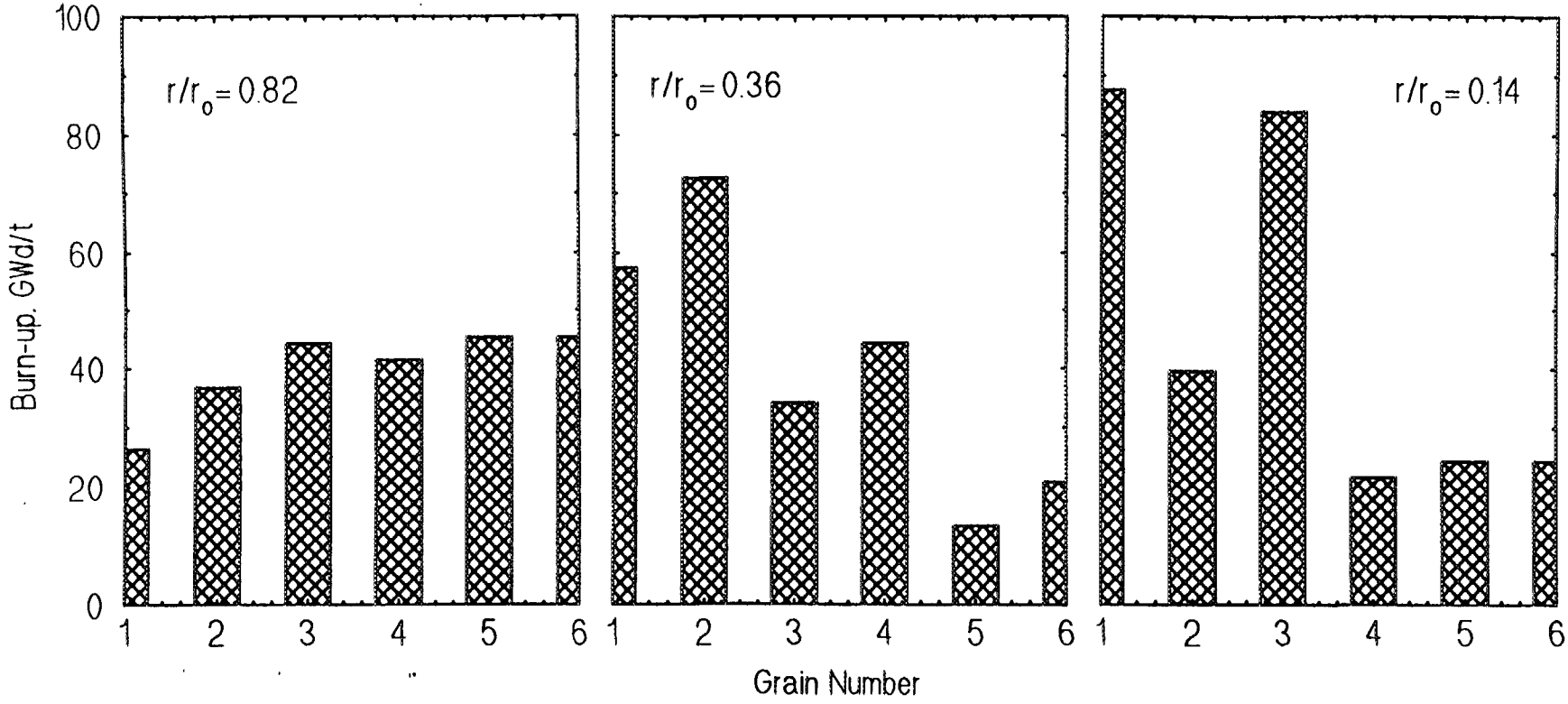

Fig. 5 Burn-up in single grains at three radial positions in the UO_2 matrix of the OCOM30 fuel.

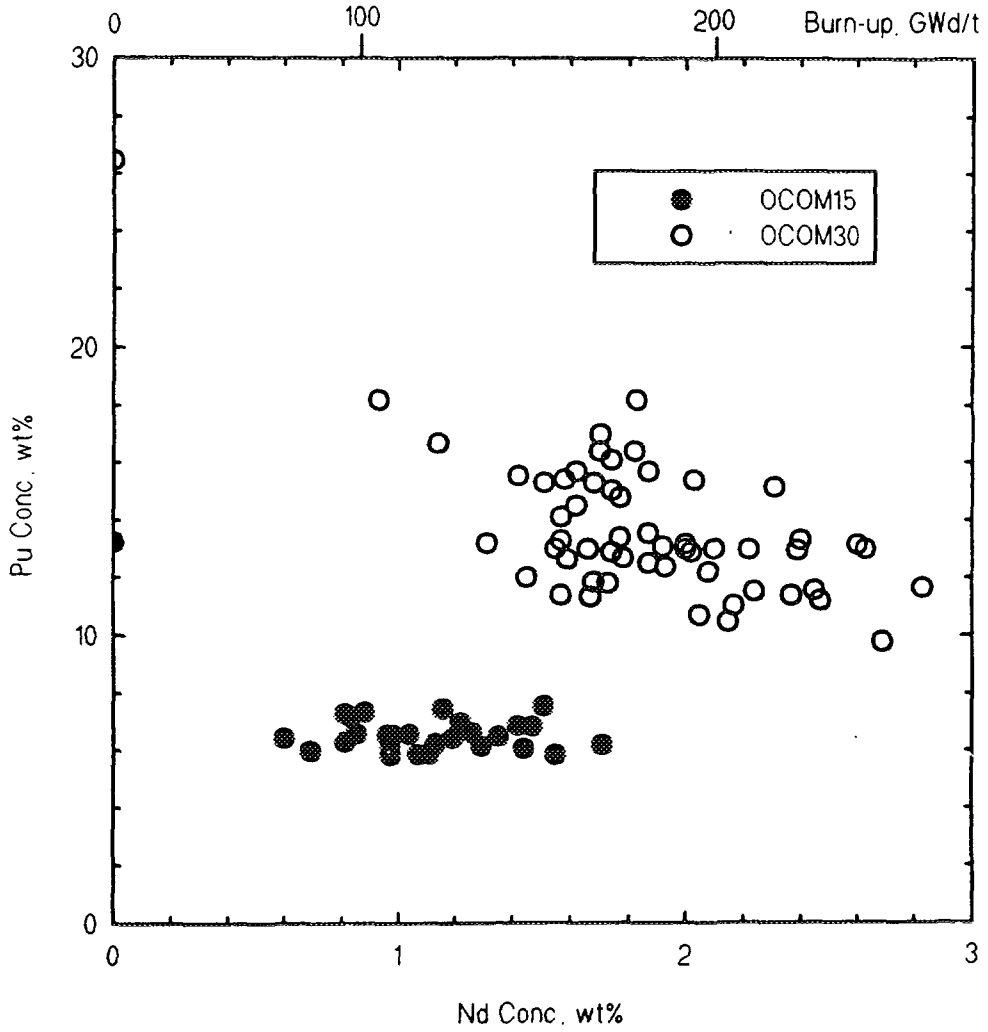

Fig.6. Plutonium concentration in the MOX agglomerates related to the neodymium concentration representing the burn-up at the same location. In both fuels approximatetly 50% of the plutonium in the agglomerates was burnt.

the MOX agglomerates. From this figure, it is evident that the concentration of xenon measured in the agglomerates was generally less than 0.6 wt%, and on average about 0.3 wt%, regardless of the neodymium concentration. This reveals that gas release increased with the agglomerate burn-up. From which it can be deduced that the OCOM15 agglomerates released less gas than the OCOM30 agglomerates, and that MOX agglomerates situated close to the fuel surface, released more gas than those further in.

It is not clear what percentage of fission gas was released from the UO_2 matrix. As seen from Table 3, EPMA measured 0.52 wt% xenon in the matrix, which is considerably more than the amount produced during irradiation to 31.1 GWd/t (0.42 wt%). This inconsistency probably arises because the burn-up in the UO_2 matrix varied from grain to grain (see Fig.5) and xenon and neodymium were analysed at different times. Consideration of the fuel temperature during the irradiation, however, reinforces the view that little gas was released from the UO_2 matrix. This assumption is supported by the the radial xenon profile shown in fig.4. which indicates that at the most 14% release may have occurred locally in the central region of the fuel between $r/r_o = 0.2$ and the centre of the pellet.

Table 3 Fission gas release data for the OCOM30 fuel

Radial Position	Xe Created (wt%)	Xe Measured [a] (wt%)	Percentage Release
	MOX Agglomerates + UO_2 Matrix		
	0.59	0.52	11.9
	MOX Agglomerates		
r/r_o= 0.86	2.59	0.20	92.3
r/r_o= 0.28	2.18	0.28	87.2
	UO_2 Matrix [b]		
	0.42	0.52	

[a] Average values.
[b] The inconsistency is assumed to be due to variations in the local burn-up in the UO_2 matrix (see section 4.2).

4.6 The behaviour of fission product caesium

Caesium behaved very differently from xenon in that it was almost completely retained by the MOX agglomerates. In Fig. 9 the concentration of caesium in the MOX agglomerates of the OCOM15 and OCOM30 fuels is plotted as a function of the neodymium concentration. Despite the scatter in the data it is clearly seen that at neodymium concentrations up to 2.3 wt% (burn-ups up to 215 GWd/t) the concentration of caesium increased linearly with the neodymium concentration indicating complete retention by the MOX agglomerates. At neodymium concentrations above 2.3 wt%, the measured caesium concentration lies below the line drawn through the data indicating some caesium was released from the agglomerates at the very highest burn-ups (215-270 GWd/t).

In the UO_2 matrix of the OCOM30 fuel a caesium concentration of 0.28 wt% was measured. This corresponds to the ceasium inventory calculated using the fission yield value of 0.17 at% caesium per 1 at% burn-up derived from the EPMA data plotted in Fig.9. It would appear, therefore, that caesium was not released from the UO_2 matrix.

5. DISCUSSION

5.1 Similaries between the MOX agglomerates and the high burn-up structure at the rim of conventional UO_2 fuel.

The Pu-rich agglomerates in the outer region of both fuels share several common features with the high burn-up structure at the rim of conventional UO_2 fuel. For example, the MOX agglomerates and

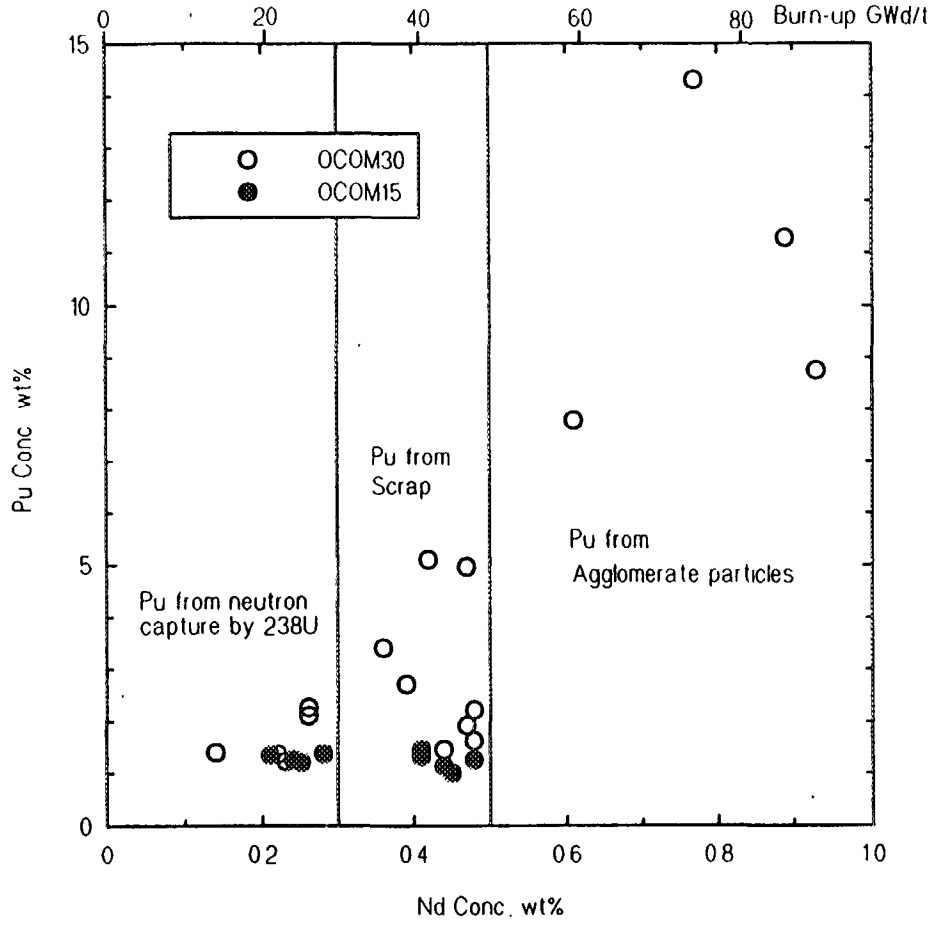

Fig.7. Concentration of plutonium in single grains in the UO_2 matrix in the OCOM15 and OCOM30 fuels related to the concentration of neodymium representing the burn-up in tthe same grains.

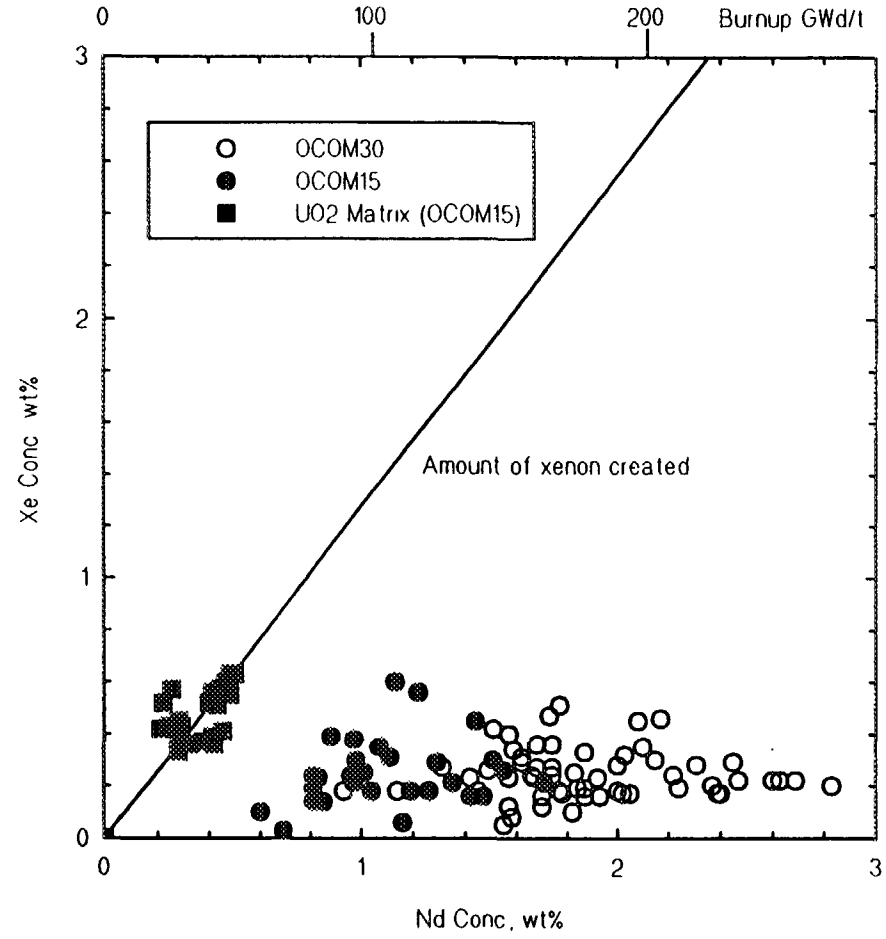

Fig.8. Concentration of retained xenon in the MOX agglomerates related to the local concentration of neodymium representing the burn-up. The local concentrations of xenon and neodymium in the UO_2 matrix in the outer region of the OCOM15 fuel between $r/r_0 = 0.7$ and the fuel surface were used to position the line giving the created amount of xenon, and are included for comparison.

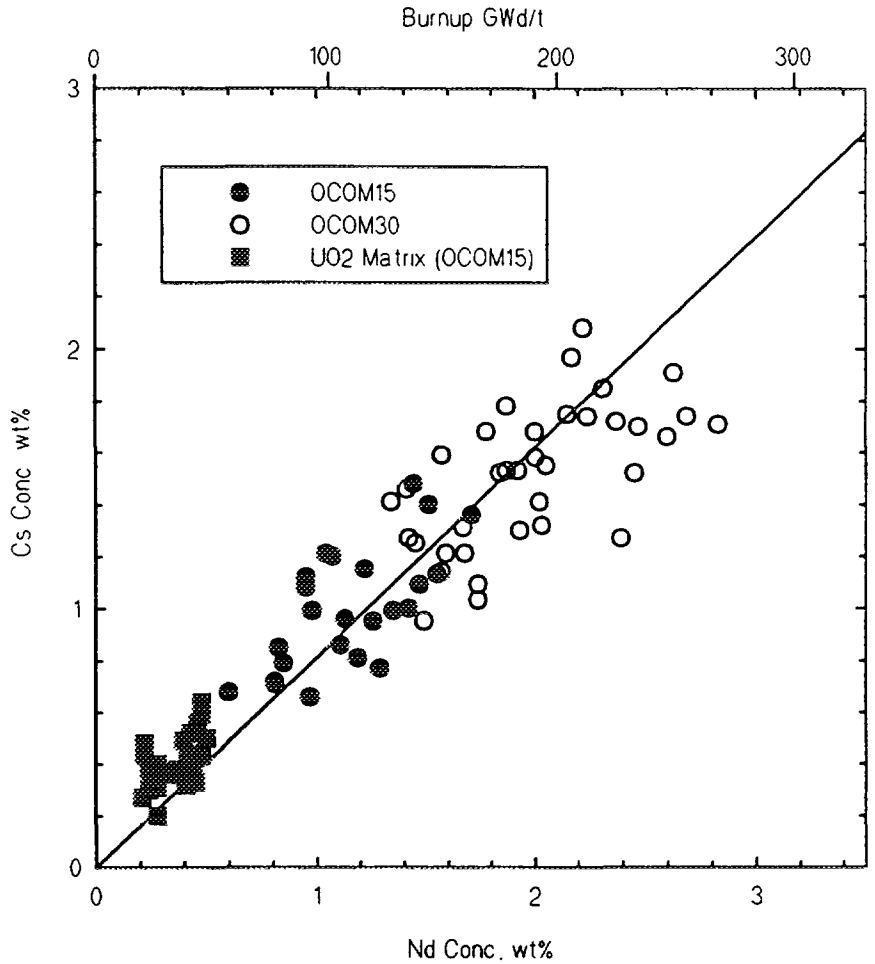

Fig.9. Concentration of retained caesium in the MOX agglomerates related to the local concentration of neodymium representing the burn-up. Over most of the range of neodymium concentrations measured, the xenon concentration increases linearly indicating full retention. . Data for the UO_2 matrix in the outer region of the OCOM15 fuel are included for the purposes of comparison.

the high burn-up structure both consist of fine grains < 1 μm in size and small faceted pores typically 1-2 μm in diameter (cf., SEM micrographs in Fig.10). Moreover, like the high burn-up structure in UO_2 fuel (see Ref.[7]), the agglomerates exhibit strong xenon depletion, with concentrations down to 0.3 wt%, and almost complete retention of caesium. Hence, it is reasonable to postulate that in both cases the mechanisms producing the characteristic microstructure are the same. If this is so, the finding that large pores are contained in MOX agglomerates situated close to the fuel surface, may provide an insight into the development of the high burn-up structure in UO_2 fuel.

5.2 Formation of the high burn-up structure at the rim of MOX fuel

It has been found [7] that at the rim of conventional UO_2 fuel the high burn-up structure forms when the local burn-up exceeds 60 to 75 GWd/t. Generally, local burn-ups of this magnitude are attained in the outer region of the fuel when the average cross-section burn-up reaches 40-45 GWd/t. Thus, the burn-up of both the OCOM15 and OCOM30 fuels was within the range at which the high burn-up structure would be expected to form in UO_2 fuel.

In the UO_2 matrix of the OCOM30 fuel the high burn-up structure may have just started to form at some places at the pellet rim. The high burn-up structure was evidently not present where the xenon profile shown in fig.4 was measured, since close to fuel surface the xenon concentration increased with the burn-up. However, at a second location, the fuel microstructure within 10-20 μm of the pellet surface

may just have begun to transform, because the concentration of xenon measured fell slightly at the fuel surface. The existence of patches of high burn-up structure around the fuel rim would be consistent with the measured burn-up at the fuel surface of 62 GWd/t. This burn-up is at the lower end of the range for the formation of the high burn-up structure in conventional UO_2 fuel.

5.3 Fission Gas Release

The release of fission gas to the rod free volume requires a system of escape tunnels along the grain boundaries in the UO_2 matrix. Because the fuel irradiation temperature was low, such a tunnel system did not develop in the OCOM15 and OCOM30 fuels. Hence, the UO_2 matrix behaved like a hard shell enclosing the agglomerates, and preventing the gas released there from escaping to the rod free volume. The released gas, therefore, has no option but to precipitate in the pore structure of the agglomerate.

More than 90% of the gas had been released from the lattice of the MOX agglomerates and had precipitated in its pore structure. Although the burn-up in the OCOM30 agglomerates was almost twice as high as that of the OCOM15 agglomerates, the percentage of fission gas released from the oxide lattice of the agglomerates was not significantly different (90% compared with 85%). In absolute terms, however, considerably more gas was lost from the lattice of the OCOM30 agglomerates, because these had a much higher burn-up. At the end of the irradiation generally around 0.3 wt% xenon remained in the agglomerate lattice. This concentration corresponds to the limit of solubility of xenon in irradiated UO_2 at temperatures of 700 to 1000°C [8].

For the OCOM30 fuel an average cross-section release value of 11.7% was obtained from area analysis by EPMA. Most of this gas is assumed to have come from the UO_2 matrix in the central region of the fuel. However, the puncturing result for this segment (see Fig.11) revealed that only around 2.5% of the fission gas inventory was released to rod free volume at an average burn-up of 44.5 GWd/t. This indicates that almost 80% of the gas released from UO_2 lattice was retained in the fuel.

The puncturing result for the OCOM15 segment with an average burn-up of 43.6 GWd/t was 1.7%. Thus, despite the difference in the porosity structure of the OCOM15 and OCOM30 agglomerates, the percentage of gas released to rod volume in both segments was practically the same.

5.4 Effect of fuel inhomogeneity

During the irradiation of the OCOM15 and OCOM30 fuels low temperatures prevailed and consequently U/Pu interdiffusion was minimal. Thus, both fuels remained inhomogeneous on the microscopic scale. The MOX agglomerates in the central region of the fuel did not lose Pu to the UO_2 matrix, and initial variations in the plutonium concentration of the grains in the UO_2 matrix were not smoothed out. In both fuels, however, the concentration of plutonium in the MOX agglomerates decreased by about 50% as result of fission, whereas the plutonium content of the UO_2 matrix increased four-fold to about 2 wt% due to neutron capture by ^{238}U. Consequently, the total concentration of plutonium fell to 3.6 wt% in the OCOM30 fuel (a decreased of 18%) and to 3.2 wt% in the OCOM15 fuel (a decrease of 27%).

Although the volume fraction of the MOX agglomerates, and the burn-up in the agglomerates were entirely different in the OCOM15 and OCOM30 fuels, there was no significant difference in the level of release from the two fuels. This is evident from the puncturing data in Fig. 11, which reveal that for both fuels the percentage of gas released to the rod free volume increased from less than 0.5% at 10 GWd/t to a maximum of 3.5% at 45 GWd/t. Perhaps the low percentage of gas measured is surprising in view of the extremely high percentage of gas that was released from the MOX agglomerates. But the puncturing results reflect the fact that most of the gas released from the agglomerate lattice remained trapped in the agglomerates, being unable to penetrate the dense UO_2 matrix. To this can be added the fact that the percentage of gas released by the UO_2 grains of the matrix was relatively small (probably about 10% for both fuel) and that a large fraction of this gas was retained in the fuel.

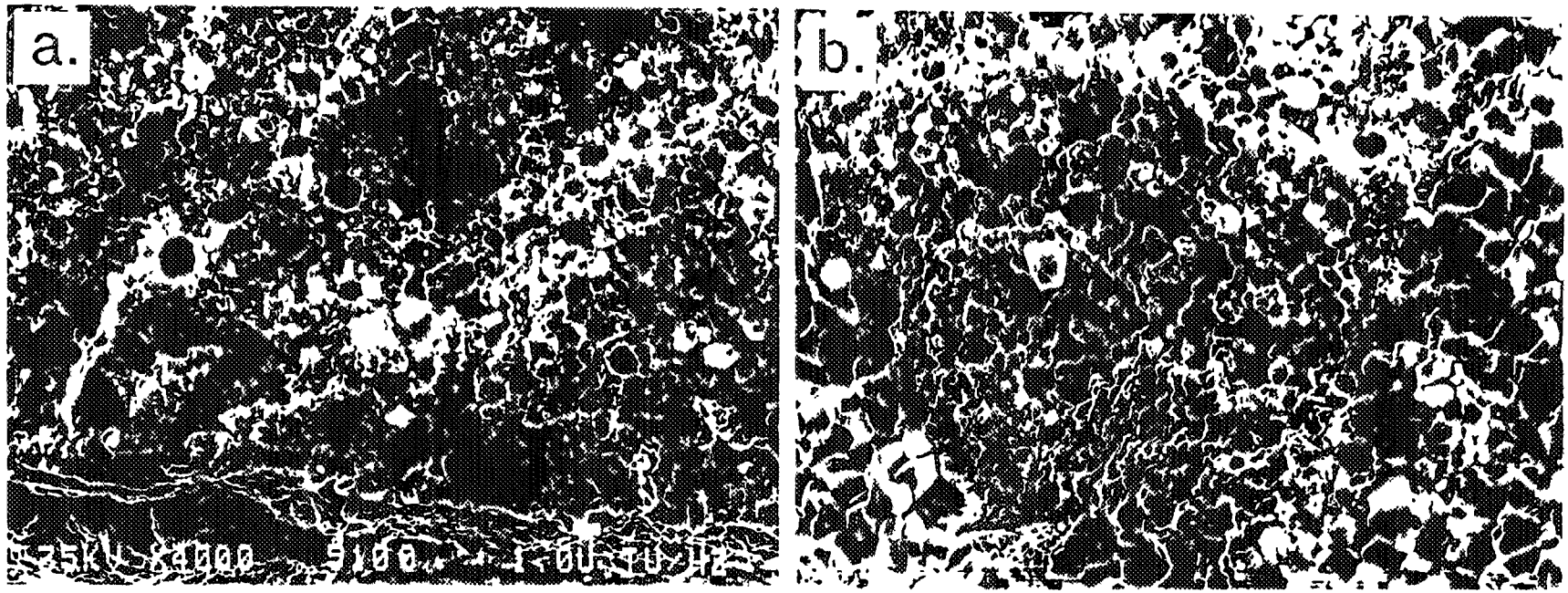

Fig 10 Scanning electron micrographs showing the high burn-up structure a) At the rim of a conventional UO_2 fuel irradiated to 45 2 GWd/t b) In a Pu-rich agglomerate at r/r_0= 0.85 in a MOX fuel irradiated to 38 8 GWd/t. Fine grains and small faceted pores are seen in both micrographs

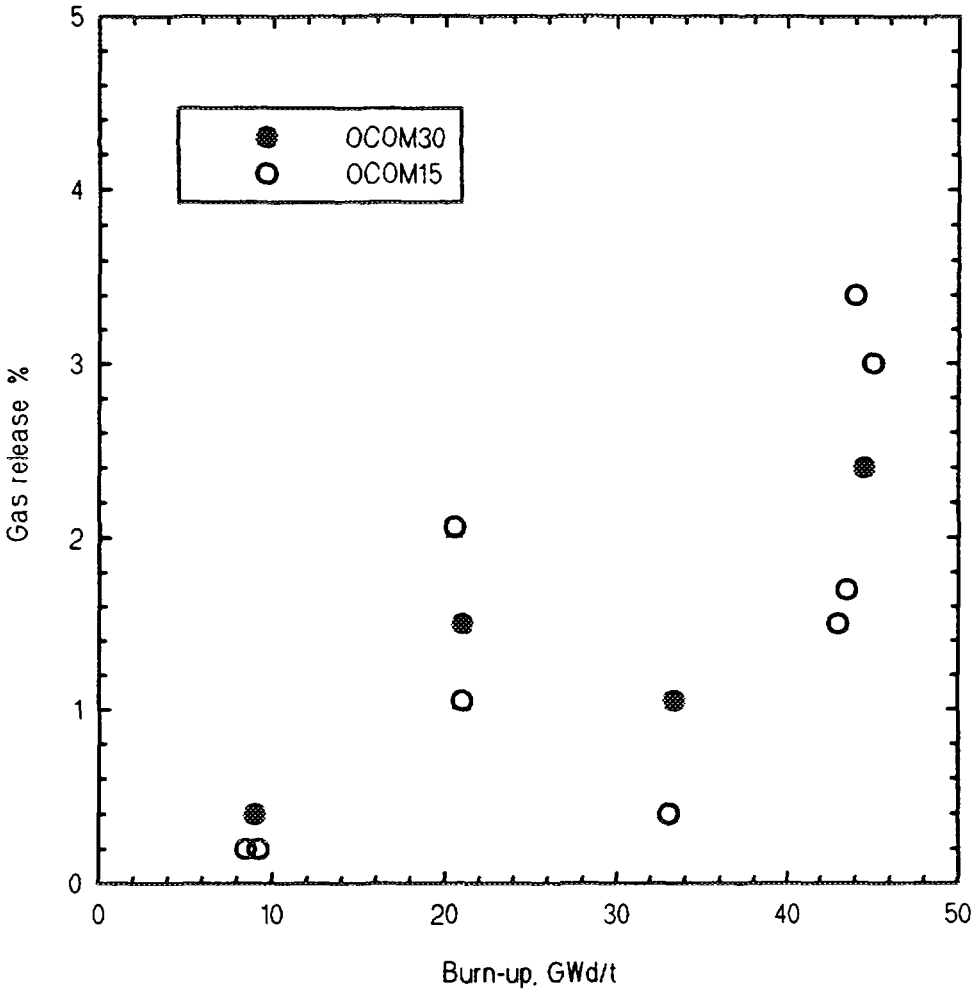

Fig.11. Puncturing results for percentage of fission gas released from the OCOM15 and OCOM30 fuels.

Finally a brief word about caesium. About twice as much caesium was produced in the OCOM30 agglomerates as in the OCOM15 agglomerates due to the difference in agglomerate burn-up and apparently all this was retained. However, since the volume fraction of the agglomerates was twice as high in the OCOM15 fuel as in the OCOM30 fuel, and the average, burn-up of the two fuels was almost the same, both probably contained aproximately the same total amount of caesium.

6. SUMMARY AND CONCLUSIONS

The MOX agglomerates in the outer region of the OCOM15 and OCOM30 fuels share several common features with the high burn-up rim structure of conventional UO_2 fuel; namely, small grains, small faceted pores, high xenon release from the oxide lattice and almost complete retention of caesium. As in conventional UO_2 fuel, the high burn-up structure seems to form in the UO_2 matrix when the local burn-up at the pellet rim exceeds 60-75 GWd/t. At the surface of the OCOM30 pellet the burn-up in the UO_2 matrix was 62 GWd/t, and patches of high burn-up structure may have existed around the fuel circumference.

The burn-up in the OCOM30 agglomerates was almost twice as high as in the OCOM15 agglomerates (e.g., at the fuel surface, typically 270 GWd/t compared with 160 GWd/t). As low temperatures prevailed during the irradiation, U/Pu interdiffusion did not occur. Because of this, both fuels remained inhomogeneous on the microscopic scale. The concentration of plutonium in the MOX

agglomerates decreased by about 50% as a result of fission, whereas the plutonium content of the UO_2 matrix increased four-fold to about 2 wt% due to neutron capture by ^{238}U. As a result, the total concentration of plutonium in the fuel fell to 3.6 wt% in the OCOM30 fuel (a decreased of 18%) and to 3.2 wt% in the OCOM15 fuel (a decrease of 27%). The main difference between the agglomerates in the two fuels was that those in the OCOM15 fuel generally exhibited a finer pore structure due to the lower burn-up.

More than 80% of the fission gas had been released from the oxide lattice of the MOX agglomerates in both fuels leaving at the most 0.6 wt% and on average 0.3 wt% in solution. A very high fraction of the gas released from the lattice precipitated and remained in the pore structure of the agglomerates. Although the burn-up in the OCOM30 agglomerates was considerably higher than in the OCOM15 agglomerates, the percentage of fission gas released from the agglomerates was not significantly different (90% compared with 85%).

Puncturing revealed no significant difference in the percentages of gas released to the free volume in the OCOM15 and OCOM30 fuel segments. For both fuels the percentage of gas released to the rod free volume increased from less than 0.5% at 10 GWd/t to a maximum of 3.5% at 45 GWd/t. This reveals that the percentage of gas released by MOX fuel manufactured by the OCOM process is largely independent of the level of inhomogeneity of the fuel.

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BEHAVIOUR OF A DEFECTIVE MOX FUEL ROD IN A PWR

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Abstract

At the end of the DAMPIERRE 1 power plant 11th cycle in 1993, routine sipping techniques identified a leak on a mixed oxide (MOX) fuel assembly. Application of failed fuel management policy led to the reloading of this assembly for the 12th cycle. It was its second irradiation campaign. This situation allowed EDF and CEA to set up a special monitoring programme during the cycle in order to obtain numerous information about the behaviour of defective MOX fuel at steady state power levels and during transients. These data have been obtained due to: a very good knowledge of its power history; the daily routine measurements of the primary circuit activity, conducted by the plant operator; a specific on-line gamma spectrometry facility installed by CEA to monitor the primary water during this cycle; various on-site post-irradiation examinations carried out at the end of the 12th cycle : visual inspection, qualitative and quantitative sipping tests, defective fuel rods detection and localisation. Analyses of this behaviour were mainly in relation with two scopes: determination of the gaseous and airborne fission product release rates out of a defective MOX stack, in relation with theorical prediction (PROFIP code) and with results obtained on experimental similar rods in a research reactor (EDITHMOX 01 experiment); application of on-line discrimination methods developed by EDF and CEA in order to know the type (uranium oxide UO_2 or mixed oxide) of fission product source. Based on measurement of gaseous isotopes, these methods allow plant operator to detect very early a possible evolution of the defect and to foresee on-site examinations during shutdown period. The main results of this programme were: release rates of gaseous fission product were similar to those observed with defective UO_2 fuel; no worsening of the defect size or of activity release occurred over one year of irradiation; discrimination methods have been in good agreement with the type of fuel.

1. INTRODUCTION

Plutonium recycling first started in France in 1987 with the 5th reload of SAINT LAURENT B Unit 1. At the beginning of 1995, 424 mixed oxide fuel assemblies (MOX assemblies) have been loaded in the seven reactors which operate with this fuel. Table I shows the history of these irradiation cycles and the number of MOX fuel assemblies loaded in each reactor.

To date, 200 three cycles irradiated fuel assemblies have been discharged with an average burnup of 37,5 GWd/tU and a maximum burnup of 40 Gwd/tU. Four MOX fuel assemblies, irradiated for a fourth cycle on the periphery of the core of GRAVELINES Unit 4, have achieved a burnup of 44 Gwd/tU. These assemblies have been discharged in March 1994.

TABLE 1 :

EDF OPERATING EXPERIENCE FEEDBACK WITH MOX FUEL

Unit with MOX	1987	1988	1989	1990	1991	1992	1993	1994
SLB1	C5 16 MOX	C6 16 MOX		C7 16 MOX	C8 16 MOX	C9 16 MOX	C10 16 MOX	(C11) 16 MOX
SLB2		C6 16 MOX	C7 16 MOX		C8 16 MOX	C9 16 MOX	C10 16 MOX	(C11) 16 MOX
GRA3			C8 16 MOX	C9 16 MOX	C10 16 MOX	C11 16 MOX	C12 8 MOX	(C13) 16 MOX
GRA4			C8 16 MOX	C9 8 MOX	C10 16 MOX	C11 16 MOX		(C12) 16 MOX
DAM1				C9 16 MOX	C10 /	C11 16 MOX	C12 /	(C13) /
DAM2							C12 16 MOX	(C13) 16 MOX
BLA2								(C12) 8 MOX
Total cycles	1	2	3	4	5	5	5	7
Total MOX F/A	16	32	48	56	64	80	40	88

(C11) Current cycle

January, 1995

Until now, the behaviour of MOX fuel assemblies has been completely satisfactory. The first failure on a MOX fuel rod since its introduction in reactor has been identified at the end of cycle 11 of DAMPIERRE Unit 1 in July 1993.

2. DAMPIERRE UNIT 1 OPERATION DURING CYCLE 11

2.1. Primary coolant activity evolution

The core was constituted during that cycle of 32 MOX assemblies and 125 UO_2 assemblies. The cycle has started with one UO_2 failed fuel assembly, reloaded for its third irradiation cycle, according to EDF leaking management policy described in reference [1].

Figures 1 and 2 show the specific gamma activity evolutions of noble gases and iodines in the primary coolant during the cycle 11, from September 1992 to June 1993. They show also power history of the reactor during the cycle and purification flow rate through the Chemical and Volumetric Control System.

- During the first weeks following starting, activities are representative of those released by the reloaded Uranium failed assembly. The iodines specific activity levels are stabilized at about 50 Mbq/t. The noble gases specific activity levels are below 3000 MBq/t. For ^{133}Xe, radioactive equilibrium is not yet reached.

- After a few weeks of operation, a rapid increase of all activities, followed by a stabilisation at an elevated level, indicates the development of new leaks. The iodine spiking phenomenon observed with a slow dynamic is typical of failure developed on fresh fuel.

A second slighter step of gamma activities can be also observed about middle of March 1993. At the end of the 11^{th} cycle, the sum of noble gases activity reached 30 000 MBq/t. During all the cycle, the low level of short-lived iodines isotopes activity means a failure without dissemination of fissile matter into primary circuit.

2.2. Poolside examinations at the end of the 11^{th} cycle

In mast sipping test performed on-line during off-loading (see reference [2]) identified three leaking fuel assemblies :

- the previous leaking UO_2 fuel assembly reloaded for its third cycle,
- one new leaking UO_2 fuel assembly, at the end of its third irradiation cycle,
- one MOX fuel assembly, at the end of its first irradiation cycle.

Location and power history of the defective assemblies are given in Figure 3.

The MOX fuel assembly was tested by quantitative sipping test (see the presentation of this method in reference [3]). The equivalent diameter of the defect was smaller than 20 m. Poolside TV inspection performed on that assembly has shown no anomaly. The fuel defect origin was unknown to date.

According to EDF leaking fuel management policy, the MOX assembly, which performed the reloading criteria, was re-inserted for a second cycle of irradiation. The two UO_2 leaking assemblies, which have finished their irradiation, were discharged.

Consequently, the 12^{th} cycle of DAMPIERRE Unit 1 started with one defective MOX assembly in August 1993. This cycle was a very interesting case for application of defective MOX fuel discrimination methods (see § 4).

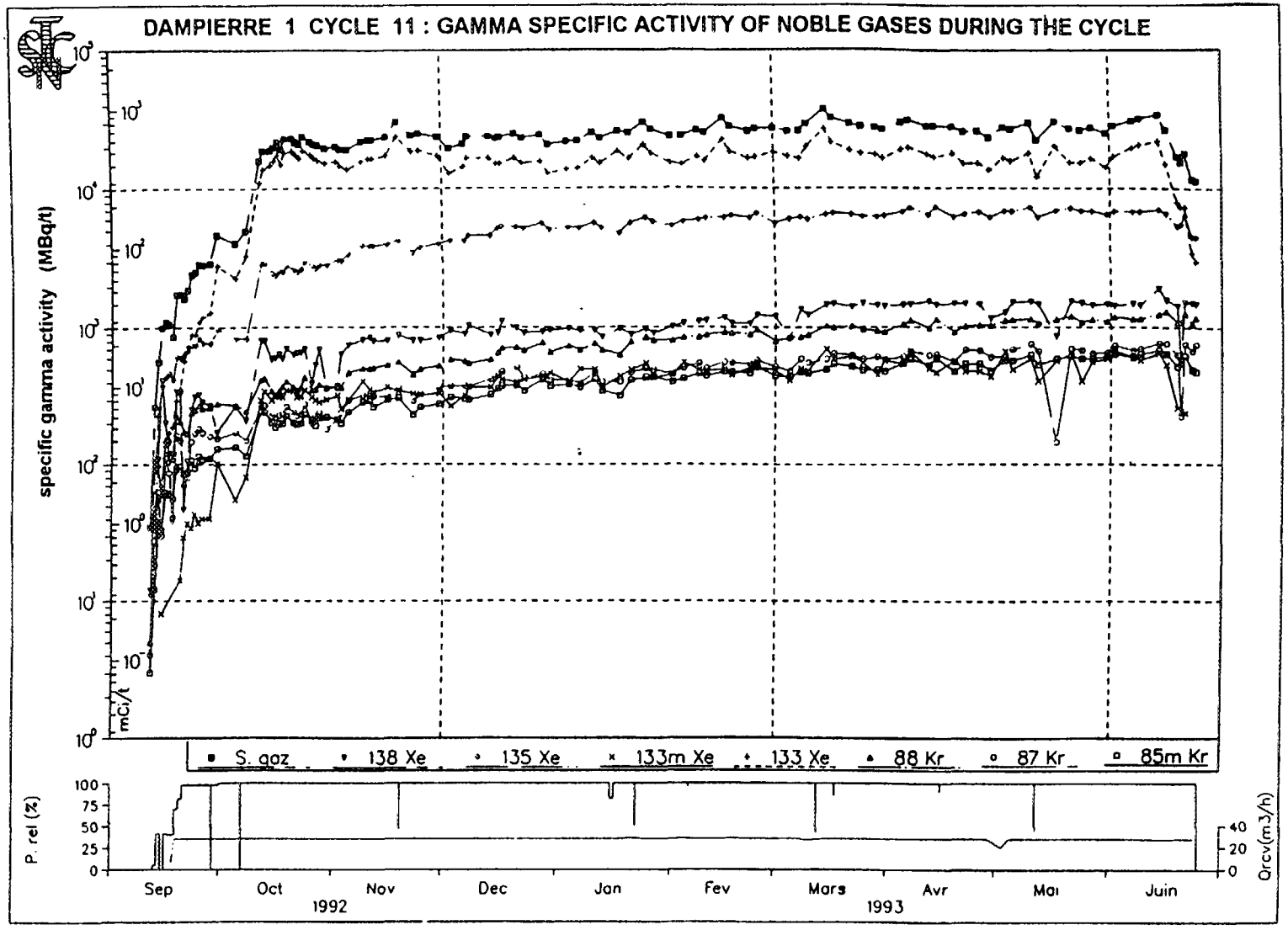

FIG. 1. *Specific gamma activity of noble gases during cycle 11 of DAMPIERRE Unit 1.*

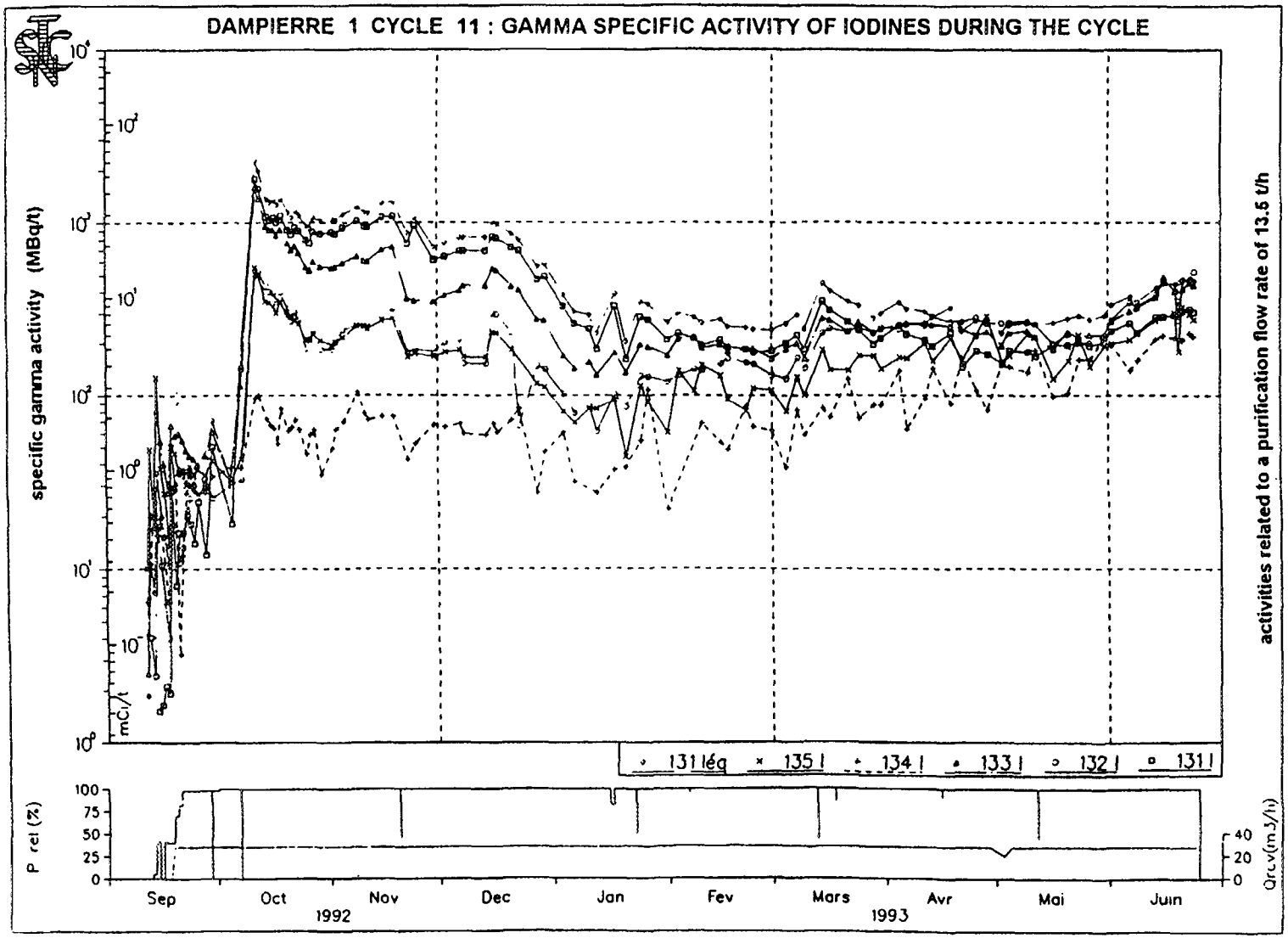

FIG. 2. Specific gamma activity of iodines during cycle 11 of DAMPIERRE Unit 1.

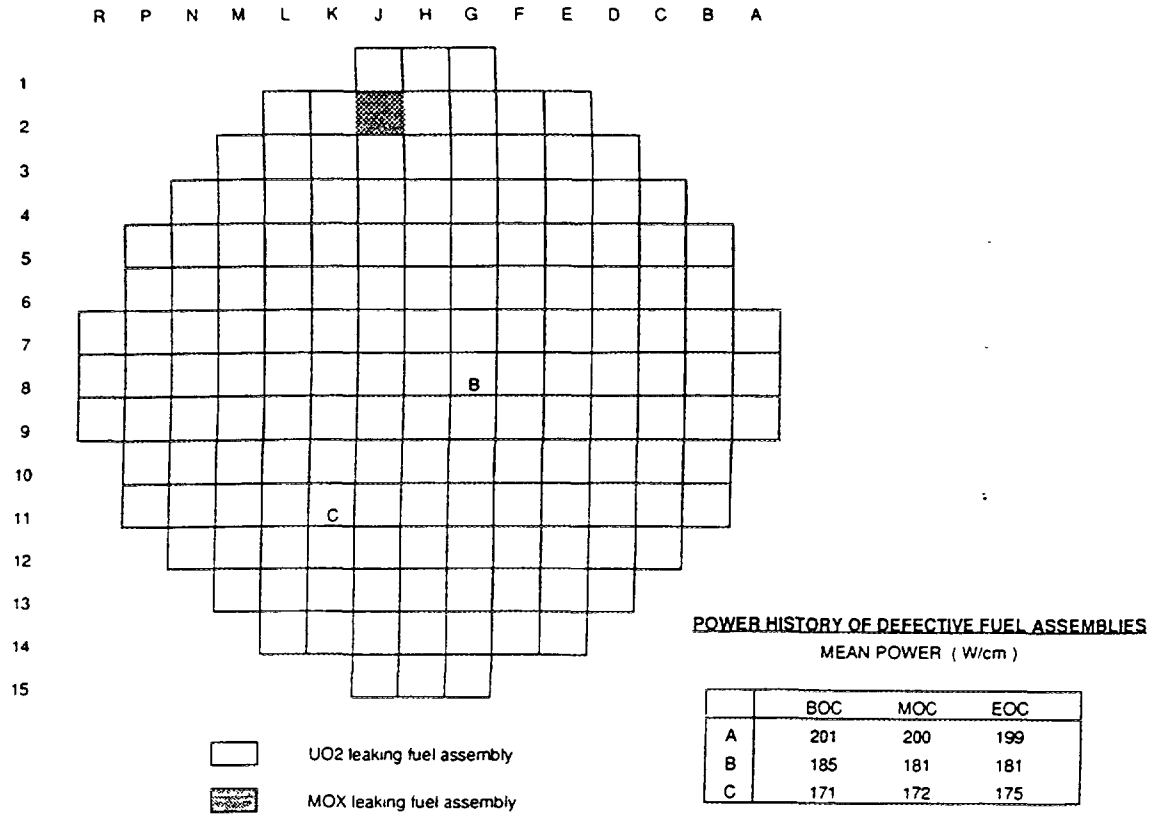

FIG. 3. Location of leaking fuel assemblies in core during cycle 11 of DAMPIERRE Unit 1.

3. DAMPIERRE UNIT 1 OPERATION DURING CYCLE 12

3.1 Equipment installed and specific monitoring of cycle 12

The fission product activity in the primary water of DAMPIERRE 1 was monitored on line by the CEA/DRN/DEC at the request of EDF/SEPTEN, with the dual aim of qualifying methods for discriminating UO_2/MOX in leaking fuel rods, and improving experimental feedback on fission product release from leaking MOX fuel rods. The experimental device was operated on the DAMPIERRE 1 site from 8/12/1993 to 1/09/1994 during virtually the entire cycle 12.

3.1.1 Measuring instruments :

The EMEL on-line gamma spectrometry device consists of the following components:

- a planar HP Germanium diode of 8 cm^3 volume, equipped with a cryostat, a copper collimator and additional steel protection. The diode is placed opposite an internally electro-polished stainless steel channel of 3.3 cm outside diameter, conveying the primary fluid. The entire assembly is fixed to a steel frame (see Fig. 4),
- an electronic metering system,
- a microcomputer, loaded with the necessary gamma spectrum analysis software.

The primary water circulating in the EMEL device is drawn off in the REN (sampling circuit) room from loop 3 and is returned to the RCV (Volume Control Tank). The water flow rate in the device is of the order of 200 l/h at a pressure of 3.5 bars.

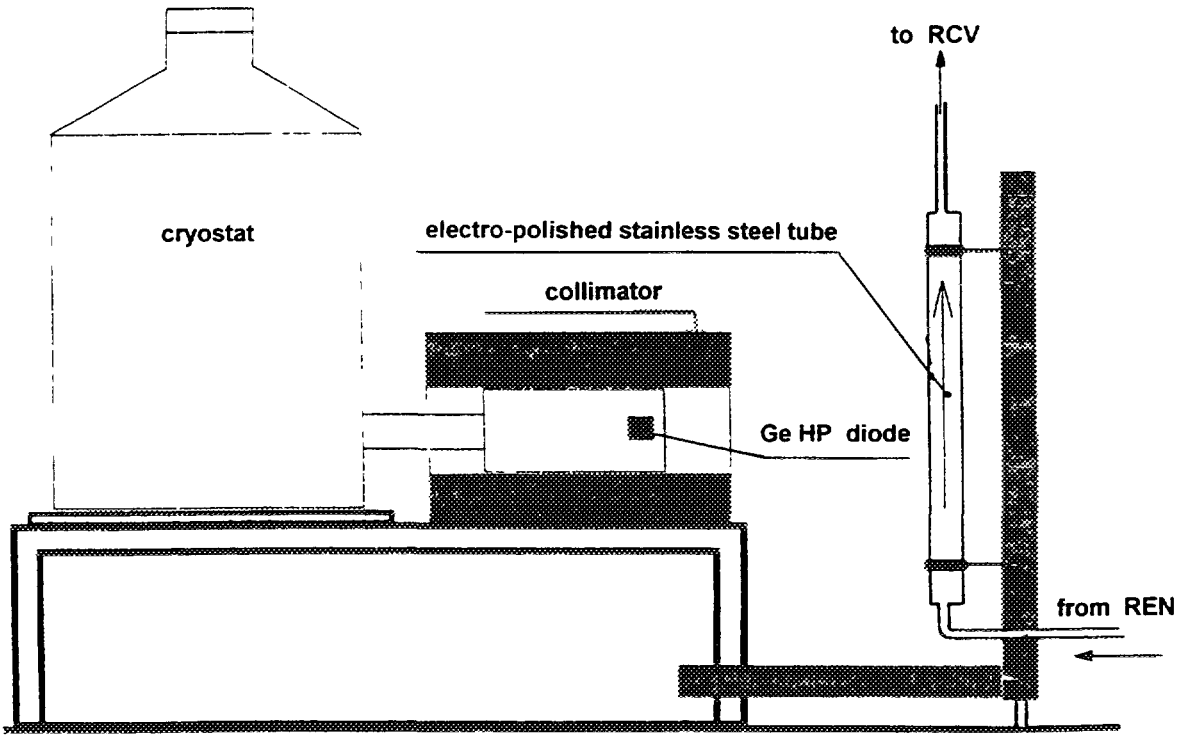

FIG. 4. On-line gamma spectrometry measurements device "EMEL".

3.1.2 Measurement characteristics :

The gamma spectrometry system operates over a one-hour cycle which includes:

- an acquisition time of 2700 s,
- spectral analysis and storage of results on the computer hard disk,
- a standby period until the start of the next one-hour cycle.

The spectra are analysed with respect to a table of radionuclides comprising 36 bodies with, in particular, all the radioactive isotopes of rare gases, iodine and caesium isotopes with radioactive periods greater than 1 min. This operating method can also be used to monitor accurately the variations in fission product activities in the primary water during power transients.

3.2 Primary coolant activity evolution during cycle 12

From the start-up, the fission product gamma activity levels increased rapidly, confirming the presence of the failed MOX assembly, then remained stable to the end of cycle 12 at a value slightly higher than at the previous cycle. Figures 5 and 6 provide the activity evolutions for noble gases and iodines during cycle 12 operation, from August 1993 to July 1994. No significant indication of additionnal leaks can be shown during this cycle. The reactor was shutdown from 5 January to 20 February due to an intervention on the main generator.

The low gamma activity levels of short lived isotopes such ^{138}Xe or ^{134}I show the good behaviour of the failed rod and the absence of evolution of the defect size during the cycle.

3.3 Poolside examinations performed at the end of cycle 12

In mast sipping performed on-line during core unloading detected the previous leaking MOX assembly again. However, a problem concerning the calibration of the single-channel gamma analyser

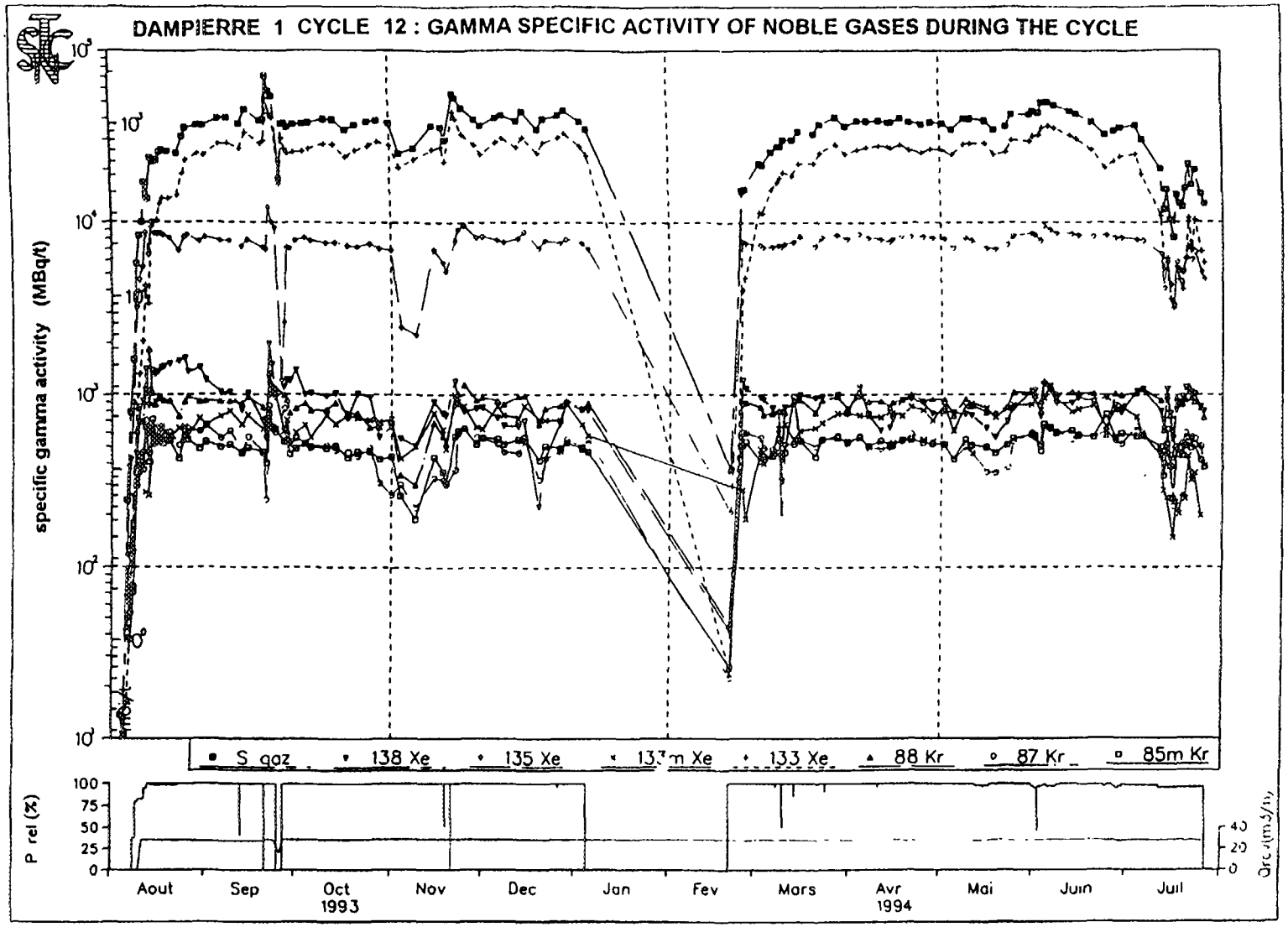

FIG. 5. *Specific gamma activity of noble gases during cycle 12 of DAMPIERRE Unit 1.*

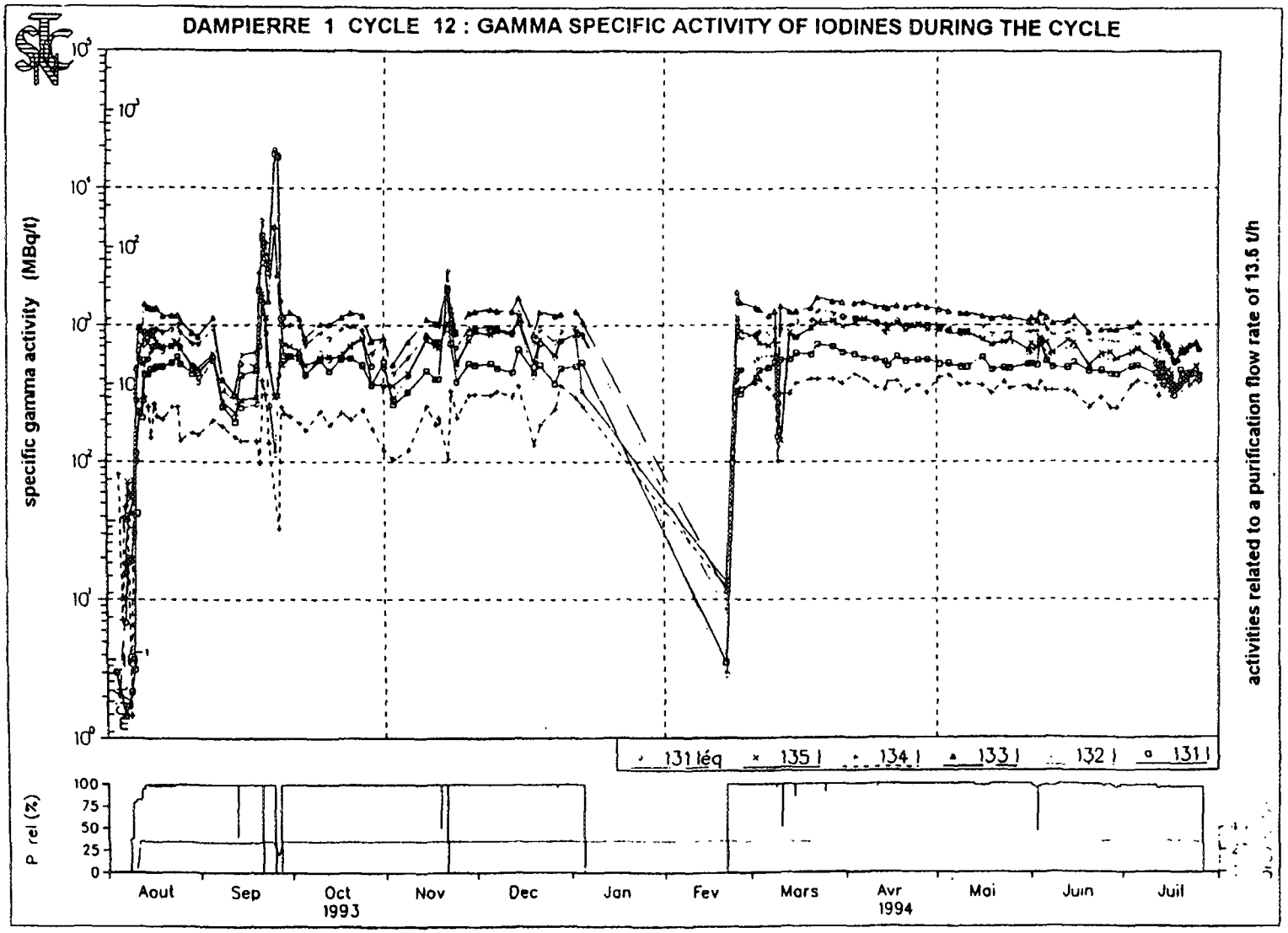

FIG. 6. *Specific gamma activity of iodines during cycle 12 of DAMPIERRE Unit 1.*

during control sequence did not enable to be sure that all leaking fuel assemblies were identified. The location and power history of the defective assembly during cycle 12 is given in Fig. 7.

The MOX assembly was tested by quantitative sipping test. The equivalent diameter measured was about 20 μm and close to the size measured at the end of the previous cycle.

The MOX assembly was not reloaded for the 13th cycle and will be repaired before reloading for a third cycle of irradiation. Before this, it has been tested recently by Ultrasonic device in order to determine the number and the position of leaking rods. Two neighbouring rods were identified as leaking. The repair and poolside examinations of the failed rods are scheduled to be carried out in July 1995.

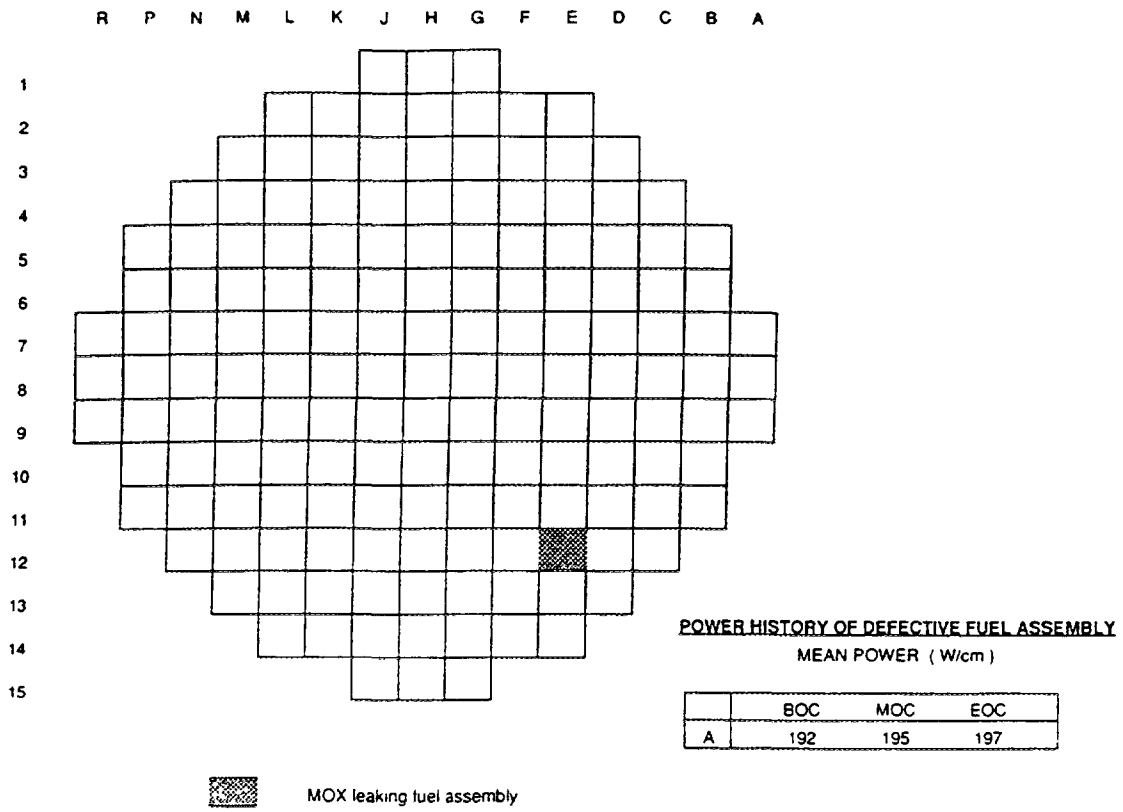

FIG. 7. Location of leaking fuel assemblies in core during cycle 12 of DAMPIERRE Unit 1.

3.4 Analysis of poolside results in relation with activity released during operation

The results developed above permit to formulate two conclusions :

- they confirm the absence of defect size evolution during the cycle 12 and the good behaviour of the reinserted defect,
- comparison with typical activities of middle- and long-life fission products released out of a failed uranium rod (see reference [4]) shows that the DAMPIERRE 1 MOX failure has induced an increase of coolant activity within the range observed with a failure on an uranium rod.

4. ANALYSIS OF ACTIVITY RELEASE DURING CYCLE 12

Fission product levels observed during the 12th cycle of DAMPIERRE Unit 1 in the primary circuit are compared to :

- irradiation results of an experimental leaking MOX rod in an experimental loop : the EDITHMOX 01 experiment,
- calculations of gamma specific activities thanks to the PROFIP 4 code.

4.1 The EDITHMOX 01 experiment :

This irradiation concerned an experimental fresh MOX fuel rod (length about 500 mm) with an 0,3 mm in diameter artificial defect drilled in front of the pellet stack. This irradiation took place in the JET POMPE loop at the SILOE research reactor of the CEA's Grenoble Nuclear Research Center.

This programme permitted to obtain the release rates of fission products (noble gases and halogens) out of the mixed oxide, at the beginning of life (burn-up less than 2 GWd/tU), for steady state power levels within the range of 8,5 - 26 kW/m.

Results obtained (see reference [5]) show than release rates of noble gases, and probably iodines, out of a fresh pellet stack are similar to those observed out of a fresh UO_2 one.

4.2 PROFIP 4 code calculations

The PROFIP code (ref. [6]) is used to calculate releases and activities induced in the primary cooling circuit of PWRs by fission products released out of leaking fuel rods, and to determine the residual contamination in fissile matter deposited on the fuel cladding.

This code is perfectly qualified with respect to volatile fission product release (noble gases, iodines and caesium isotopes) for leaking UO_2 fuel.

The code has been used for Dampierre 112, taking into account the results of examinations after fuel irradiation (see § 3.3) :

- number of defective rods: 2,
- burn-up reached at end of cycle 12 : 24 GWd/tU,
- linear power of defective rods at cycle 11: 15 kW/m,
- linear power of defective rods at cycle 12: 22,2 kW/m,
- date of defect opening: beginning of cycle 11.

4.3 Comparison between measured and calculated release rates

The comparison between the release rates R/B of gases and iodine isotopes (this rate describes the dependance of the emission level of a radionuclide with its half-life, due to retention time in the fuel or in the gap) calculated by PROFIP and those measured with EDITHMOX 01 and at DAMPIERRE 1 (end of cycle 12) is presented below :

	Noble gases and iodines release rates		
Radionuclide	*EDITHMOX 01 values*	*DAMPIERRE 1 values*	*PROFIP 4 values with two defects*
^{133}Xe	1.3×10^{-1}	1.0×10^{-1}	8.2×10^{-2}
^{135}Xe	2.4×10^{-2}	1.9×10^{-2}	2.1×10^{-2}
^{131}I	1.5×10^{-1}	2.2×10^{-1}	1.1×10^{-1}
^{133}I	4.5×10^{-2}	7.3×10^{-2}	3.1×10^{-2}

There is a good agreement between the measurements and the PROFIP 4 calculated results. It may be concluded that the volatile fission product release rates from the leaking MOX fuel rods of DAMPIERRE 1 are comparable to those from UO_2 fuel rods.

4.4 Conclusion

Fission product activity releases (noble gases and volatiles) out of the two leaking MOX rods detected after the DAMPIERRE Unit 1 cycle 12 shutdown are similar to those observed for UO_2 rods irradiated in the same conditions : burn-up less than 24 GWd/tU, and linear power rate less than 22 kW/m. They are in good agreement with analytical results on fresh MOX fuel too.

No deterioration of the leak (with a small size) occured during its second irradiation cycle, and no significative release of solid fission products or alpha emitters has been observed.

5. PRESENTATION OF DEFECTIVE MOX FUEL DISCRIMINATION METHODS DURING PLANT OPERATION

The situation encountered at DAMPIERRE 1 at the beginning of the 12^{th} cycle permitted for the first time the application of new methods developed in a common EDF (Service d'Etudes et Projets Thermiques et Nucléaires) and CEA (Direction des Réacteurs Nucléaires) Research and Development Programme, concerning the on-line knowledge of the failed fuel kind (MOX or UO_2) by the operator.

5.1 Aims and interest of these methods for plant operator

When a leak occurs on a fuel rod during normal operation, gaseous and airborne fission product are released into primary circuit. Monitoring their specific activities provides then, after analysis, data useful to understanding the fuel behaviour and to forecast its future evolution during the operating cycle, at steady state power levels and during transients.

As MOX fuel contains much more actinides with strong radiotoxicity than UO_2 (e. g.: Plutonium, Americium, Curium), it is important for plant operator to detect very early the type (uranium oxide or mixed oxide) of the defective fuel assembly, specially when the activity level of short half-life fission product (e. g.: ^{134}I or ^{138}Xe) indicates a possibility of fissile matter release out of the defective rod. These conditions provoke plant operator to plan specific provisions for end of cycle (e. g. : complementary controls for radiological protection of workers, specific liquid and solid wastes management, ...).

5.2 Presentation of discrimination methods

All of these methods are based on the difference between :

- fission yield for Uranium (mainly ^{235}U) and Plutonium (mainly ^{239}Pu and ^{241}Pu), for specific radionuclides or for stable isotopes,

or :

- isotopic composition of irradiated Uranium and Plutonium.

In practice the result is given by a ratio between two chosen isotopes. This ratio depends on the average burn-up of the defective fuel rod, and shall be as different as possible between the two kind of fuels. Three methods can be distinguished :

5.2.1 Ratios between two radioactive noble gases with short and middle half-life

All these ratios include one Xenon and one Krypton isotope, because the fission yields of Xenons are equivalent or slightly higher with Pu than with U fissions, while those of Kryptons are more little with Pu than with U (see Table 2, extract from reference [7]). The measurement concerns the specific gamma activity of each isotope directly in the primary circuit water.

Three ratios have been selected :

[1] $\frac{^{135}Xe}{^{85m}Kr}$ [2] $\frac{^{138}Xe}{^{87}Kr}$ [3] $\frac{^{137}Xe}{^{89}Kr}$

Main advantages and disadvantages of these ratios are :

+ measurement with on-site existing means (gamma spectrometry), generally with a good sensitivity and accuracy (low dispersion of counting results),
+ almost immediate result after sampling or on-line following,
+ good "contrast" (i. e. {ratio for a fresh MOX fuel} / {ratio for a fresh UO_2 fuel}): respectively 2,5 for [1] , 2,0 for [2] and 2,9 for [3] ,
- need of on-line gamma spectrometry in order to do routine measurements on ^{138}Xe, ^{137}Xe and ^{89}Kr,
- need to know the release rate R/B of fission product out of the rod, in order to correct the measured ratio,
- necessity of complete understanding of ^{135}Xe behaviour in the core and in the sample: this isotope has a high neutronic capture rate, and a radioactive filiation with ^{135}I.

5.2.2 Ratios between stable Xenon and Krypton isotopes :

The principle is the same as the one used for radioactive noble gases and exposed above (see the values of fission yields on Table 3). The ratio (each term given in volume or mass) taken into account depends of the measurement technique :

TABLE 2 :

DISCRIMINATION METHODS BASED ON RADIOACTIVE NOBLE GASES CUMULATIVE FISSION YIELDS FOR SELECTED RADIONUCLEIDES

(Compilation of fission product yields [B.F. RIDER] - Vallecitos Nuclear Center - 1981)

Isotope	Half-life	CUMULATIVE FISSION YIELD Yc (%)							
		U 235		U 238	Pu 239		Pu 240	Pu 241	
		thermal	fast	fast	thermal	fast	fast	thermal	fast
135 Xe	9.15 h	6,54	6,60	6,96	7,58	7,47	7,44	7,23	7,32
85m Kr	4,48 h	1,29	1,35	0,74	0,60	0,59	0,57	0,41	0,40
138 Xe	14,2 mn	6,23	6,11	5,65	5,18	4,70	5,85	6,22	6,01
87 Kr	76 mn	2,56	2,49	1,61	0,99	1,02	0,99	0,76	0,75
137Xe	3,84 mn	6,13	6,04	6,01	6,05	5,68	6,24	6,61	6,47
89 Kr	3,16 mn	4,56	4,25	2,70	1,43	1,45	1,37	1,17	1,20

TABLE 3 :

DISCRIMINATION METHODS BASED ON STABLE NOBLE GASES
CUMULATIVE FISSION YIELDS FOR STABLE AND LONG-LIFE NOBLE GASES

(Compilation of fission product yields [B.F. RIDER] - Vallecitos Nuclear Center - 1981)

Isotope	CUMULATIVE FISSION YIELD Yc (%)								
	U 235		U 238	Pu 239		Pu 240	Pu 241		Pu 242
	thermal	fast	fast	thermal	fast	fast	thermal	fast	fast
83 Kr	0,54	0,58	0,40	0,30	0,31	0,30	0,20	0,20	0,24
84 Kr	1,00	1,03	0,82	0,47	0,49	0,43	0,35	0,35	0,35
85 Kr (10,7 y)	0,29	0,27	0,15	0,13	0,13	0,13	0,10	0,09	0,09
86 Kr	1,97	1,95	1,29	0,76	0,78	0,76	0,57	0,59	0,65
Σ Krypton ⇨	**3,8**	**3,83**	**2,66**	**1,66**	**1,71**	**1,62**	**1,22**	**1,23**	**1,33**
131 Xe	2,89	3,22	3,24	3,87	3,88	3,54	3,09	3,22	3,19
132 Xe	4,32	4,67	5,16	5,41	5,32	4,81	4,56	4,64	4,57
133 Xe (5,24 d)	6,70	6,75	6,76	7,01	6,91	7,01	6,73	6,69	6,59
134 Xe	7,88	7,67	7,88	7,66	7,38	7,03	7,90	7,72	7,38
135 Xe	6,54	6,60	6,96	7,58	7,47	7,44	7,23	7,32	7,16
136 Xe	6,31	6,23	6,95	7,09	5,94	6,75	7,08	6,81	6,88
Σ Xenon ⇨	**34,64**	**35,14**	**36,95**	**38,62**	**36,9**	**36,58**	**36,59**	**36,4**	**35,77**
129 Xe	0,75	0,88	0,97	1,40	1,55	1,10	0,82	0,99	0,85

- $\{^{\text{sum of stable Xenons}}\}$ / {sum of stable Kryptons}, including ^{85}Kr and ^{133}Xe, if gas chromatography is used,
- ratio of two isotopes (e. g. ^{132}Xe / ^{86}Kr) if mass spectrometry is used.

Main advantages and disadvantages are the following :

+ good sensitivity with mass spectrometry,
+ results available even with a small defect,
+ possibility to determine the burn-up of the defective rod,
+ low distorsion of the ratio in water of the primary and purification circuit,
- necessity of sampling in the gaseous phase of the Volume Control Tank (RCV) and to concentrate the noble gases,
- method tested up to now only on a laboratory mockup of the RCV,
- decay time necessary before the measurement if the gamma activity of the sample is high.

5.2.3 Ratio between two long-life radioactive fission product :

This method concerns the measurement of the gamma specific activity ratio ^{106}Ru / ^{144}Ce by sampling of the cooling water. ^{106}Ru presents a fission yield ten times greater with Plutonium than with Uranium fissions (see Table IV). The contrast between UO_2 fuel and MOX fuel is also very great and easy to determine.

Due to the chemical properties and the release rate of these isotopes, the possibilities of detection and analysis concern mainly the defects which release fissile matter (i. e. with a big equivalent diameter) and their behaviour during a transient.

The long half-life of these radionuclides permits a gamma-counting a long time after the sampling. Nevertheless the background level of the spectrum shall be low. Commonly the counting is possible one or two weeks after the sampling, depending on the activity level of noble gases and iodines.

TABLE 4 :

DISCRIMINATION METHODS BASED ON LONG-LIFE FISSION PRODUCT CUMULATIVE FISSION YIELDS - Yc (%) -

(Compilation of fission product yields [B.F. RIDER] - Vallecitos Nuclear Center - 1981)

Isotope	Haf-life	CUMULATIVE FISSION YIELD Yc (%)								
		U 235		U 238	Pu 239		Pu 240	Pu 241		Pu 242
		thermal	fast	fast	thermal	fast	fast	thermal	fast	fast
106 Ru	373 d	0,40	0,54	2,48	4,30	4,36	4 97	6 13	6,09	5,31
144 Ce	285 d	5,50	5,29	4 55	3,74	3,68	4 07	4,21	4,19	4,26

The analysis of this ratio has to take into account the behaviour of these isotopes in the primary circuit, which can induce a distorsion of its value.

6. APPLICATION OF DISCRIMINATION METHODS DURING THE 11th AND THE 12th CYCLE OF DAMPIERRE UNIT 1

6.1 Application during cycle 11

A specific analysis of noble gases released into the primary circuit out of the defective rods has been performed during the 11th cycle, in order to determine the time at which the MOX assembly failed.

Based on the routine measurements of the operator (daily gamma spectrometry of the primary water, after a decay time of about one hour), the method concerning the ratio ^{135}Xe / ^{85m}Kr has been chosen.

The study of the evolution of the specific gamma activity ratio (without correction of the release rate ratio) over the cycle 11, shows clearly the time at which the MOX failed: at the beginning of October 1992 (see Fig. 8).

6.2 Application during cycle 12

6.2.1 Methods concerning radioactive noble gases

During this cycle, methods developed in § 4.2.1 have been regularly applied. Results are presented in :

* Figs. 9 and 10 for the ratio ^{135}Xe / ^{85m}Kr, respectively without and with correction of release rate R/B,
* Fig. 11 for the ratio ^{138}Xe / ^{87}Kr (with R/B correction),
* Fig. 12 for the ratio ^{137}Xe / ^{89}Kr (with R/B correction).

On these figures are plotted :

- the evolution of the ratio for a defective UO_2 rod and for a MOX rod versus the burnup of the rod (theoretical values),
- the measured ratios during the cycle 12, with the hypothesis of a MOX assembly burnup of 12 GWd/tU and 24 GWd/tU at the end of the 11th and the 12th cycle respectively.

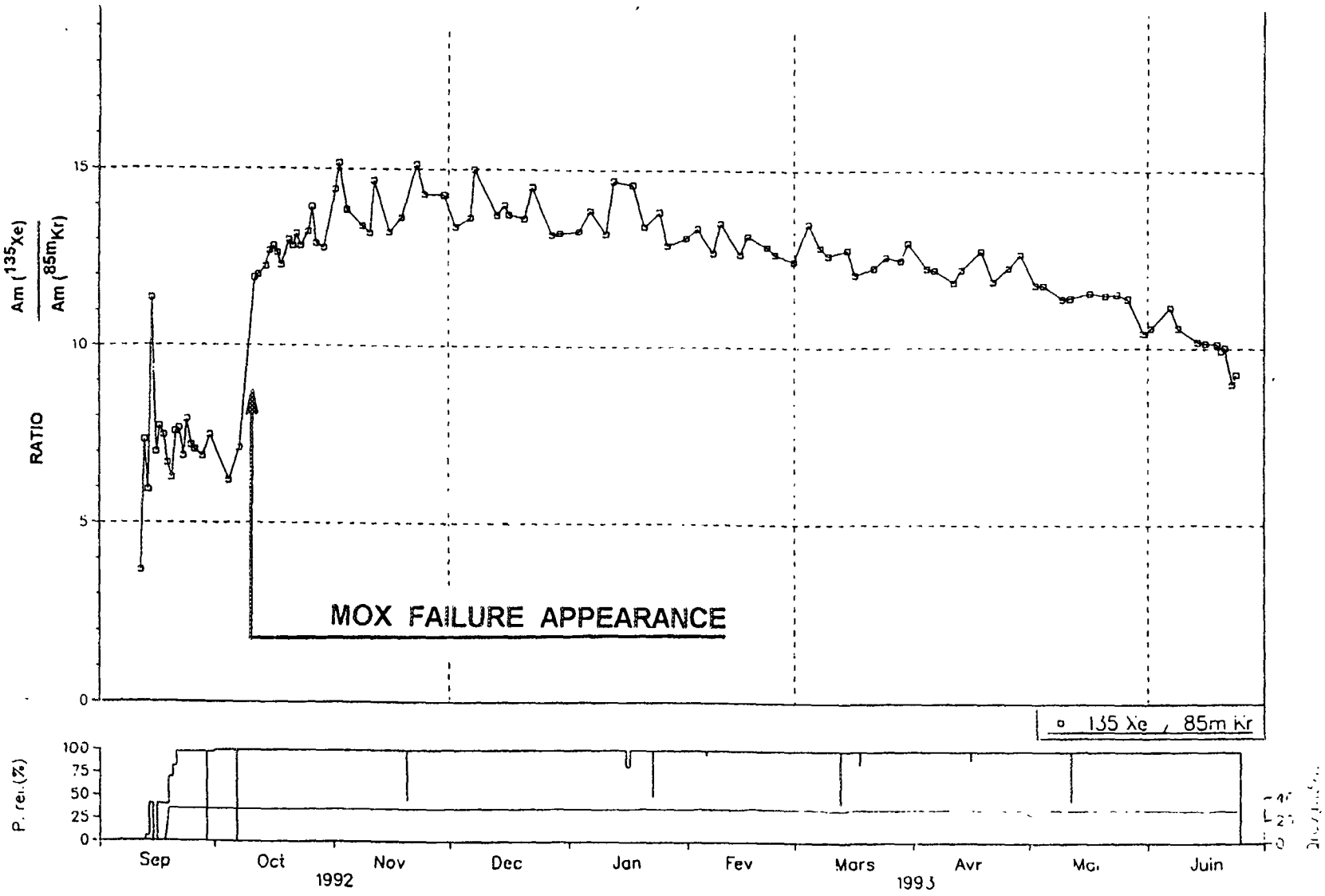

FIG. 8. *Specific gamma activity ratio $^{135}Xe/^{85m}Kr$ during cycle 11 of DAMPIERRE Unit 1.*

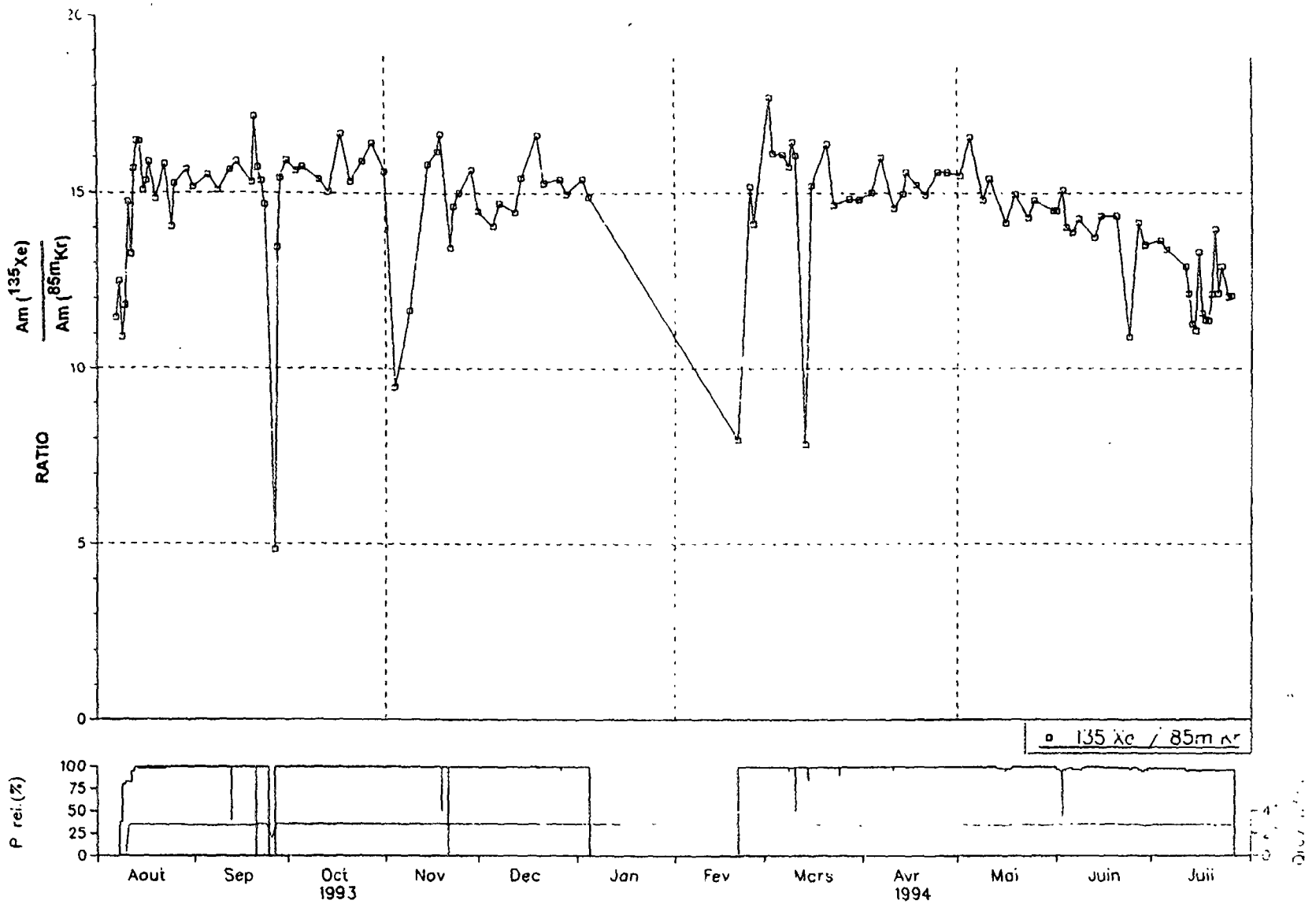

FIG. 9. *Application of discrimination methods: Specific gamma activity ratio $^{135}Xe/^{85m}Kr$ during cycle 12 of DAMPIERRE Unit 1 (without R/B correction).*

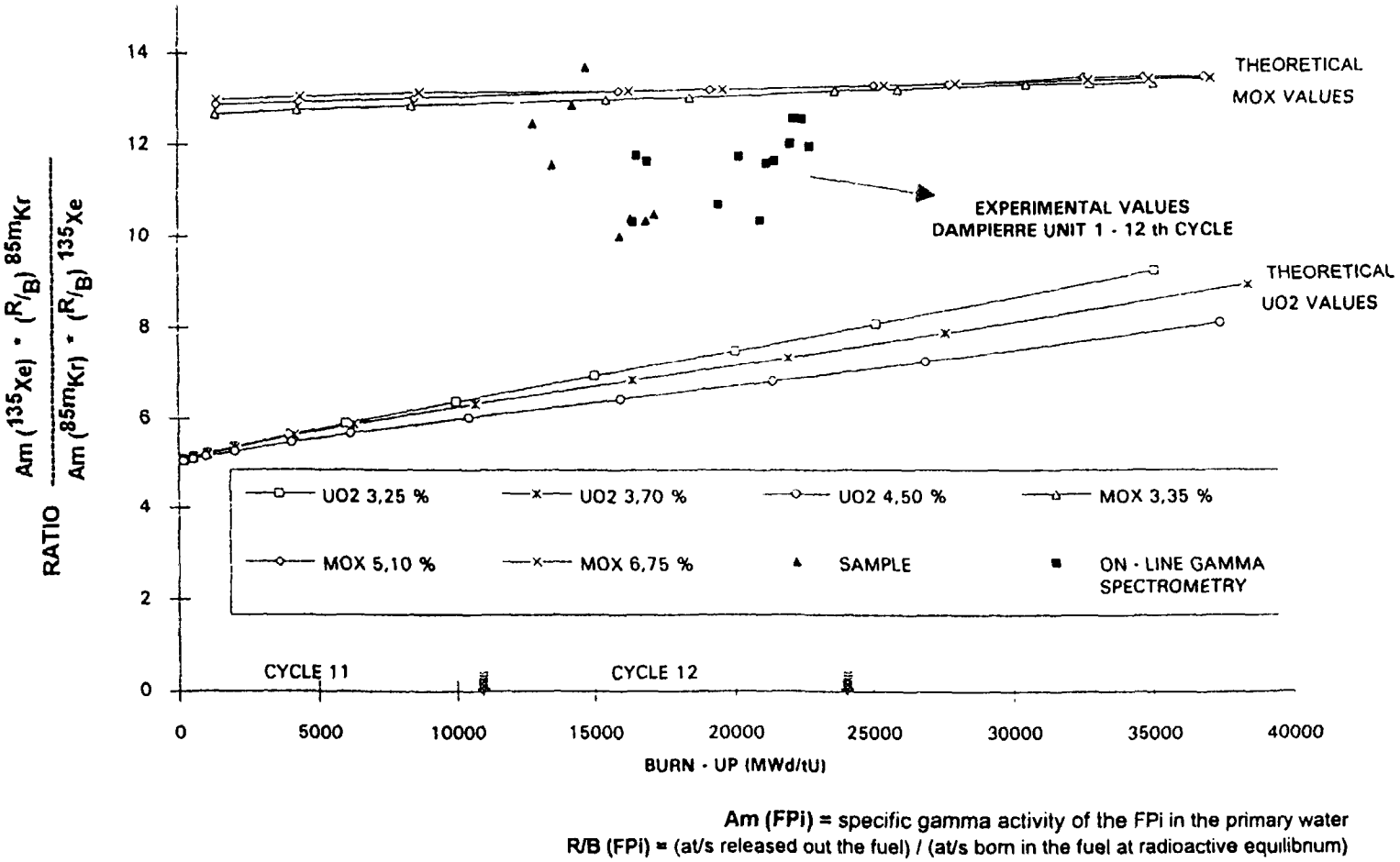

FIG. 10. *Application of discrimination methods: Specific gamma activity ratio $^{135}Xe/^{85m}Kr$ during cycle 12 of DAMPIERRE Unit 1.*

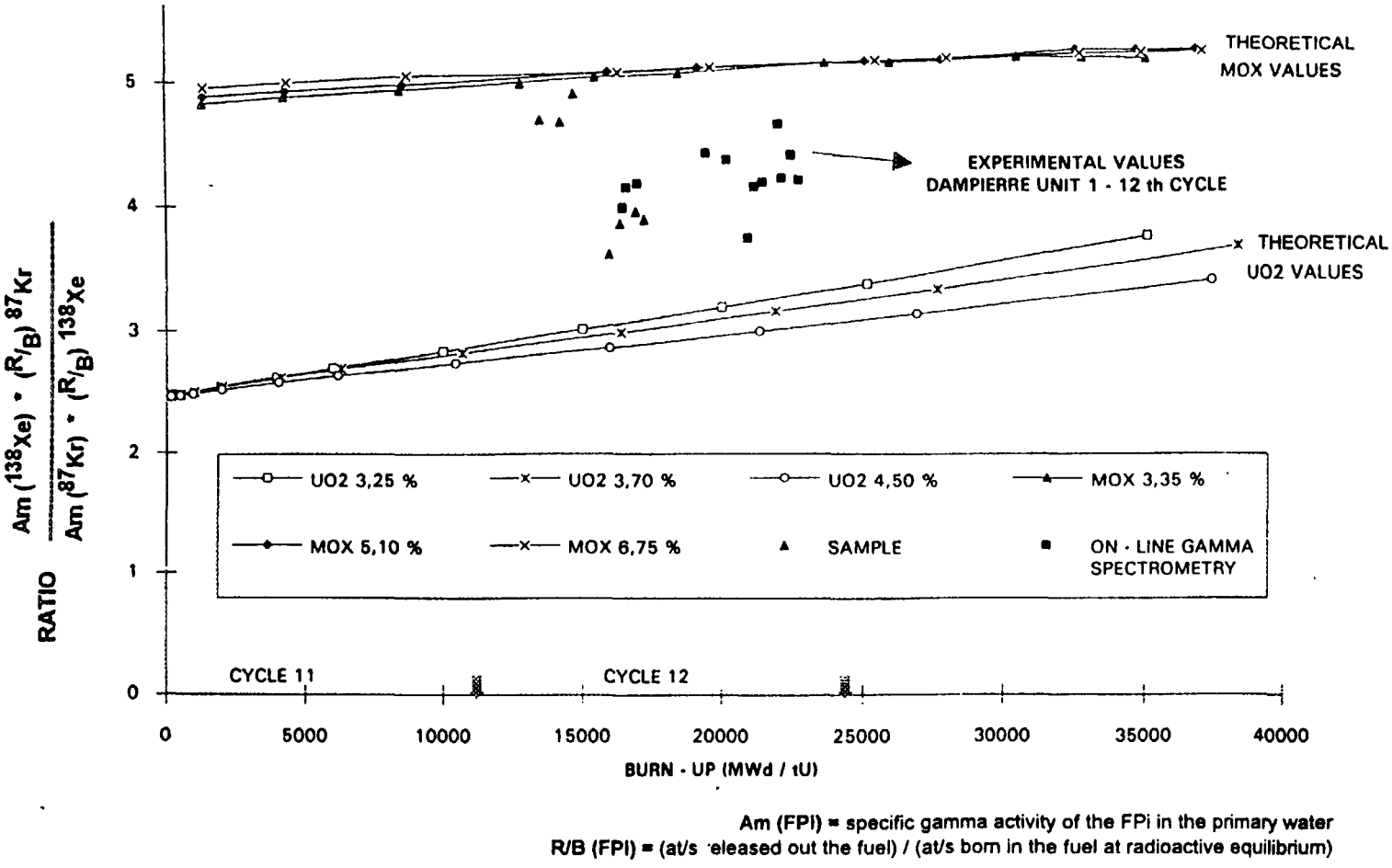

FIG. 11. *Application of discrimination methods: Specific gamma activity ratio $^{135}Xe/^{85m}Kr$ during cycle 12 of DAMPIERRE Unit 1.*

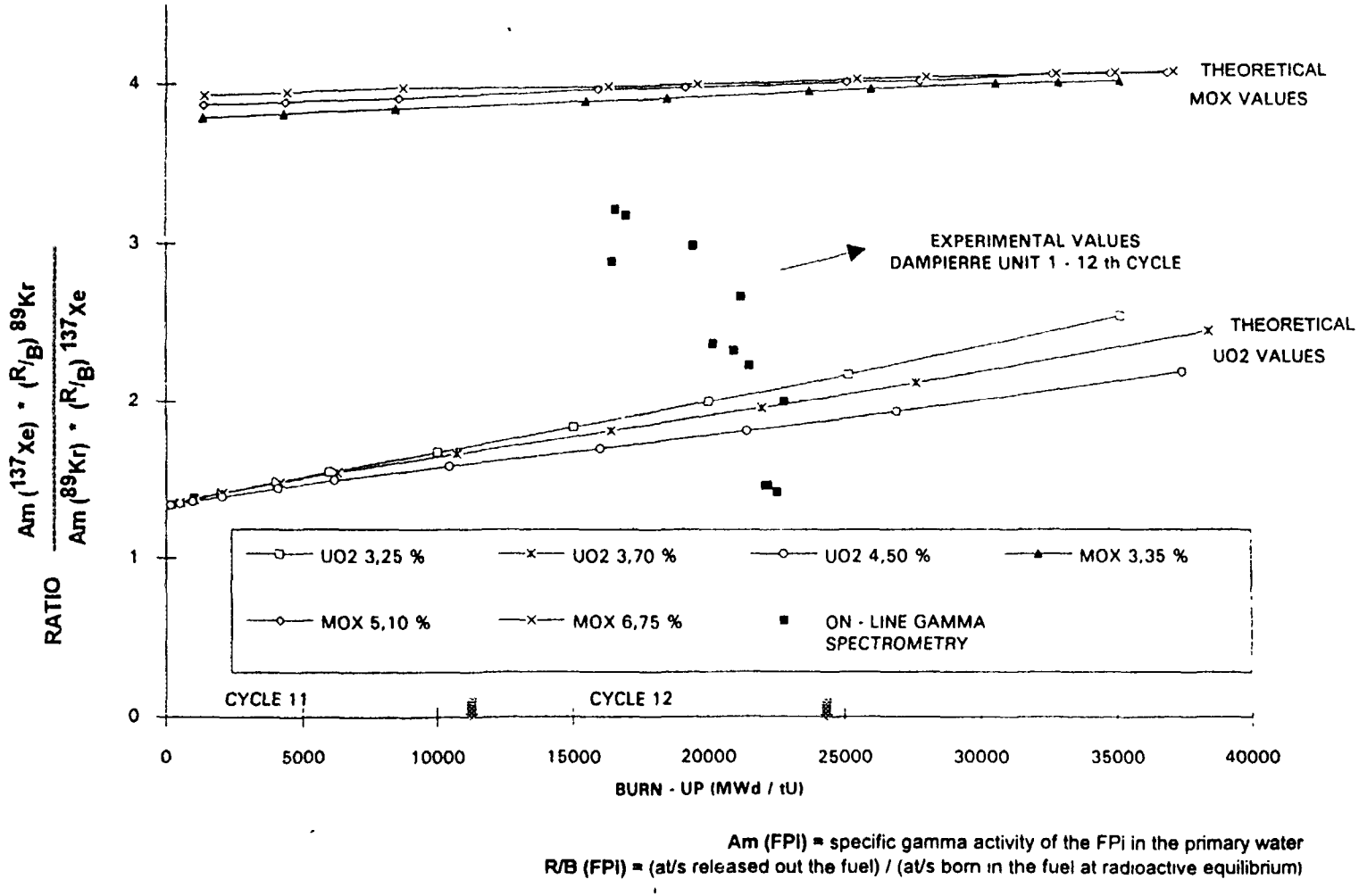

FIG. 12. *Application of discrimination methods: Specific gamma activity ratio $^{135}Xe/^{85m}Kr$ during cycle 12 of DAMPIERRE Unit 1.*

The analysis of these results indicates a good agreement between measured and theorical values for MOX fuel, specially during the first monthes of the cycle (from august to october 1993). Since mid-november 1993, a slight discrepancy is visible, corresponding to an evolution of measured values towards theoretical values for UO_2.

Up to now, the explanation of this evolution is not well understood. It could correspond to the apparition of a little defect on a UO2 fuel rod. But unfortunately the single-channel calibration problem encountered during sipping control (see § 3.3) did not permit to confirm this hypothesis.

The third ratio (^{137}Xe / ^{89}Kr) shows values lower than theoretical MOX range. The reason is probably due to the high dependance of the ratio with the transit time between the defect and the EMEL device. This time has to be better determined.

6.2.2 Methods concerning long-life fission product

Due to the small size of the defect, and to the absence of evolution during the irradiation, no dissemination of fissile matter occured. Consequently solid fission product (such as Ru and Ce) have been occasionnaly detected, but no reliable conclusion could be drawn.

7. CONCLUSION

After seven years of operating in France, the MOX fuel behaviour is very satisfactory up to typical burn-up reached with 1/3 annual core management.

The first failure on a MOX rod encountered at DAMPIERRE Unit 1 during its 11^{th} cycle and reloaded for the 12^{th} cycle showed a good behaviour over one year of irradiation and a burn-up up to 24 GWd/tU : no release of fissile matter and no evolution of the defect size were observed.

Analysis of fission product activity levels observed during cycle 11 and 12 of DAMPIERRE Unit 1 shows release rates of fission product out of defective mixed oxide similar to those obtained with uranium oxide. This is in good agreement with theorical prediction (PROFIP code) and the experimental release rates measured on a defective experimental fresh MOX rod irradiated in a research reactor of the CEA (EDITHMOX 01).

Application of on-line discrimination methods developed by EDF and CEA in order to know the type (UO_2 or MOX) of fission product source appears as very encouraging. Methods based on the ratio of two radioactive noble gases permitted to underline without ambiguity the presence of a leaking MOX rod, and to monitor its behaviour for the 12^{th} cycle. Specially, in the case of DAMPIERRE Unit 1, the ratio ^{135}Xe / ^{85m}Kr was an useful and easy to operate method to differentiate very early a MOX failure from an UO_2 failure during reactor operation through routine gamma activity measurements.

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IRRADIATION OF ARGENTINE MOX FUELS: POST-IRRADIATION RESULTS AND ANALYSIS

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Abstract

The irradiation of the first Argentine prototypes of PHWR MOX fuels began in 1986. These experiments were made in the HFR-Petten reactor, Holland. The rods were prepared and controlled in the CNEA's facility. The postirradiation examinations were performed in the Kernforschungszentrum, Karlsruhe, Germany and in the JRC, Petten. The first rod has been used for destructive pre-irradiation analysis. The second one as a pathfinder to adjust systems in the HFR. Two additional rods including iodine doped pellets were intended to simulate 15000 MWd/T(M) burnup. The remaining two rods were irradiated until 15000 MWd/T(M) (BU15 experiment). One of them underwent a final ramp with the aim of verifying fabrication processes and studying the behaviour under power transients. BACO code was used to define the power histories and to analyse the experiments. This paper presents the postirradiation examinations for the BU15 experiment and a comparison with the BACO outputs for the rod that presented a failure during the ramp test of the BU15 experiment.

1. INTRODUCTION

The irradiation of the first prototypes of MOX fuels fabricated in Argentina began in 1986 [1]. These experiments were made in the HFR-Petten reactor, Holland. The six rods were fabricated in the α Facility (GAID-CNEA-Argentina) [2].

The first rod has been used for destructive pre-irradiation analysis in the KFK (Kernforschungszentrum Karlsruhe), Germany. The second one was a pathfinder to adjust systems in the HFR [3]. The rod was irradiated for approximately 100 hours with a final ramp.

Two additional rods included iodine doped pellets. One of them CsI and auxiliary components and the second one elemental iodine. The concentration of iodine was calculated to simulate a burnup of 15000 MWd/ton(M) [4]. The power histories were defined with the BACO code. An irradiation period of 15 days including two power cyclings and a final ramp was designed.

The experiment named BU15 was performed with the last two rods. The goal of this experiment was to verify the fabrication processes and to study the fuel behaviour with respect to PCI-SCC. Both rods were irradiated together for a long period and then one of them underwent a final power ramp after a short preconditioning irradiation. The final burnup was 15000 Mwd/ton(M). The power level during irradiation was low and without important demands, only the normal shutdowns of the HFR. The ramp was similar to that used for the iodine test [5].

The pathfinder had an excellent behaviour in the HFR reactor. The presence of microcracks in the cladding inner surface was observed in the CsI doping test as it was predicted with the BACO code, although the postirradiation examinations were not conclusive about this point.

During the BU15 experiment the final test ramp was interrupted due to an increase of radioactivity in the coolant due to a failure in the rod. The visual inspection of the rod revealed a defect consisting in a small circular hole.

Using the BACO code taking special care in the stress analysis of the rod and in the calculation of the inventory of the fission gases, we found that the maximum power level reached in the defective zone of the MOX rod was similar to the threshold value to induce stress corrosion cracking (SCC) in standard LWR fuels.

Postirradiation analyses have shown that the defect in the BU15 rod was a SCC failure, in good agreement with BACO [6].

The analysis of the experiment made with the BACO code an a complete description of the calculation was presented at the IAEA's TCM on "Water Reactor Fuel Element Modelling at High Burnup and Experimental Support" (Windermere, 1994) [7]. At that meeting it was suggested that additional postirradiation examinations and more details of the failure condition would be needed to confirm the failure as being due to PCI. This report is a continuation of the above mentioned paper. We present a short description of the experiments performed with the MOX fuel rods, the main findings of the new set of postirradiation examinations (not available at the Windermere meeting) and the results attained using the BACO code to study the behaviour of the defective fuel rod.

2. FUEL RODS DESCRIPTION

The rods were originally designed for the MZFR reactor (Karlsruhe, Germany). Due to the decommissioning of the MZFR, the experiments were performed in the HFR-Petten reactor. It was necessary to do some rod dimensional changes (i.e. fuel length) and to modify the end plug design.

Figure 1 shows the rod type A1. Table I presents the main characteristics of the rod.

3. BU15 EXPERIMENT DESCRIPTION

The BU15 irradiation experiment was carried out with fuel rods A.1.2 and A.1.3, both similar to the pathfinder. The power history for this irradiation test was proposed upon calculations made with the BACO code. The target duration of the irradiation was an average burnup of 15000 MWd/ton(M), more or less twice the final burnup for Atucha I fuel. After the steady state irradiation at on average power level of 230 W/cm one of the rods was submitted to a power ramp [8].

During the steady state irradiation both rods were assembled together through a threaded coupling. This system permits an easy disassemble of the two rods in any irradiation stage.

The average burnup of 15000 MWd/t(M) was reached in two main steps:

a) Up to a burnup of 8100 MWd/t(M) the rods were irradiated in different locations of the HFR core. The irradiation device consisted in an aluminum fuel rod capsule located in either of two different carriers. The first of these consisting an aluminum structure in the core section whilst the other had a hollow stainless steel structure having the possibility of containing pressurized BF_3 gas for power control purposes. The primary pressure in the fuel rod capsule was kept at 145 bar (see *Fig. 2*), and

b) Up to a burnup of 15000 MWd/t(M) both rods were irradiated in the Pulse Side Facility (PSF) of the reactor (see *Fig. 3*). The irradiation device consisted of an aluminum fuel rod capsule located in a special PSF capsule carrier which can be translated parallel to the HFR core box wall. Also the primary pressure in the fuel rod capsule was kept at 145 bar.

Fuel Rod for the BU-15 experiment

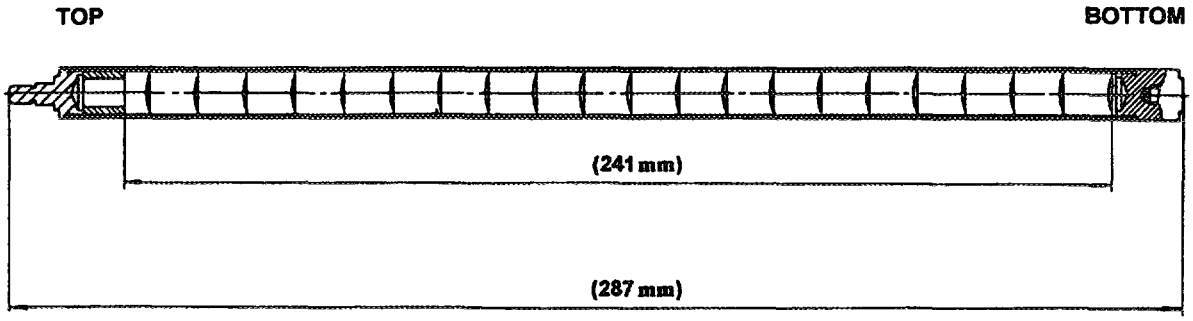

Figure 1: MOX fuel rod type A.1 for the BU15 experiment.

Table I **MOX fuels irradiated at Petten reactor.**
Fuel rods characteristics (U/Pu mixed oxide) .

Table Ia : Fuel rods type A.1, A.3 y A.4.

Rod	A.1
Length [cm]	24.1 cm
Pellets number	21
Compensating pellets number	1
Filling gasses	He
Filling pressure	1.15 atmospheres

Table Ib : Pellets

Density	10.52 ± 0.04 gr/cm^3
Pellet height	11.2 ± 0.1 mm
Pellet diameter	10.40 ± 0.01
$Pu_{fiss}/U+Pu_{met}$	0.53 %
Enrichment (U^{235}+Pu)	1.25 %
O/M relation	2.00
Dishing volume	25. ± 5. mm^3

Table Ic : Cladding

Cladding material	Zry-4
Cladding inner diameter	11.7 mm
Cladding thickens	0.6 mm

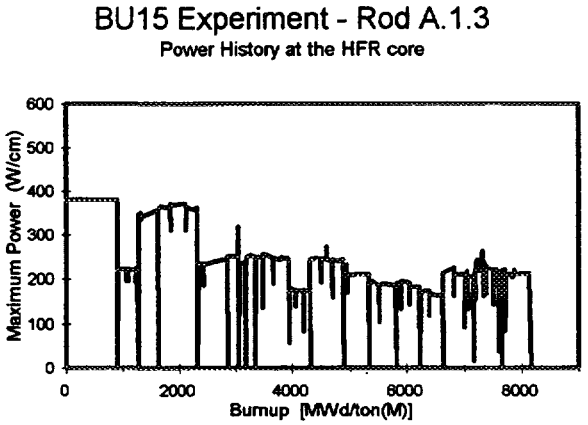

Figure 2: Power irradiation history for the A.1.3 fuel rod in the HFR core.

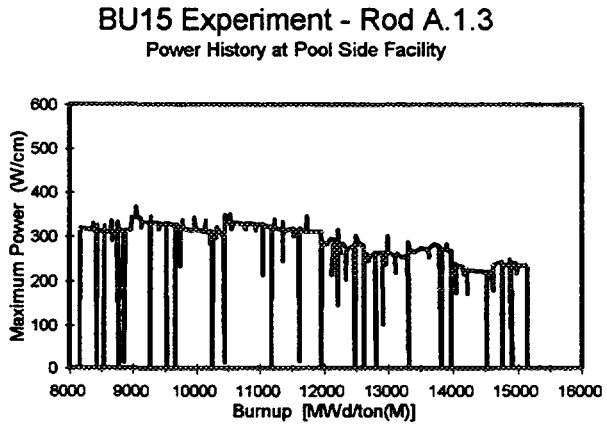

Figure 3: Power irradiation history for the A.1.3 fuel rod in the Pool Side Facility (PSP).

After termination of the bulk irradiation phase both fuel rods were disassembled in the Petten hot cells and the A.1.3 rod was prepared for insertion into a standard LWR fuel testing capsule consisting in an aluminum capsule located in a similar carrier as described in b), which allowed to adjust it to a specified power level. In this case the primary system pressure in the fuel rod capsule was kept during the ramp test phase at 115 bar (e.g. PHWR system pressure).

The ramp proposed consisted in two parts (see *Fig. 4*):

1) a short period of pre-irradiation power level in the PSF position which will be used for the ramp test in order to determine the experiment power versus PSF position characteristics and,
2) the ramp test starting with a ramp rate of 50 $Wcm^{-1}min^{-1}$ from nearly zero power to maximum 420 W/cm linear fuel rod power and followed by a 6 hours steady state holding at this maximum power.

The ramp test was performed following the proposed schedule. However, instead of the anticipated high power level of 420 W/cm the maximum fuel power reached only a maximum ramp power level of approximately 380 W/cm, due to the low quantity of remaining fissile material in the fuel rod.

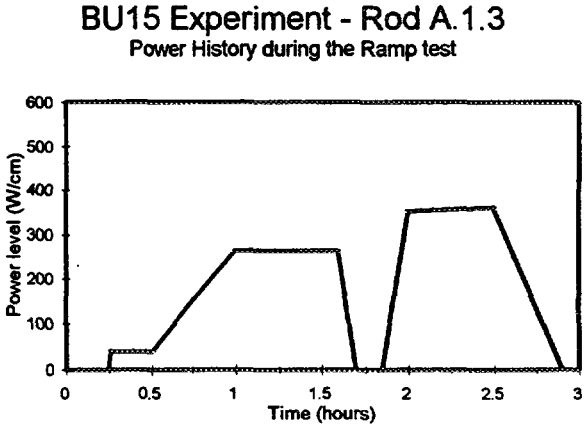

Figure 4: Power irradiation history for the A.1.3 rod in the PSF during the ramp test.

Rods A.1.2 and A.1.3 behaved during the stationary phase as it was expected:

- no rod failures were detected,
- no fabrication defects were evident.

During the EOL power ramp, rod A.1.3 behaved as follows:

- a maximum power of 390 W/cm was reached,
- the power ramp had to be finished earlier than planned due to an increase in activity in the coolant circuit,
- visual inspection of the fuel rod revealed the existence of a small circular hole in the cladding

During operation for the entire experiment the total power production was determined via a heat balance approach by measuring the increase in temperature of the capsule coolant water and then the real power fuel rod production by difference with the measured nuclear heating of an empty capsule structure.

The axial power distribution of the fuel rods, as characterised by the normalised thermal neutron fluence, for the different irradiation phases are shown in *Figs. 5, 6* and *7*. The relative physical position of the fuel rods is superimposed on these plots.

The power history includes the normal cycle operation, it means, reactor shutdowns, variations from the planned average power in the fuel rods of approximately 100 W/cm and the final ramp test for the A.1.3 fuel.

The total time for this test was of 1020.2 calendar days, corresponding to 26 irradiation periods (531.5 days) and 26 periods on which the fuels rods were not irradiated due to changes of irradiation capsules, reactor shutdowns or non availability of a suitable core position.

After a cooling time of 49 days and 9 hours rod A.1.3 was submitted to a power ramp.

Before the ramp test a preconditioning irradiation was performed, increasing power rate at 7.2 W/cm.min during 42 minutes until an average fuel rod power of 250/260 W/cm was reached. This last power level was held 39 minutes. The fuel rod power was then reduced during 5 minutes at a power rate of 48 W/cm.min until an average fuel rod power of 10 W/cm was attained, after 10 minutes, the power level was increase at a rate of 43 W/cm.min during 8 minutes to a final average rod power of 350/360 W/cm. This power was held during 52 minutes.

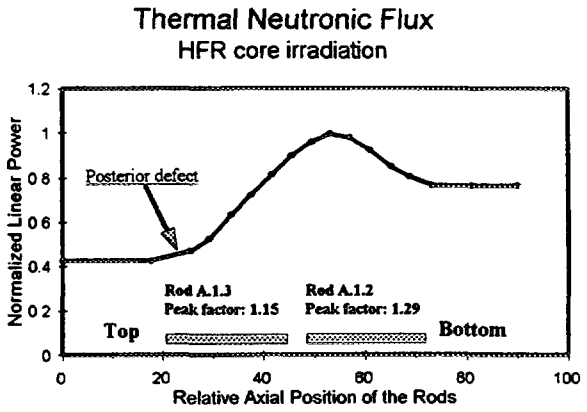

Figure 5: Thermal neutronic flux distribution for the assembly in the HFR core.

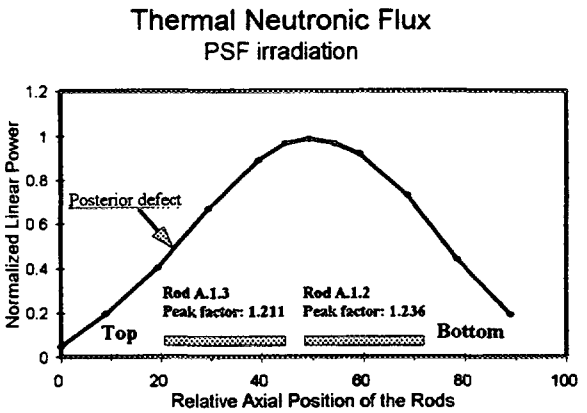

Figure 6: Thermal neutronic flux distribution for the assembly in the Pool Side Facility (PSF)

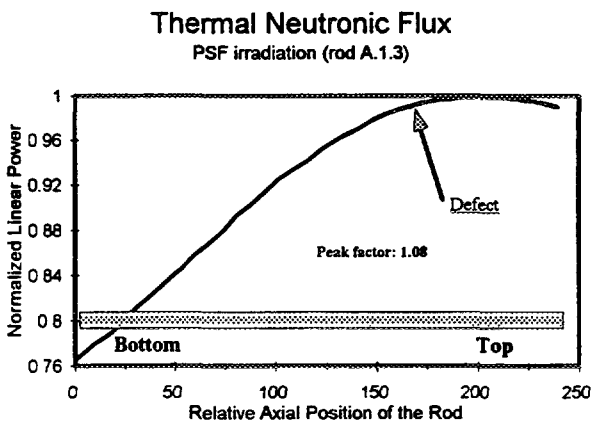

Figure 7: Thermal neutronic flux distribution for the A.1.3 fuel rod in the PSF during the ramp test.

At approximately 28 minutes operation at the ramp power level an increase of the activity of the primary water was detected. This activity peak decreased after 6 minutes to its previous level. As this event gave reason for assumption of a fuel rod failure the experiment was 24 minutes after the start of the activity release shutdown at a power rate of 175 W/cm.min during 2 minutes.

It is noteworthy that at beginning of life, the A.1.2 and the A.1.3 fuel rods reached maximum peak fuel rod power of 430 W/cm and 380 W/cm respectively. The average for the assembly was 330 W/cm. These maximum peak power levels, during that pre-irradiation period, where higher or equal than the maximum power reached during the final ramp test for the A.1.3 fuel rod.

4. POSTIRRADIATION EXAMINATIONS OF THE BU15 EXPERIMENT

After finishing the low level irradiation in the HFR, the irradiation device was disassembled and some non-destructive examinations were done on both pins [17]. These examinations were repeated on fuel rod A.1.3. after the transient. Destructive examinations were performed on both rods (for A.1.2 after the low level irradiation and for A.1.3. after the transient). The main findings of the postirradiation examinations are described in the following paragraphs.

4.1 Visual Inspection

Both pins were photographed from three orientations along the whole axis after the low level irradiation. Rod A.1.3. was photographed in the same manner after the transient.
No unusual features appeared in both rods after the low level irradiation.

The following findings can be stated for pin A.1.3. after the power ramp:

- Rod colour was mainly light grey, the zone next to the female screw end plug (bottom end plug) was dark grey coloured.
- From 219 mm, counting form the lower end plug, up to the upper end plug the surface shows metallic luster.
- A defect appeared approximately at position 171.2 mm from the lower end plug, shown in *Fig. 14.*

4.2 Eddy Current Check

Eddy current testing was done in the pins:

a) before irradiation,
b) after the low level irradiation before the transient and
c) after the transient for rod A.1.3.

Results are shown in *Figs. 8* and *9.*

Before irradiation slight wavelike oscillations are observed along the fuel column.
After low level operation the amplitude of the oscillations has increased and a correlation with the pellet structure of the fuel column and with the power profiles of the irradiation (maximum oscillations corresponding to maximum power positions) is clearly visible. This should be compared with the irradiation power profiles shown in *Figs. 5, 6* and *7.*

After the transient performed on rod A.1.3 the amplitude of the oscillations has increased noticeably with respect to those observed after the low level irradiation, here again a correlation with the power profile during the ramp is apparent.

A careful inspection of rod A.1.3 after the transient revealed a small defect at position 171.2 mm counting from the lowest end of the bottom end plug.

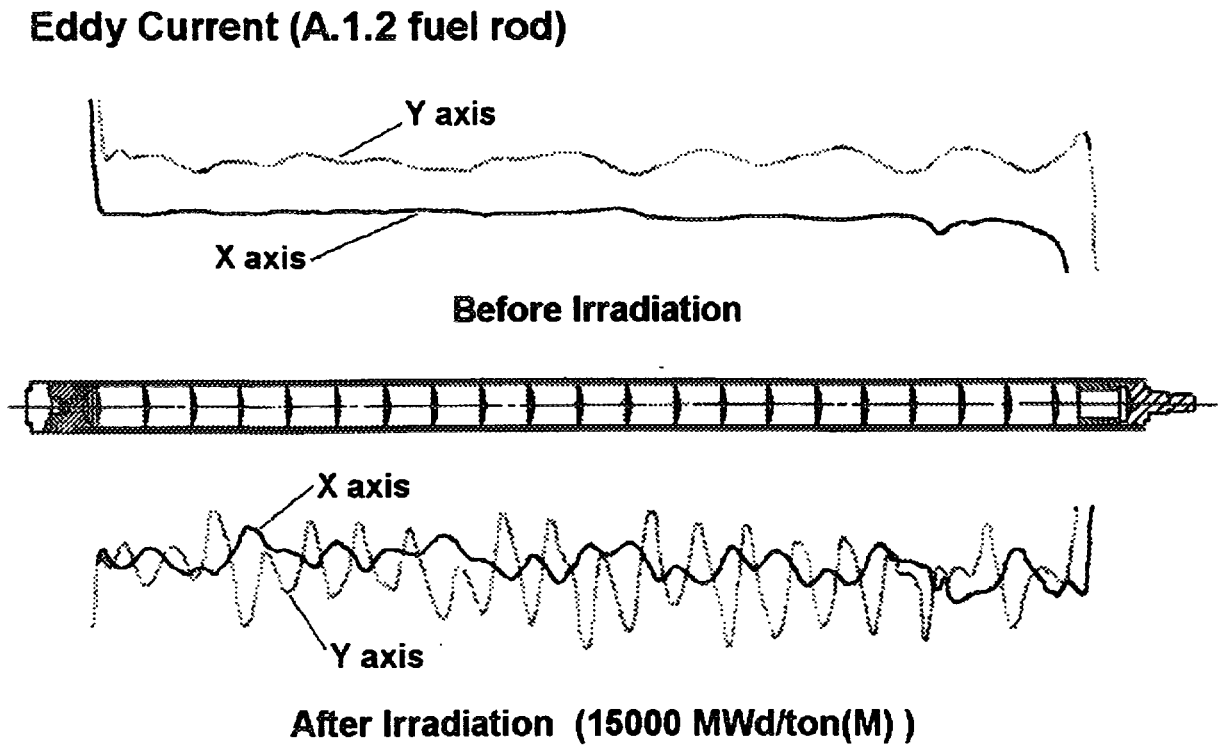

Figure 8: Eddy current check. Fuel rod A.1.2

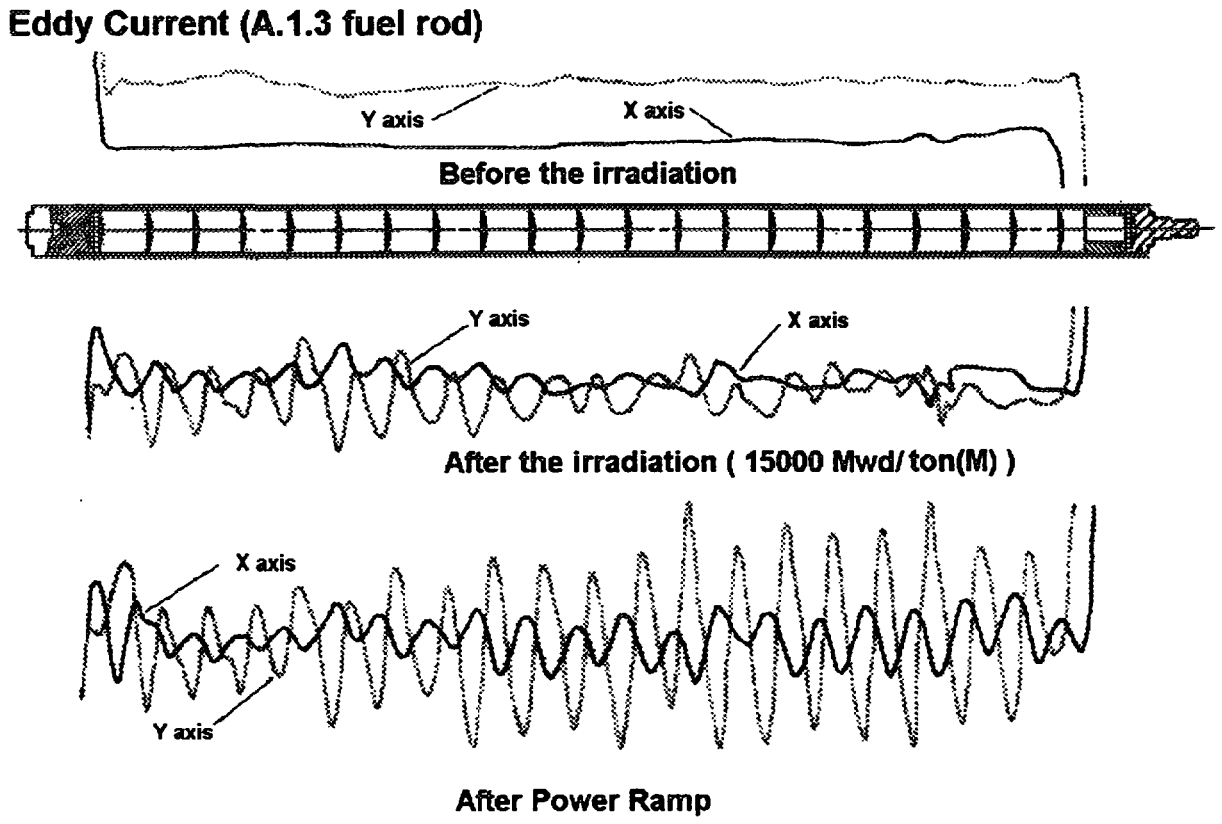

Figure 9: Eddy current check. Fuel rod A.1.3

4.3 Neutron Radiography

Both pins were subjected to neutron radiography before and after low level operation and rod A.1.3. was tested again after the transient. Exposures were taken from two orientations with 0° and 90° angles.

Radiography has penetrated the fuel completely so that the internal geometry of the fuel column could be evaluated. The following findings can be stated:

- Pellets and their dishings can be clearly discerned.
- Most of the pellets show some cracking. The direction of the cracks is preferably longitudinal or diagonal. There are not pronounced transversal cracks that separate pellet sections.
- Moisture is visible inside the A.1.3 rod after the transient, in the area near the defect.

4.4 Gamma Scanning

Gamma scanning was performed on both fuel rods after the low power irradiation while they were still attached to each other. Rod A.1.3. was tested after the transient.

For the two rods after the low level irradiation gamma spectrometry was performed with only about 10.8 days cooling time. In this way it was possible to detect also short-lived radionuclides like ^{132}Te. For eight radionuclides axial scans were taken along the length of both test pins still attached to each other. Results of this test are shown in Tables II, III and *IV* for each individual fuel rod and for the average of the assembly. A gamma scanning profile for ^{140}La is shown in *Fig. 10.*

Some of the findings of this test will be mentioned below:

- The interfaces between all the pellets are visible as activity dips.
- The maximum of rod power according to the scans of the non-migrating radionuclides is located in the upper part of rod A.1.2., near the male screw end cap.
- A strong power gradient can be observed in pin A.1.3 where the upper end has seen only 52% of the maximum power.

Gamma scanning of rod A.1.3 has been performed after a cooling time of 38.2 days after the end of the power ramp. Gamma scanning profile for ^{140}La is shown in *Fig. 11.* Neither the axial power distribution nor the peaking factors have been appreciably modified due to the short irradiation time during the transient.

4.5 Dimensional Control

Dimensional control has been performed on rod A.1.2 after the low level irradiation and on rod A.1.3 after the transient. Digital measurement with a length increment of 0.5 mm and an angle increment of 15° was applied.

Common to all graphs was the appearance of circumferential ridging due to mechanical interaction between fuel and cladding. Ridging is clearly more pronounced for rod A.1.3, indicating the greater extent of the pellet-cladding interaction during the transient. The ridges had a height of about 10-15 μm for rod A.1.2 and 20-25 μm for rod A.1.3 with the location corresponding to pellet-pellet-interfaces. A correlation of the ridge height with the power profile for both the low level irradiation and the power ramp is apparent. Plots of rod diameter vs. length are shown in *Figs. 12* and *13.*

4.6 Sectioning Diagram

On the basis of the findings of non destructive post-irradiation examination, sectioning diagram for the destructive examination was set up. For rod A.1.2 a zone near the maximum linear power during

TABLE II: γ spectrometry for the A.1.2. rod (15000 MWd/t(M) of irradiation)

NUCLEIDE	Average Activity	Maxim Activity	Maxim Position	Peak factor (% of mean)
^{141}Ce	5789.9	6168	321.92	106.5
^{132}Te	6326.8	7009	297.60	110.8
^{131}I	14483.3	15505	309.19	107.1
^{103}Ru	43021.4	46410	313.14	107.9
^{106}Ru	2311.2	2507	341.48	108.5
^{137}Cs	2158.3	2297	366.82	106.4
^{95}Zr	17056.4	18283	298.25	107.2
^{140}La	22451.9	23283	297.00	106.7

TABLE III: γ spectrometry for the A.1.3. rod (15000 MWd/t(M) of irradiation)

NUCLEIDE	Average Activity	Maxim Activity	Maxim Position	Peak factor (% of mean)
^{141}Ce	4609.3	6042	257.60	131.1
^{132}Te	5409.8	6961	257.60	128.7
^{131}I	12064.8	15300	257.60	126.8
^{103}Ru	34178.1	45545	257.60	133.3
^{106}Ru	1692.1	2374	257.60	140.3
^{137}Cs	1584.2	2127	257.60	134.3
^{95}Zr	14341.4	18119	257.60	126.3
^{140}La	19134.6	23799	257.60	124.4

the low level irradiation was selected. For rod A.1.3 the area near the defect has been chosen for a complete cross section microscopic examination.

4.6.1 Pin A.1.2

- The fuel pellet is heavily cracked. The cracks run mostly radially but there is also a circumferential crack about halfway from the center to the periphery.
- The gap between fuel and cladding measures 8 to 12 μm radial.
- Normal grain size is 7.6 μm diameter in the periphery and 9.0 μm in the central zone.
- Pu distribution is homogeneous without agglomerations (see *Fig. 15*).

TABLE IV: γ spectrometry for the assembly of rods A.1.2 and A.1.3 (Average)

NUCLEIDO	Average Activity	Maxim Activity	Maxim Position	Peak factor (% of mean)
^{141}Ce	5267.3	6168	321.92	117.1
^{132}Te	59953.4	7011	290.45	117.8
^{131}I	13434.8	15505	309.19	115.4
^{103}Ru	39155.6	46410	313.14	118.5
^{106}Ru	2033.3	2507	341.48	123.3
^{137}Cs	1894.1	2297	366.82	121.3
^{95}Zr	15888.5	18283	289.25	115.1
^{140}La	21027.3	23967	295.27	114.0

MOX fuels irradiated at Petten reactor

Table V : BACO outputs for the A.1.3. pin.

Table Va : Maximum values reached during irradiation for each axial section.

	Axial section				
	1[1]	2[1]	3[2]	4[2]	5[2]
Linear power [W/cm]	430.	385.	340.	366.	409.
Pellet centre temperature [°C]	1433.	1245.	1102.	1191.	1334.
Pellet surface temperature [°C]	422.	413.	408.	415.	424.
Hoop stress [MPa]	174.	154.	130.	191.	208.
Contact pressure [MPa]	37.	34.	31.	40.	43.
von Misses equivalent stress at pellet centre [MPa]	609.	382.	347.	439.	384.
Surface crack scope [cm]	0.17	0.21	0.24	0.21	0.20
Cladding radial strain [%]	0.29	0.17	0.09	0.08	0.08
Cladding axial strain [%]	0.11	0.11	0.11	0.11	0.11

[1] Maximum values reached at BOL (beginning of life)

[2] Maximum values reached at the final ramp (EOL)

4.6.2 Pin A.1.3

Starting from position 180.1 mm, always counting from the bottom end plug, a series of cross sections have been prepared and examined, moving the sectioning position towards the observed location of the defect (171.2 mm), until the failure was clearly visible (*Fig. 16*).

The defect was a branched crack, through-wall type located near a circumferential ridge, facing a dip in the pellet surface (*Fig. 17*). Other similar incipient cracks have been revealed by the ceramography (*Fig. 18*). Although studies have not been made of the fracture surface the path cracking observed is typical of a PCI-SCC failure [9,10].

Grain size is 7.6 μm in the periphery and 9.0 μm in the centre of the pellet.

Table Vb : Reached values for each axial section EOL, cero power and STP conditions

	Axialsection				
	1	2	3	4	5
Pellet radius [cm]	0.5193	0.5195	0.5191	0.5183	0.5176
Cladding inner radius [cm]	0.5198	0.5199	0.5195	0.5190	0.5189
Cladding outer radius [cm]	0.5820	0.5823	0.5819	0.5817	0.5817
Gap [μm]	10.	9.	8.	12.	26.
Active stack length [cm]	4.777	4.775	4.775	4.780	4.782
Cladding length [cm]	4.785	4.784	4.786	4.782	4.784
Cladding radial strain [%]	-0.44	-0.38	-0.45	-0.48	-0.48
Cladding axial strain [%]	-0.72	-0.76	-0.70	-0.80	-0.76
STP Produced gases volume [cm^3]	18.67	16.36	14.03	11.62	9.82
STP Released gases volume [cm^3]	0.405	0.195	0.084	0.030	0.034
STP Trapped gases volume [cm^3]	11.78	11.49	10.9	9.92	8.37
STP Fission gases volume at grain boundary [cm^3]	6.47	4.68	3.01	1.67	1.42
Local Burnup [MWd/ton(M)]	19725.	17651.	15575.	13089.	10180.

Table Vc : Reached values for the rod at EOL and at STP conditions (and cero power).

	EOL	STP	Units
Active stack length	23.95	23.89	cm
Cladding length	23.94	23.92	cm
Axial cladding strain	-0.63	-0.75	%
Gases pressure	0.33	0.15	MPa
Fraction gas release	1.1		%
Produced gases volume (STP)	70.50		cm^3
Released gases volume (STP)	0.75		cm^3
Trapped gases volume (STP)	52.50		cm^3
Fission gases volume at grain boundary (STP)	17.25		cm^3
% He	72.		%
% Xe	24.		%
% Kr	4.		%
Burnup (average)	15244.		MWd/ton(M)

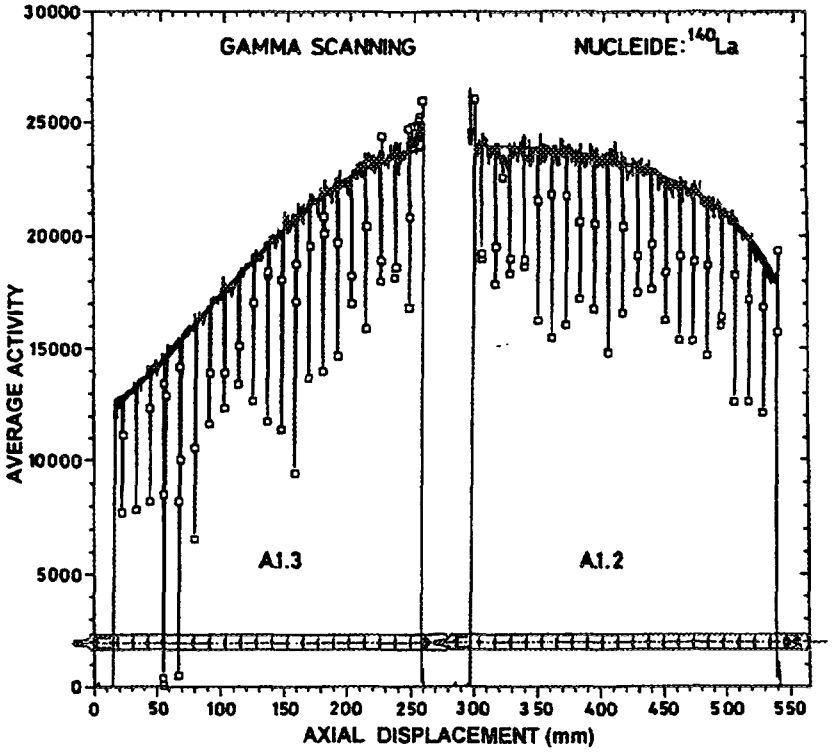

Figure 10: Gamma scanning profile for ^{140}La before the ramp.

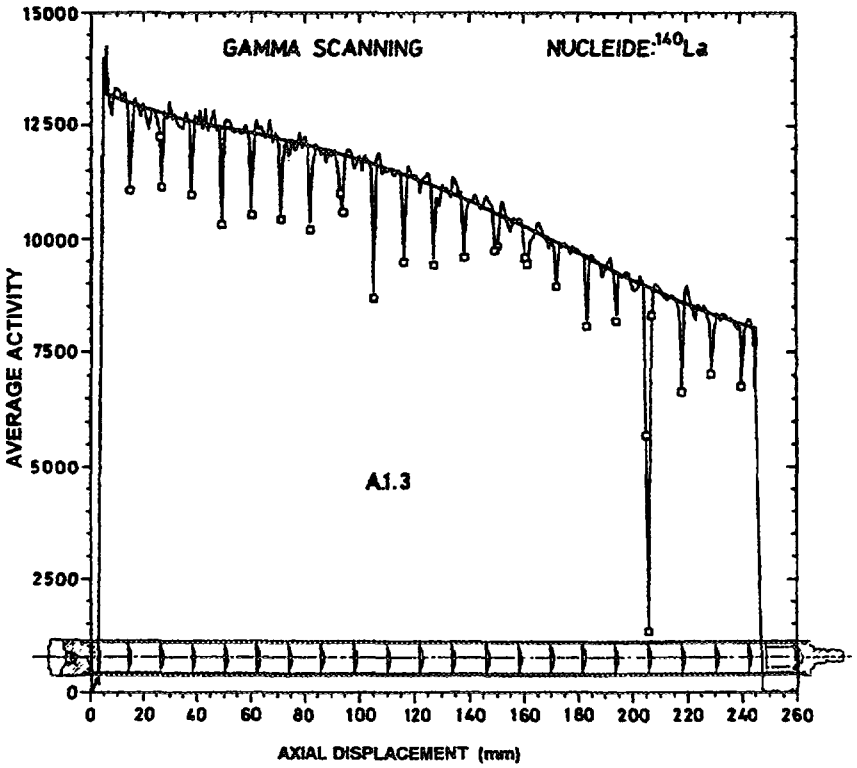

Figure 11: Gamma scanning profile for ^{140}La for rod A.1.3 after the power ramp.

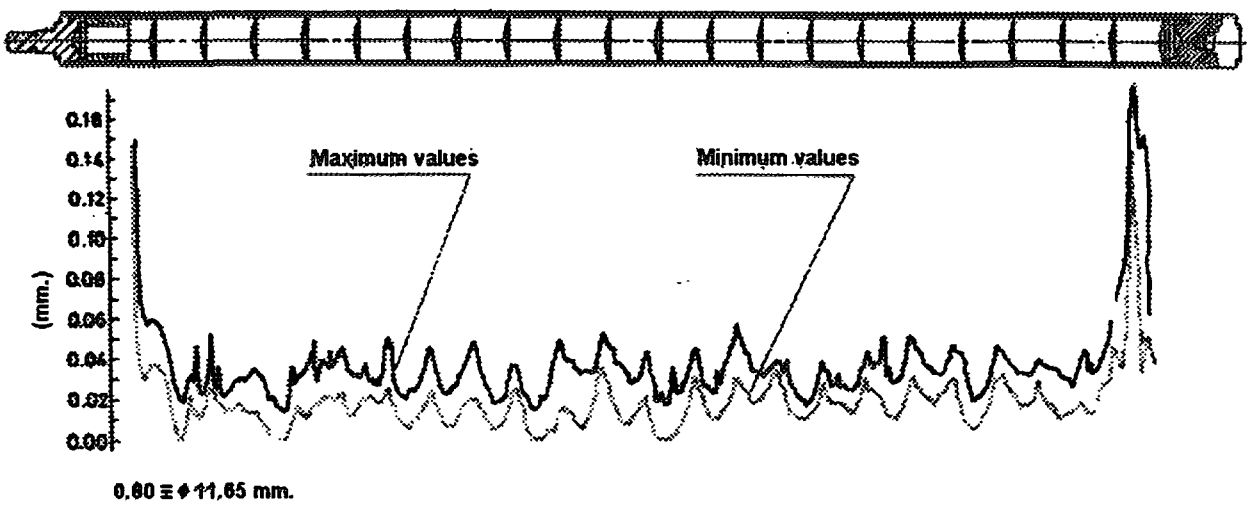

Figure 12: Diameter vs. lenght for fuel rod A.1.2

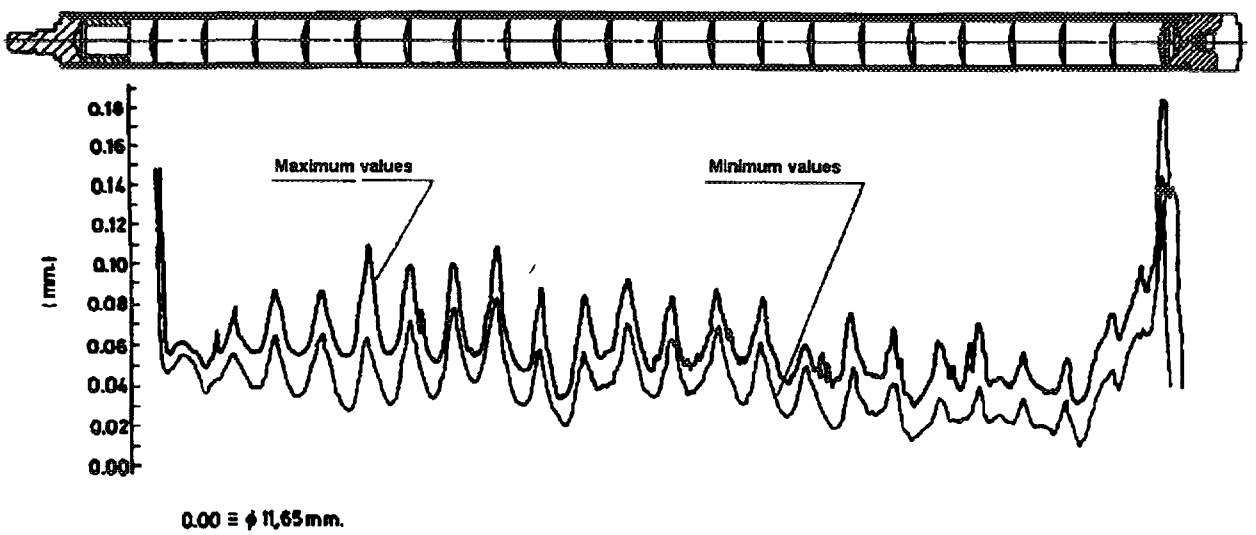

Figure 13: Diameter vs. lenght for fuel rod A.1.3

5. BACO CALCULATIONS FOR THE A.1.3 FUEL ROD

5.1 Baco Code

The BACO code (BArra COmbustible) simulates the fuel rod behaviour under operation conditions in a nuclear reactor. BACO code includes the following calculations and models: strain-stress state of the cladding and pellet system, cladding and pellet thermal-elastic-plastic analysis, swelling, pellet cracking, grain growth, relocation, porosity histogram evolution, creep, cladding anisotropy, fission gases inventory and gap conductance [11, 12 and 13].

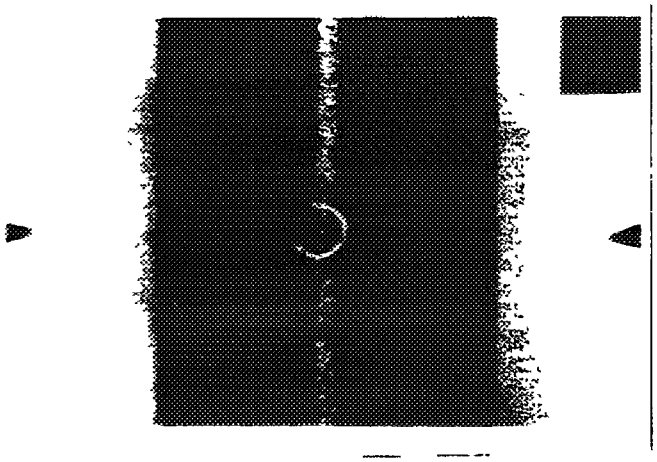

Figure 14: Fuel rod type A.1.3 showing the hole that it is included in the circle.

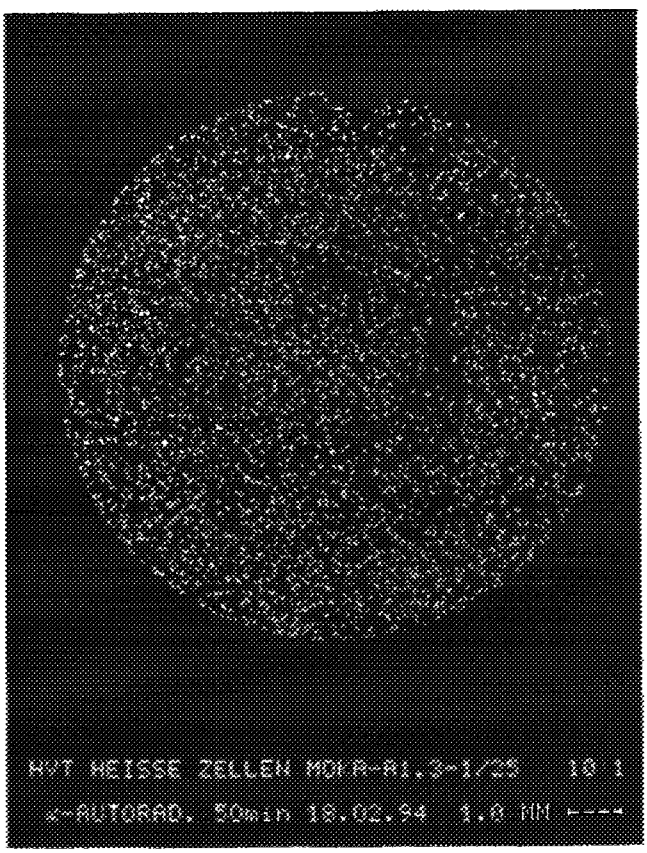

Figure 15: α-Autorrad. of a section of the fuel rod A.1.3 showing the Pu distribution.

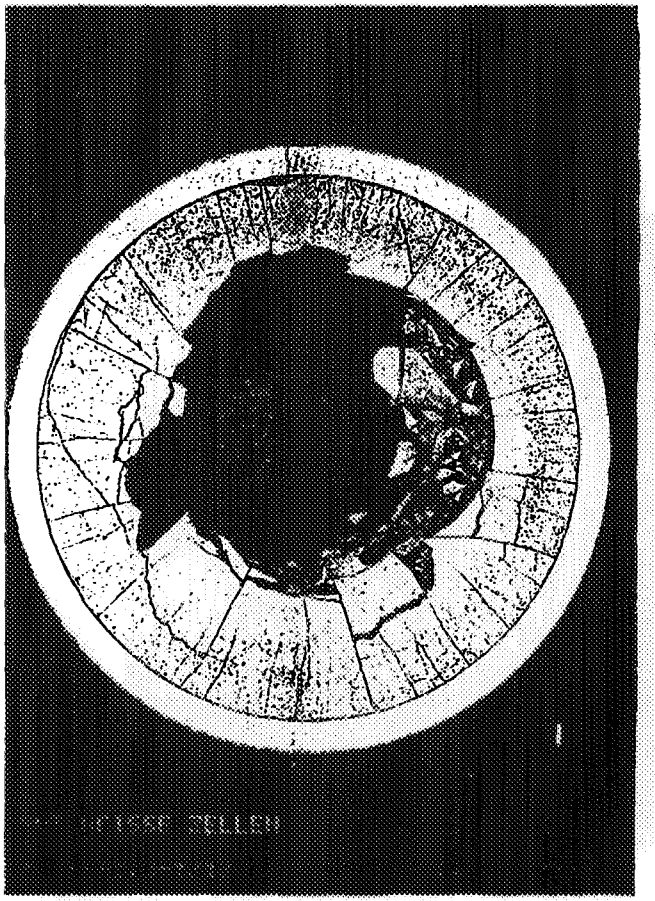

Figure 16: Micrograph of a cross section near the defect of the fuel rod A.1.3

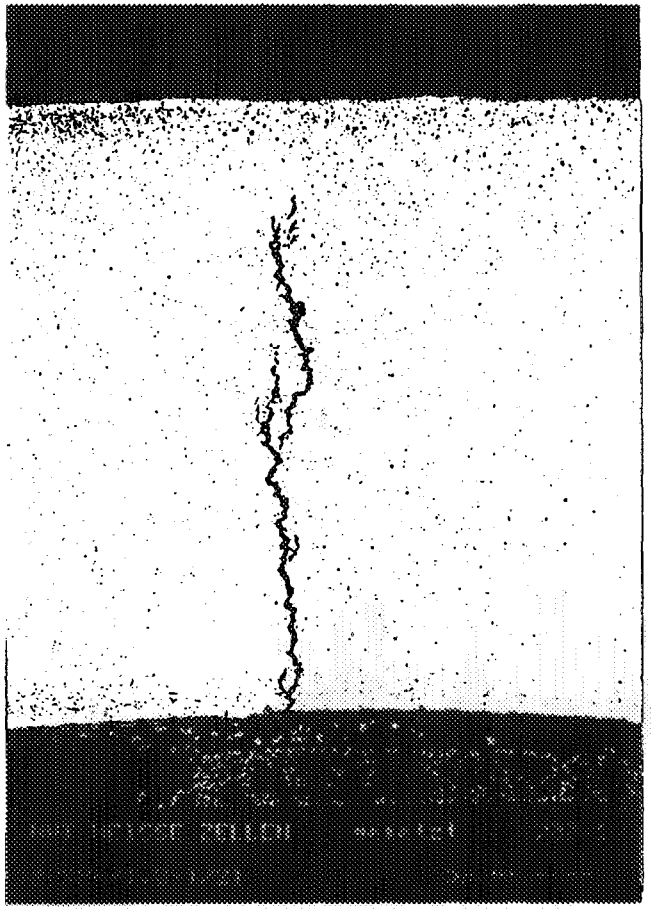

Figure 17: Micrograph of the defect of the fuel rod A.1.3

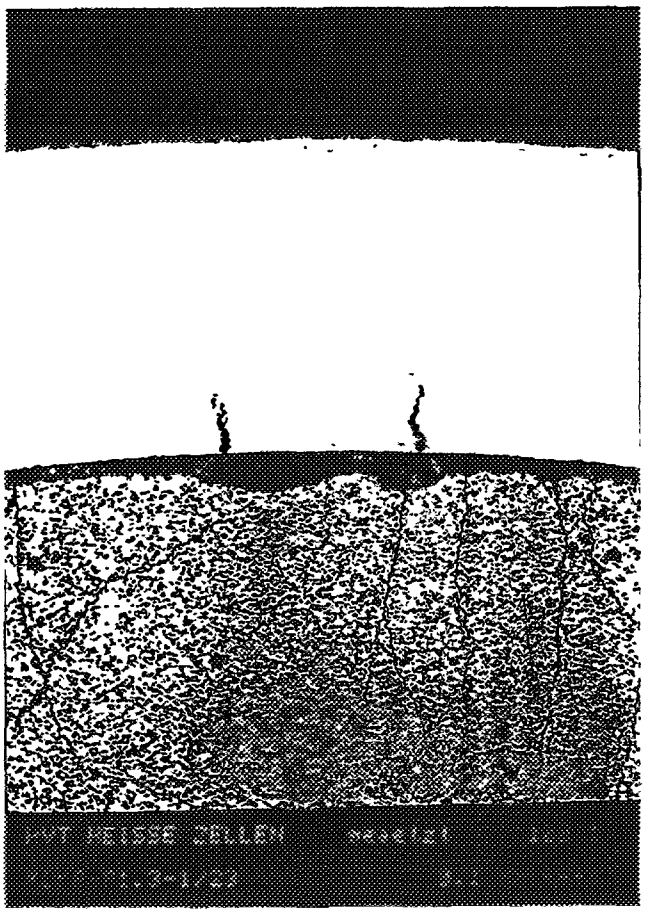

Figure 18: Micrograph of a crack in the cladding of the fuel rod A.1.3 in other section that support maximum stresses (same height at the defect).

The BACO code development began in 1974 in CNEA. The latest version of the code takes advantage of the hardware and software new tools, incorporates new behaviour models to manage a new set of experiments, and new requirements of design.

The IAEA's CRP FUMEX (Co-ordinated Research Programme on Fuel Modelling at Extended Burnup) required a preliminary work with the BACO code in order to verify, to correct and to change models, calculations structuring and convergence analysis, leading to BACO version 2.20 employed in this work.

The job schedule was a feedback work, the code was used to define the irradiation experiment and, in the next stage, the BACO output was employed to up-grade the code.

5.2 Calculation Results

Due to the appearance of the defect in rod A.1.3 a more detailed calculation with the BACO code has been made.

The power axial profile was obtained from *Fig. 2* on Reference [14] (pre-irradiation stage), the *Fig. 2* from Reference [15] (pre-irradiation ramp stage) and from *Fig. 2* from Reference [16] (power ramp). We divided the MOX pin in 5 axial sections. The fifth axial section chosen corresponded to the

defective zone, where we observed the maximum peak level during the power ramp. *Figure 19* shows the power irradiation history used for the BACO calculations. *Figure 20* shows the power history for the defective zone in the A.1.3 fuel rod (the fifth axial section in BACO code).

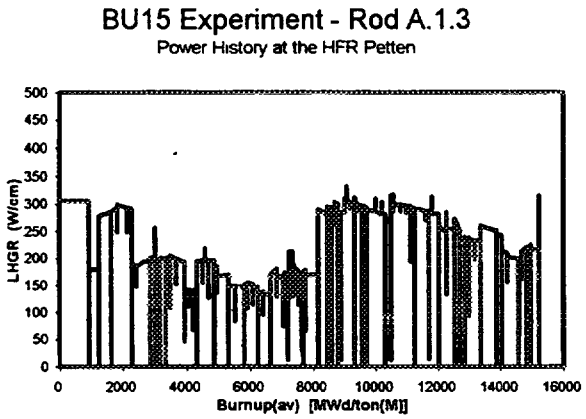

Figure 19: Power irradiation history for the A.1.3.

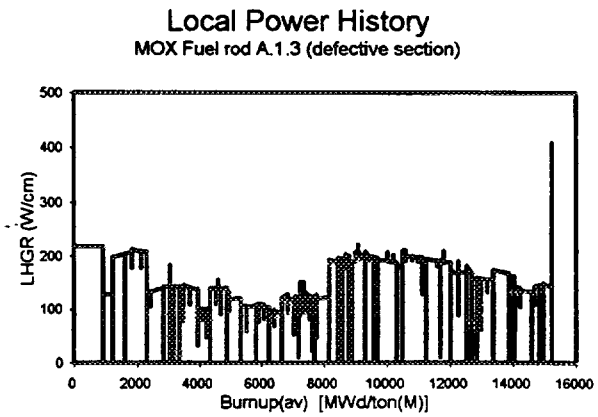

Figure 20: Local power history for the rod A.1.3 in the defective zone.

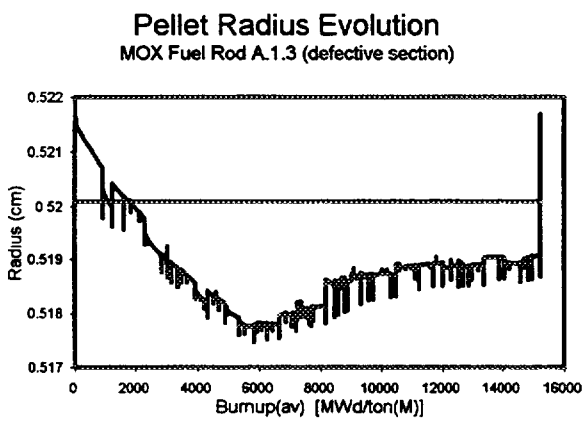

Figure 21: Pellet radius evolution for the fuel rod A.1.3 in the defective zone as it was calculated for the BACO code.

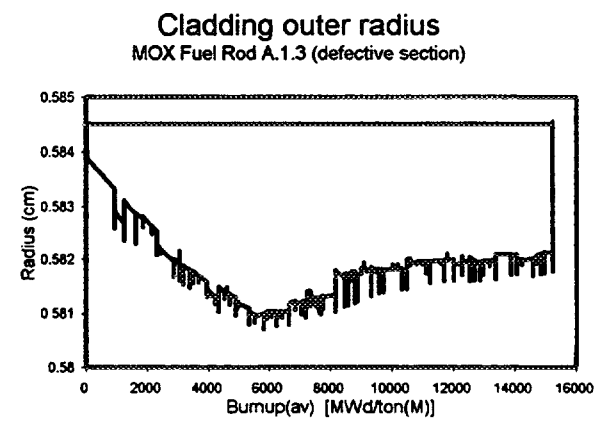

Figure 22: Cladding outer radius evolution of the fuel rod A.1.3 in the defective zone as it was calculated for the BACO code.

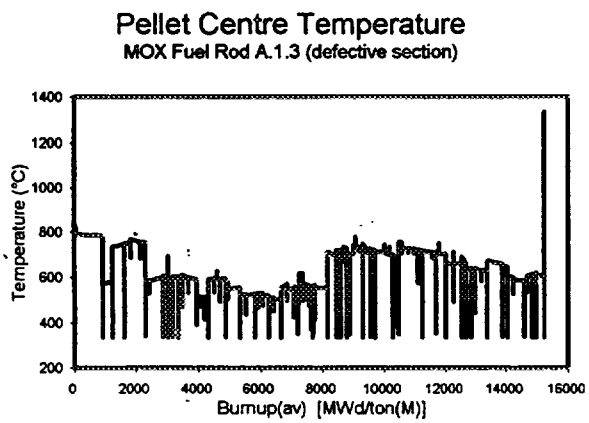

Figure 23: Pellet centre temperature. Defective zone in the A.1.3 fuel rod.

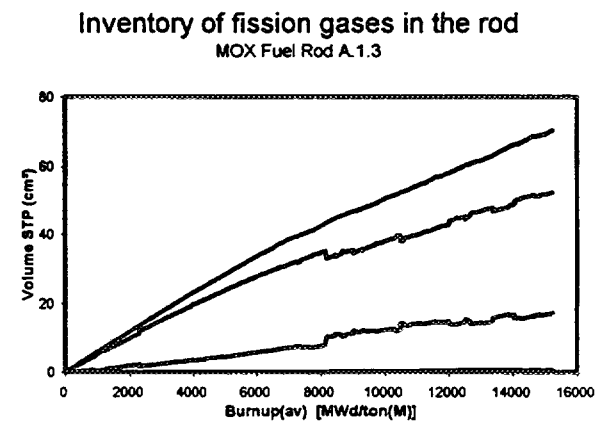

Figure 24: Fission gas inventory evolution in the rod A.1.3

Pellet and cladding contact appeared in the first days of irradiation due to the small gap and high power condition. The gap opened when shutdowns took place. It is easy to identify the densification and swelling behaviour (*Fig. 21*). *Figure 22* shows the evolution of the external radius of the cladding.

Pellet centre and cladding inner surface temperatures were acceptable. *Figure 23* shows the relative low temperature for the pellet centre of the defective axial section during pre-irradiation period and the increase in temperature in the final ramp.

Figure 24 shows the fission gases inventory evolution. We discriminate:

- produced fission gases (upper curve),
- released fission gases (lower curve, just above the x-axis),
- fission gases trapped in the UO_2 matrix, inside the grain (medium upper curve), and
- fission gases kept in grain boundary (medium lower curve).

There was a tendency to saturate the grain boundary.

Figure 25 completes the above information with the released gases fraction. The fraction released for the A.1.3 rod was 1.1 %. A complementary vision of free gases in the rods is shown in *Figure 26* where we discriminate de percentage of He, Xe and Kr in the free volume, there is a direct relationship between gap conductance and gas composition (see *Figure 27*).

There was stress reversal in the rod due to the shutdowns (*Fig. 28*). The hoop stress maximum was 208 MPa in the A.1.3. rod for the defective axial section. We can assume a direct relationship between the defect and that hoop stress. The defective zone supported the strongest mechanical demands and the value reached for the hoop stress is practically the SCC threshold. The biggest power increment was the last ramp:

$$Ql(end) - Ql(initial) = 290.W/cm$$

The radial contact stress between pellet and cladding was very strong too (*Fig .29*).

The final pressure in the rod due to free gases was about 0.45 MPa (see *Figure 30*).

We calculated a growth in the grain size in the central zone of the pellet from R_g (initial)= 6.2 μm to R_g (end)= 16 μm.

Table V shows the parameters calculated with BACO code. Results are comparable to those reported in (4.1). Due to the lower power, the final burnup for the A.1.3 fuel rod was lower than that of the assembly, and the local burnup in the defective zone was the lowest.

6. CONCLUSIONS

The maximum hoop stress and pellet-cladding radial contact pressure appears in the axial section corresponding with the failure (*Figs. 28* and *29*).

There was a good agreement between experimental results and BACO code predictions, specially with respect to the relationship between the failure position and characteristics and the mechanical demands predicted [7].

The presence of microcracks inside the cladding in the doped rods, the coincidence between the predicted and measured pellet-cladding gap values, the temperature calculated and the microstructure observed, the grain sizes distribution, indicated a good BACO code evaluation.

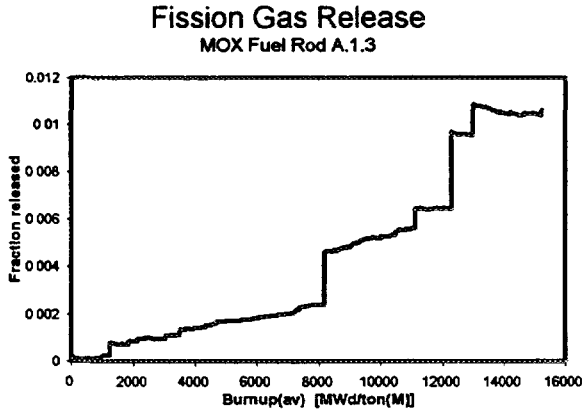

Figure 25: Fission gas release fraction in the fuel rod A.1.3.

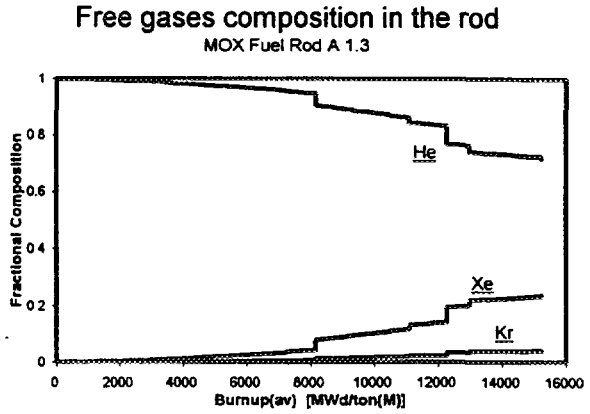

Figure 26: Gases composition in the fuel rod as it was calculated using the BACO code.

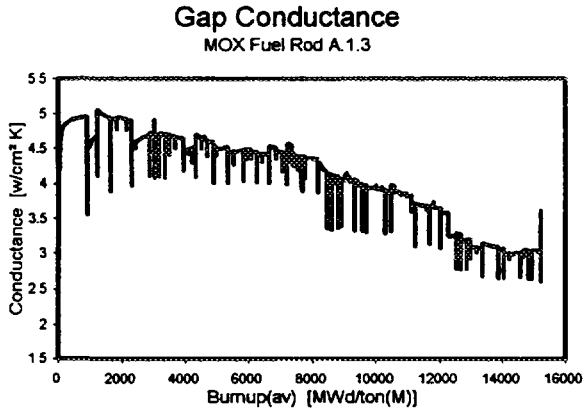

Figure 27: Gap conductance in the fuel rod as it was calculated for the BACO code.

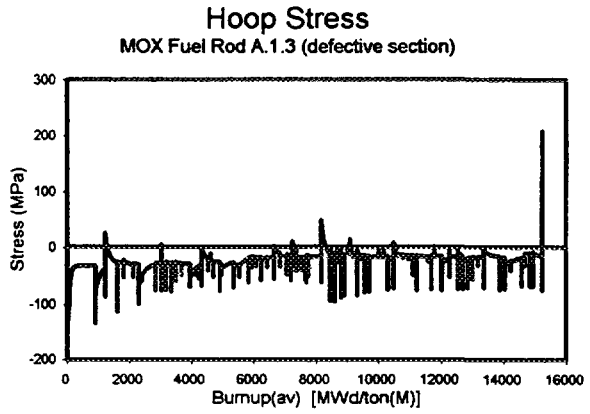

Figure 28: Hoop stress for the A.1.3 fuel rod in the defective zone.

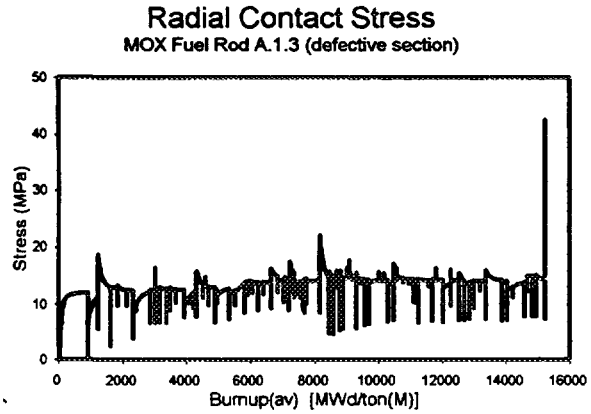

Figure 29: Radial pellet-cladding contact pressure. Defective zone in the A.1.3 fuel rod.

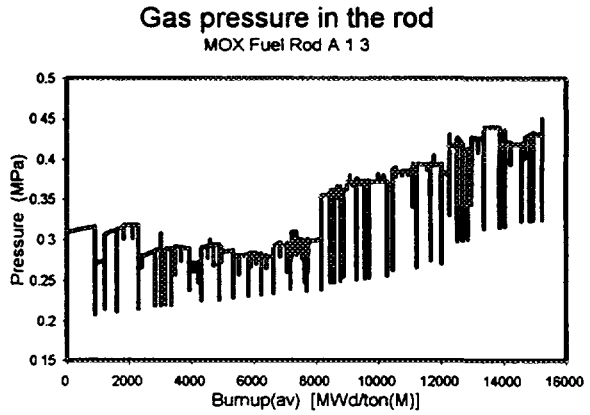

Figure 30: Gas pressure in the fuel rod as it was calculated for the BACO code.

The defective zone of the A.1.3 rod had the biggest mechanical demands. The calculated hoop stress was σ_v=208 Mpa. This value indicated that PCI-SCC was the mechanism likely to produce the failure. That prediction was confirmed with the post-irradiation examinations (*Fig. 17*).

The absence of fabrication induced failures in the irradiated rods is an important achievement in our MOX fuel development program.

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THERMAL AND IN-PILE DENSIFICATION OF MOX FUELS : SOME RECENT RESULTS

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Abstract

In-pile densification of PWR fuels is one of the main phenomena which determine the evolution of the pellet-clad gap during the first stage of the irradiation, and thus has consequences onto the thermo-mechanical behaviour of fuel rods. It can be predicted using the results of resintering tests and appropriate correlations. In this context, CEA, FRAMATOME and EDF have undertaken a joint research programme aiming to characterise the densification of MOX fuels. Different fuels were prepared by the MIMAS process using different UO_2 powders as matrix. After a detailed characterisation, fuel pellets were submitted to isothermal resintering tests and analytical irradiations. Correlations between in-pile and thermal densification were established. This paper presents the results obtained with two types of MOX fuel: one fabricated with the AUC UO_2 powder (ammonium uranyl carbonate conversion process) and another one fabricated with the SFEROX powder (peroxide conversion process). Isothermal resintering tests (up to 100 hours in various atmospheres) and out-of-pile characterisations (density measurement, ceramography, pore size distribution, ...) allowed to quantify the thermal densification kinetics of the selected fuels and to analyse the microstructural changes. As expected the disappearance of pores in the micron range was observed. Moreover, the influence of resintering conditions (gas humidity) was demonstrated. The irradiation was performed in SILOE reactor using a nucleate boiling rig simulating PWR conditions in terms of temperature, coolant pressure and neutron flux. Periodic measurements of the fuel stack shrinkage on neutron radiographs allowed to quantify the densification and its kinetics. The irradiation was carried on up to the observation of the maximum shortening. Results in terms of fuel stack shortening as a function of burn up were obtained at different operating power levels (in the range 15-25 $kW.m^{-1}$). The fission rate was confirmed as one of the main parameters of in-pile densification. The analysis of post-irradiation ceramographies, complicated by the heterogeneity of mixed oxides and the typical microstructure imposed by UO_2 powders , is expected to give information about the contributions of UO_2 matrix and Pu-rich agglomerates porosity. Assuming the isotropy of densification, axial shortening can be converted into volume variation in order to compare in-pile and out-of-pile results. The comparison indicates that, in spite of differences in mechanisms (influence of fission spikes for instance), the maximum densifications are equivalent in both cases : as for UO_2, resintering test results give good data to predict the in-pile behaviour of MOX fuels.

1. INTRODUCTION

Of the physical phenomena affecting the behaviour of sintered ceramics used in nuclear fuels, densification under irradiation plays a dominant role from the very start of irradiation in the reactor. In view of the effects induced by this phenomenon on the thermo-mechanical behaviour of the fuel rod, it is vital to have some means of predicting it. With this aim in mind, the experimental programme presented in this paper

was set up to study in-pile densification of two types of uranium and plutonium mixed oxide fuels (MOX) used to manufacture assemblies for pressurised water reactors (PWR). The difference between these two MOX fuels was the type of conversion process used to prepare the matrix UO_2 powder.

Established jointly by the French Atomic Energy Commission (CEA), a research organisation, Framatome, designer of fuel rods, and the French Electricity Board (EdF), this programme comprises two main parts:

- an out-of-pile part relating to the thermal stability study of the two types of MOX fuel,
- and an in-pile part dedicated to the analysis of irradiation-induced densification of these same fuels.

These two distinct experimental parts had the following common aims:

- first, validation of an out-of-pile densification test representative of fuel behaviour under irradiation, and which can be used in an industrial application,
- and secondly, development of a model describing the irradiation-induced densification of MOX fuels.

2. IN-PILE DENSIFICATION

The increase in density noted at the start of irradiation of sintered ceramics used in nuclear reactors leads to:

- a reduction in pellet height and thus in total length of the fissile column,
- and a reduction in mean pellet diameter which affects not only pellet thermal conditions by opening up the gas-filled pellet-cladding gap, but also the subsequent changes in the gap from a mechanical standpoint.

For these reasons, it was considered necessary to obtain a more accurate understanding of irradiation-induced densification and to develop tools for quantifying it, such as out-of-pile tests and modelling systems.

In-pile densification has already been the subject of numerous studies and the mechanisms involved have already been clearly identified [1-3]. The main cause of densification in the reactor is the elimination of the residual small-size porosity ($< 1\ \mu m$) remaining after sintering. This porosity is eliminated by bulk diffusion of vacancies from the pores.

The vacancies go into solution in the matrix from the pore surface and this mechanism is thermally activated but is also promoted under irradiation by pore-fission spike interaction. In the case of small-size pores (< 50 nm), a fission spike mays lead to complete destruction of the pore [4], whereas, for larger-size pores, the erosion process is progressive.

The vacancies in solution in the matrix diffuse towards sinks formed by grain boundaries and free surfaces. Naturally, the diffusion rate is governed by temperature, but also by the fission rate because of the creation of point defects. In the athermal field, irradiation accelerates diffusion, and the increase in diffusion coefficient is approximately linear with respect to fission density [1].

The main parameters coming into play in irradiation-induced densification are as follows:

- temperature, which affects the kinetics and, to a lesser extent, the maximum densification limit [5],
- fission density for the previously explained reasons (it is generally assumed that, below 400-450°C, densification is athermal),
- initial density and especially the oxide pore size distribution given that small-size pores have the biggest impact on densification,

- to a lesser extent, the grain size which governs sink efficiency with respect to the vacancy sources formed by the pores.

These mechanisms are of a general nature and are applicable to all sintered fuel ceramics; however, behavioural differences are to be expected between UO_2, which has already been widely studied, and the MOX fuel made by the MIMAS process [5-6]. This is because the manufacturing process consists in diluting a plutonium-rich master blend (PuO_2 content ~ 25 to 30%) in depleted UO_2 and gives rise to a microstructure including:

- agglomerates of master blend up to one hundred micrometres in size,
- a UO_2 matrix in which the master blend is diluted,
- in certain cases, accumulations of pure UO_2.

From this description, it is easy to understand why MOX fuels are likely to present specific behavioural features with respect to in-pile densification. For example:

- fission reactions are restricted to the plutonium-rich areas and this leads to locally high fission densities (about 4 times higher than the mean value for the pellet),
- as pores are eliminated, the Pu homogeneization occurs at the same time,
- since the presence of Pu changes the diffusion coefficient value, its distribution in the material thus has an effect on densification,
- the UO_2 powder used for dilution plays a role as a result of its specific properties and more particularly its morphological properties (size of elementary crystallites, specific surface area, agglomeration and oxidation characteristics, etc.).

3. EXPERIMENTAL ASPECTS

3.1 Fuels studied

Within the context of this programme, two batches of MOX fuel pellets were studied. They were manufactured in the laboratory (at DEC/SPU/LCN) according to PWR specifications by the MIMAS process from wet route UO_2 powders. The batches were as follows:

- batch A: UO_2 powder obtained by the AUC (ammonium uranyl carbonate) conversion process,
- batch B: UO_2 powder obtained by the conversion process using peroxide.

The master blend is enriched with 25% plutonium.

The pellets of each fuel batch were accurately characterised:

- determination of plutonium content: the Pu/(U + Pu) ratio is 6%, a value which is close to the maximum content of the fuel assemblies currently loaded in French PWRs,
- measurement of the hydrostatic density and characterisation of open porosity: the density of the pellets studied is close to 95.5% of the theoretical density, the open porosity is very low for batch B (less than 0.1%) whereas, for batch A it reaches a value of 0.3%,
- determination of the O/M ratio = 1.999,
- detailed ceramographic examination in order to characterise the microstructure.

The ceramic micrographs confirmed the heterogeneous structure of the materials studied, a typical characteristic of the MIMAS process. Some large-size (greater than 100 μm) master blend agglomerates were noted and batch B contained, in addition, some very compact pure UO_2 agglomerations of approximately spherical shape (Fig. 1).

The porosity was found to be distributed throughout the various components of the microstructure and was thus characterised for the microstructure as a whole. The initial pore size distribution of the two fuel

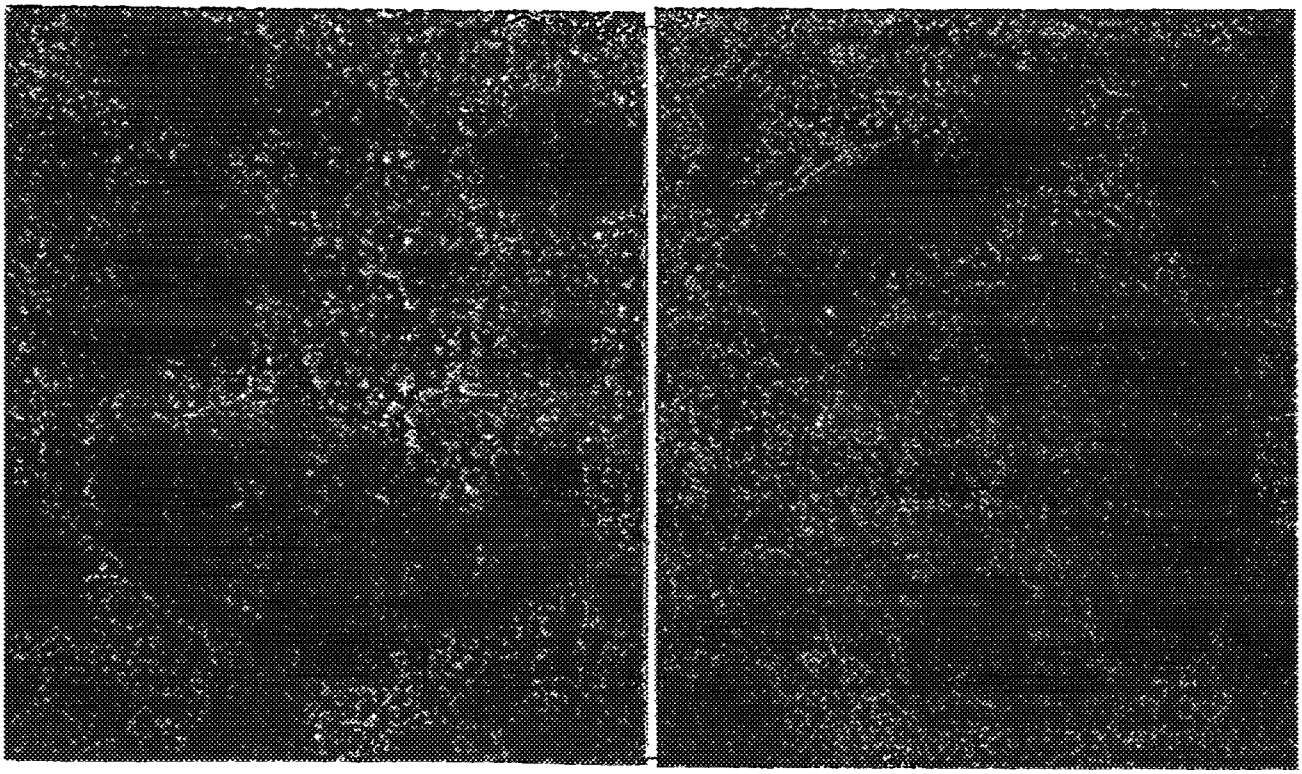

Fig. 1 : Micrographs of batches A anb B : initial state of the microstructure

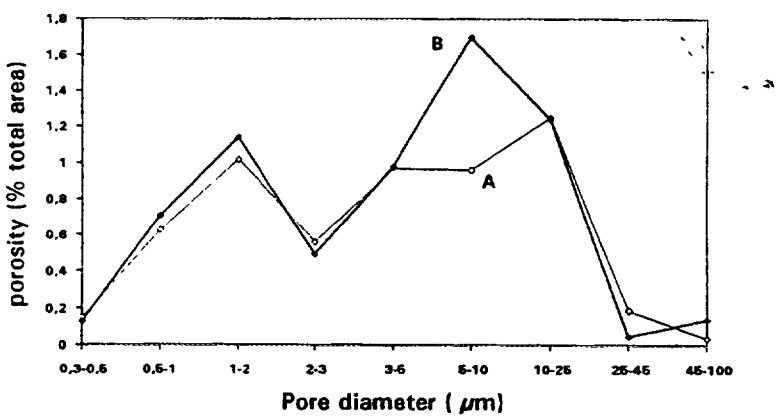

Fig. 2 : Initial pore size distribution of batches A and B

batches studied are shown in Fig. 2. The bimodal shape of the porosity distribution is more distinctive in batch B as a result of the addition of a porogen during its manufacture. This porosity generator is in fact responsible for part of the medium-size pores (between 5 and 10 μm).

3.2 Out-of-pile tests: thermal stability

These tests were conducted in the LEFCA laboratories (DEC/SPU) which has the necessary facilities to analyse the physico-chemical properties of non-irradiated plutonium fuels.
The tests consisted in carrying out annealing heat treatment at 1700°C for variable periods, ranging from 24 to 100 hours. The annealing atmosphere is identical to that used during sintering: a slightly humidified argon-hydrogen mixture.

After this heat treatment, the pellets are characterised:

- hydrostatic and bulk densities are measured by immersion in bromobenzene,
- the pore size distribution is characterised by micrographic image analysis,
- the O/M ratio is determined in order to check that the fuel has not been reduced during resintering, as this would have a direct bearing on the mesh parameter and thus on the theoretical density of the material.

3.3 In-pile experiment: monitoring of fuel pellet shortening

The aim of the fuel irradiation experiments performed in the Siloé reactor in Grenoble by DTP/SECC was to monitor the variation in length of stacks of pellets in order to characterise the densification kinetics and maximum value.

The fuel rods containing the pellets to be studied (batches A and B as previously described), with 17x17 standard PWR diametral geometry, are irradiated in the Aquilon device [7]. This is a nucleate boiling capsule capable of reproducing the operating conditions of a fuel rod in a PWR core, in terms of pressure and temperature. With a pressure of 13 MPa, the outside temperature of the fuel cladding at saturation is 330°C. This device is placed at the periphery of the Siloé reactor core in a neutron flux gradient, thus allowing the irradiation power to be adjusted by varying the distance between core and fuel rod.

Each fuel rod is irradiated for five cycles of the Siloé reactor, equivalent to about 100 days, at virtually constant power. This power is between 225 and 235 W/cm in the fuel rod median plane, giving final burn ups of 4150 $MWd.t_m^{-1}$ for batch A, and 4450 $MWd.t_m^{-1}$ for batch B. Given the height of the fissile column (600 mm), equivalent to that of the Siloé core, the experiment benefits from an interesting neutron flux axial gradient (and thus fission density) for the in-pile densification study: the power is about 115 W/cm at the top end of the fuel rods.

On completion of each irradiation cycle (every 20 days), the fuel rods are removed from the irradiation device and placed in a special container in order to be able to take neutron radiographs on the installation placed in the Siloé reactor pool [8]. Another fuel rod, identical to the two rods studied, is placed in this container to serve as a reference for shortening measurements and to eliminate some of the causes of uncertainty (estimated magnification, film expansion, etc.). The films (five per rod) are then analysed by suitable optical apparatus in order to characterise the shortening of different parts of the fissile column (1 part = 10 pellets). By dividing the fuel stack into parts in this manner, the observed shortening can be associated with the irradiation power through the previously described axial gradient. The residual power due to fission products is taken into account to estimate the fuel rod temperature and thus its expansion at the time the neutron radiography was taken so as to make the necessary corrections. This correction was then validated by taking a series of neutron radiographs after a sufficiently long cooling time to ignore the residual temperature rise.

The experiment was completed by destructive tests carried out at the LAMA, hot laboratory of the DTP/SECC. Macrographs and micrographs were taken in order to observe the change in microstructure during irradiation and, more particularly, to show the change in porosity.

4. RESULTS

4.1 Thermal stability test results

The results of resintering tests under a humid atmosphere are presented in Fig. 3 showing the change in density as a function of heat treatment time. For batches A and B, it was found that the asymptotic limit of "bulk" density increase (taking into account both closed and open porosity) is reached after about 70 hours. The value of the asymptotic limit of d/d is around 1.05% for batch A and 0.85% for batch B. Both batches have equivalent kinetics.

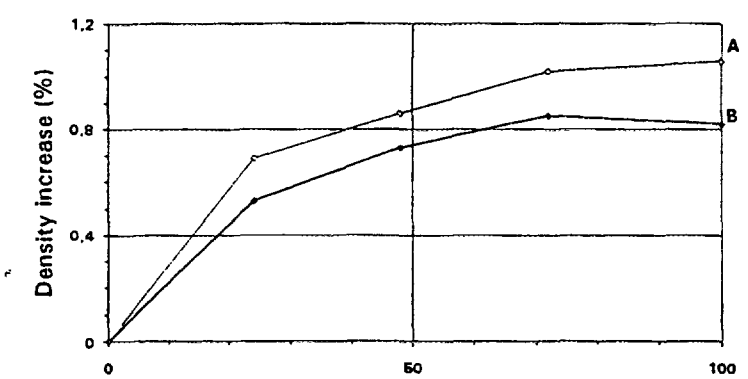

Fig. 3 : Out-of-pile thermal stability tests : variation in bulk density of batches A and B as a function of resintering time

However, with batch A it was noted that the initial open porosity disappeared during the first hours of treatment, thereby explaining the greater densification of this batch.
The surface porosity distributions obtained after 100 hours of treatment were compared to the initial ones (Fig. 4 and 5) in order to highlight the preferential elimination of small-size porosity, i.e., of diameter less than 2 μm. It was also found that some of the other pores in other classes also disappeared. However, since these pore size distributions were related to the surface and not to the volume of the sample, they cannot be used to draw anything other than qualitative conclusions concerning the variation in porosity.

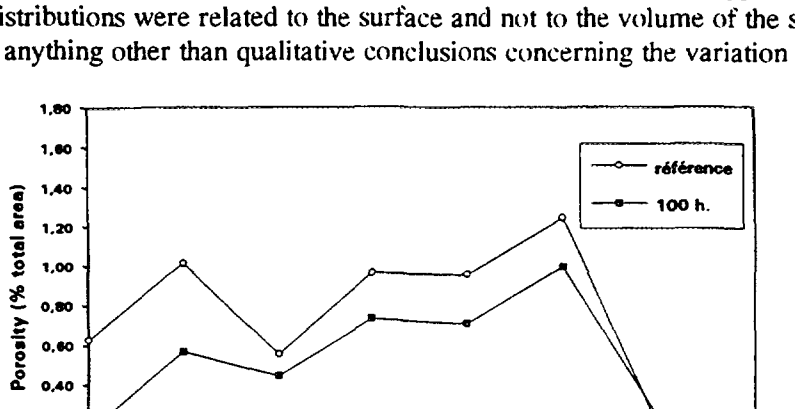

Fig. 4 : Evolution in pore size distribution of batch A : initial state and after 100 hours at 1700 °C

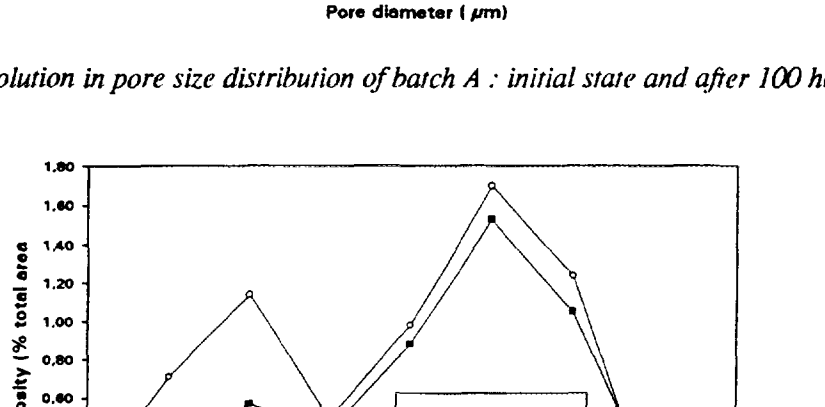

Fig. 5 : Evolution in pore size distribution of batch B : initial state and after 100 hours at 1700 °C

The ceramographs also show an enlargement of grain size (by a factor of two on average), especially in master blend areas. This phenomenon is perfectly normal given the temperature level (1700°C) and the holding time (up to 100 hours).

4.2 Results of in-pile experiments

The results obtained can be presented in graphical form: $\Delta L/L = f$(local burn up, local operating power). However, a number of corrections must be made in order to compare them with the results of out-of-pile tests.

The results are first transposed into volume variations by applying the following relationship, assuming the phenomena involved to be isotropic: $\Delta V/V = 3 \times \Delta L/L$. In addition, the solid swelling due to the accumulation of fission products in the material is subtracted by application of the following simplified law, applicable to UO_2, the validity of which has been checked on density measurements taken on MOX fuels irradiated in PWR:

$$(\Delta V/V)_{densification} = 3 \times \Delta L/L_{measured} - (\Delta V/V)_{solid\ swelling}$$

where $(\Delta V/V)_{solid\ swelling} = 0.6\%/(10000\ MWd.t_m^{-1})$

N.B.: in the case of MOX, allowance must not just be made for solid swelling because gaseous swelling was also observed in Pu-rich areas, even at low burn up.

The processed results are presented in Figs. 6 and 7 respectively for batches A and B.

It was found that:

- maximum densification would seem to be reached, for both fuel types, for pellets subjected to powers in excess of 170 W/cm, at a burn up of 2500 $MWd.t_m^{-1}$,
- the value of $(\Delta V/V)_{densification}$ is around -0.9% and -1.0% for batches A and B respectively,
- densification depends on the operating power; the pellets at the fuel rod ends, subjected to lower power levels, did not densify as much.

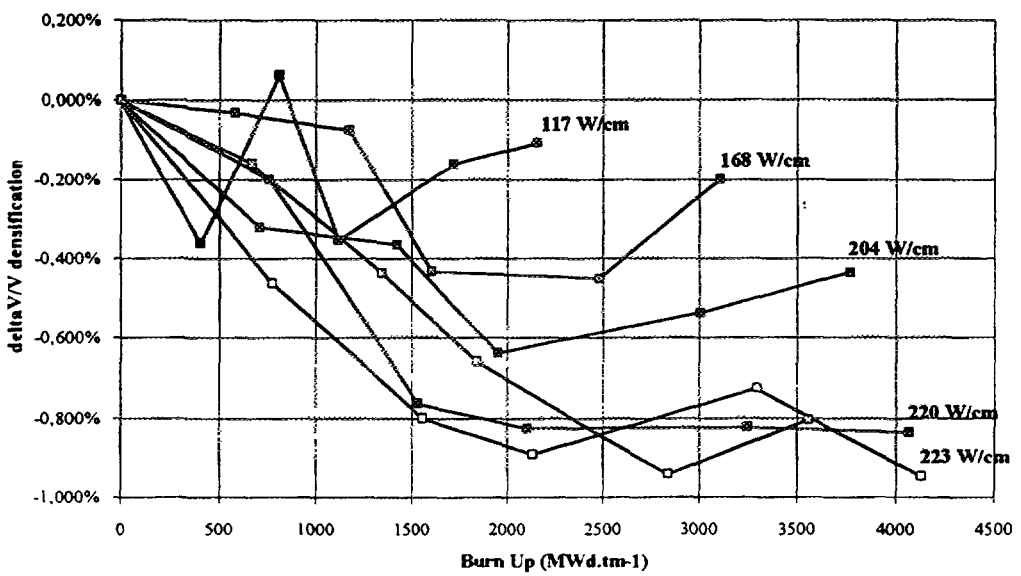

Fig. 6 : In-pile densification of batch A as a function of burn up and irradiation power

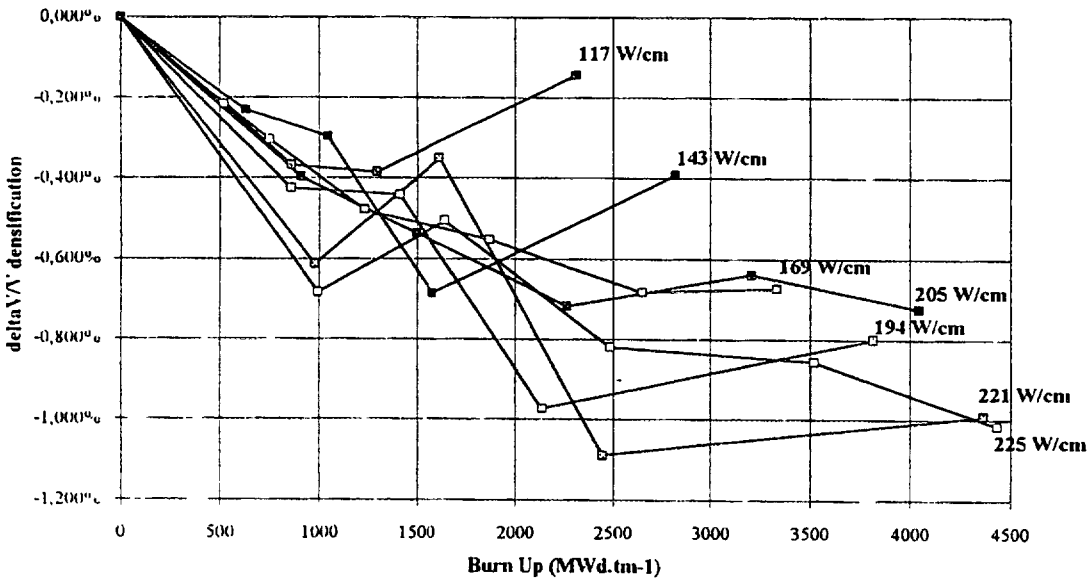

Fig. 7 : In-pile densification of batch B as a function of burn up and irradiation power

The ceramographic observations show irradiation-induced radial variation of the porosity in both cases, with the residual porosity being greater around the circumference than at the centre of the pellet, both in Pu-rich areas and in the UO_2.

At the same time, in the Pu-rich areas at the centre of the pellets, the appearance of metallic fission product precipitates and the nucleation of bubbles of inter-granular fission gases were noted (Fig. 8). This is consistent with the local temperatures close to 1000°C and especially with the local burn ups of the master blend agglomerates : close to 20000 $MWd.t_m^{-1}$ for a mean pellet burn up of 4500 $MWd.t_m^{-1}$.

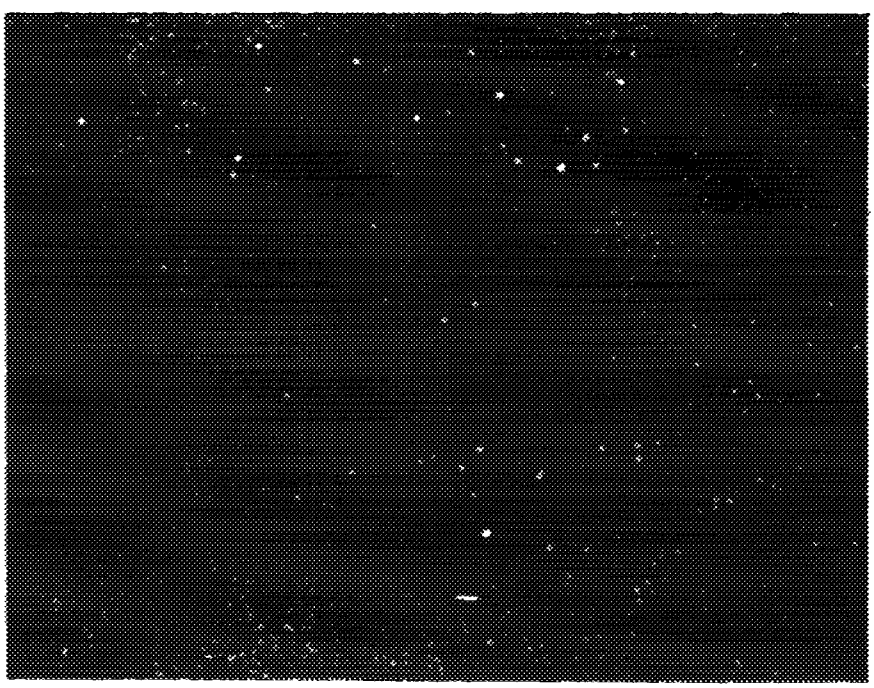

Fig. 8 : Micrograph of batch A after irradiation (mean burn up 4 $GWd.t_m^{-1}$) : pellet central zone

[1] about 1.4 x 10^{13} fissions.s^{-1}.cm^{-3}

5. DISCUSSION

For a variety of reasons, it is not possible to make a direct comparison of the results acquired out of pile and in pile.

Out of pile, the change in overall density of the pellet is measured directly (including or excluding the open porosity). For the in-pile tests, however, it is the change in height that is being measured. It has already been seen that, assuming isotropy, dimensional variations can be converted into volume and thus density variations. On the other hand, when the length of dished pellets is measured, as is the case here, only the densification of the peripheral area of the pellets, delineated by the support surfaces between pellets (1.6 mm thickness), is characterised.

Consequently, the operating temperatures of the zones concerned in the in-pile study are less than 500°C and the observed densification takes place in a temperature domain close to the domain considered to be athermal with respect to the phenomenon. On the other hand, the densification occurring during the out-of-pile test takes place at high temperature (1700°C).

Despite this fundamental difference, the asymptotic densification limits obtained both in pile and out of pile appear to be equivalent, taking into account experimental uncertainties:

- 0.9 $^{+}/_{-}$0.15 % in pile for zones having operated at 225 W/cm^{1}, compared to 1.05 $^{+}/_{-}$0.03 % out of pile for batch A,
- 1.0$^{+}/_{-}$0.15 % in pile for zones having operated at 225 W/cm, compared to 0.85$^{+}/_{-}$0.03 % out of pile for batch B.

This would seem to indicate that the mechanisms involved, whether purely thermal or combined (thermal + irradiation), do not have a predominant effect on maximum densification which no doubt depends more on the microstructure, at least for temperatures in excess of 400°C: the small-size porosity is in fact responsible for densification, regardless of its elimination mechanism.

On the other hand, the densification kinetics must be affected by the mechanisms involved because, out of pile, maximum densification is reached in 70 hours whereas more than 1000 hours are needed in pile. It is indeed preferable, for the in-pile case, to use the burn up parameter to express the in-pile densification.

The lower degree of densification of the end zones of the fuel rods - which operated at lower powers - may be explained either by a lower maximum densification limit or by slower kinetics resulting in the limit not being reached. In fact, there is every likelihood that the phenomenon involves a combination of both aspects because the zones of lower power operate at :

- lower fission densities, the effect on densification kinetics is approximately linear,
- lower operating temperatures as a result of the principle of the irradiation device: at low powers, saturation is not achieved and the cladding temperature is highly dependent upon the power. At the ends of the fuel rod, the peripheral fuel temperature is close to 350°C.

Given the heterogeneous structure of the studied MOX fuels, the difficulty of accurately characterising the variation in porosity in this type of material and the absence of temperature overlaps between the domains explored out of pile and in pile, it would seem risky at this stage to try and develop a detailed mechanistic model of in-pile densification of MOX fuels. It would seem more reasonable to adapt a semi-empirical densification law integrating both the thermal stability test results for the maximum densification limit and the irradiation results for kinetic aspects.

6. CONCLUSIONS

The analysis of the experimental results shows a similitude of behaviour of the two Mimas MOX fuels obtained from wet route UO_2 powders : out-of-pile resintering tests and operation under irradiation lead to similar final densifications despite the differences in kinetics. This would tend to indicate that the microstructure of the material plays a dominant role with respect to the operating conditions, and that the resintering test is representative.

For the modelling of in-pile densification, the use of out-of-pile test results allows implicit integration of microstructural effects such as density, grain size, pore size distribution, etc. Moreover, the variation in external diameter of the pellet is governed above all by the densification of the peripheral zone, the faster densification of the central zone (due to higher temperature) being of lower influence. Thus, the adjustment of densification kinetics on the shortening kinetics of the peripheral pellet zone is relevant with regard to the thermo-mechanical modelling of the fuel rod in which the evolution of the pellet-clad gap is one of the main parameters.

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AECL's EXPERIENCE IN MOX FUEL FABRICATION AND IRRADIATION

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Abstract

Atomic Energy of Canada Limited's mixed-oxide (MOX) fuel fabrication activities are conducted in the Recycle Fuel Fabrication Laboratories (RFFL) at the Chalk River Laboratories. The RFFL facility is designed to produce experimental quantities of CANDU* MOX fuel for reactor physics tests or demonstration irradiations. From 1979 to 1987, several MOX fuel fabrication campaigns were run in the RFFL, producing various quantities of fuel with different compositions. About 150 bundles, containing over three tonnes of MOX, were fabricated in the RFFL before operations in the facility were suspended. In late 1987, the RFFL was placed in a state of active standby, a condition where no fuel fabrication activities are conducted, but the monitoring and ventilation systems in the facility are maintained. Currently, a project to rehabilitate the RFFL and resume MOX fuel fabrication is underway. The initial campaign will consist of the production of thirty-eight 37-element (U, Pu)O_2 bundles containing 0.3 wt. % Pu in Heavy Element (H.E.) destined for physics tests in the zero-power ZED-2 reactor. An overview of AECL's MOX fuel irradiation program will be given. Post-irradiation examination results of (U,Pu)O_2 bundles irradiated to burnups ranging from 18 to 49 GWd/te H.E. in the Nuclear Power Demonstration reactor are highlighted. The results demonstrate the excellent performance of CANDU MOX fuel to high burnup, at power ratings up to 45 kW/m. The paper also outlines the status of current MOX fuel irradiation tests, including the irradiation of various (U, Pu)O_2 and (Th, Pu)O_2 bundles.

1. INTRODUCTION

AECL's MOX fuel program was started more than thirty years ago. The program consisted of two components:

- irradiation testing and post-irradiation examination (PIE) of various types of MOX fuel, with the objective of studying the general behaviour of these types of fuel in comparison with natural UO_2 fuel; and,
- development of MOX fuel fabrication technology.

Irradiation testing and PIE of MOX fuel is still continuing at AECL; it has gone from multi-element to multi-bundle demonstration testing. This paper discusses recent PIE results of (U,Pu)O_2 bundles irradiated in the Nuclear Power Demonstration (NPD) reactor, and outlines the status of current MOX fuel irradiation tests, including the irradiation of various (U,Pu)O_2 and (Th,Pu)O_2 bundles in the National Research Universal (NRU) reactor.

AECL's MOX fuel fabrication experience is summarized, including an update on the current fabrication campaign.

*CANDU: CANada Deuterium Uranium; registered trademark.

2. MOX FUEL FABRICATION AT AECL

2.1 Recycle Fuel Fabrication Laboratories

Various forms of fuel containing plutonium have been handled by AECL at its Chalk River Laboratories since 1960. Research activities were carried out in glove boxes between 1960 and 1970, including the development of MOX fuel fabrication technology, measurement of physical properties, production of fuel samples for experimental irradiation, etc. In 1970, a decision was made to re-model the plutonium laboratory and install new facilities, to focus on MOX fuel fabrication technology. Installation of the new facilities was complete by 1975 [1]. The facility, collectively referred to as the Recycle Fuel Fabrication Laboratories (RFFL), is designed to produce experimental quantities of MOX fuel for reactor physics tests or demonstration irradiations.

Subject to special precautions due to the presence of Pu (e.g., essentially all operations are done inside glove boxes), the processes employed in the RFFL follow conventional natural UO_2 practice. The fabrication line was designed for the production of sealed individual fuel elements, starting from UO_2 or ThO_2 powders as the major component and PuO_2 or $^{233}UO_2$ as the minor component.

The fabrication process adopted in the RFFL is outlined in Fig. 1. Weighed amounts of the starting oxide powders are mixed either in single-stage or double-stage blending, the latter being used for more dilute mixtures to achieve better homogeneity. After blending, the MOX powder is pre-pressed using an isostatic press, to convert the mixed powder into compacts, which are, in turn, fed into a granulator. The resulting free-flowing granules are then suitable for final pressing into green pellets using a single-cavity hydraulic press.

The green pellets are then loaded into a batch furnace, where sintering is done in a dilute hydrogen cover gas. Sintered pellets are then centreless ground to a specified diameter and surface finish. The pellets are washed and then dried in warm air. Acceptable pellets are loaded into empty sheaths that already have one endcap welded and all appendages brazed in place (these sub-assemblies are supplied by commercial fabricators). The second endcap is welded to the loaded sheath using a tungsten inert gas (TIG) welding system. The sealed elements are then helium leak-tested, scanned for surface alpha contamination, and assayed by neutron interrogation. Following assay, the elements are ready for bundle assembly.

2.2 Fabrication Campaigns in the RFFL

Extensive commissioning and test production programs using natural UO_2 were implemented in the RFFL. This enabled several modifications to, and optimization of, various process and safety-related equipment prior to the introduction of Pu in the fabrication line in 1979.

From 1979 to 1987, several MOX fuel fabrication campaigns were run in the RFFL, producing various quantities of fuel with different compositions [1]. As listed in Table I, the first campaign consisted of (U, Pu)O_2 fuel with 0.5 wt.% Pu in H.E. Later, about 1.3 Mg of (Th,Pu)O fuel elements were produced, containing up to 2.3 wt.% Pu in H.E. The last campaign was particularly challenging, since it involved the fabrication of 1350 elements containing 1.4 wt.% ^{233}U in H.E. (due to the presence of ^{232}U, which has a gamma-active daughter) [2].

The fuel elements and bundles produced in the RFFL were used for test irradiations in NRU, and for physics tests in the zero power ZED-2 reactor. About 150 bundles, containing over three tonnes of MOX were fabricated in the RFFL before operations in the facility were suspended. In late 1987, the RFFL was placed in a state of active standby, a condition where no fuel fabrication activities are conducted, but the monitoring and ventilation systems in the facility are maintained.

Currently, a project to rehabilitate the RFFL and resume MOX fuel fabrication is underway. The pre-operational testing (using inactive material) is scheduled for mid-1995, with regulatory approval to

resume MOX operations anticipated by late 1995. The initial campaign will consist of the production of thirty-eight 37-element $(U,Pu)O_2$ bundles containing 0.3 wt.% Pu in H E., destined for physics tests in ZED-2.

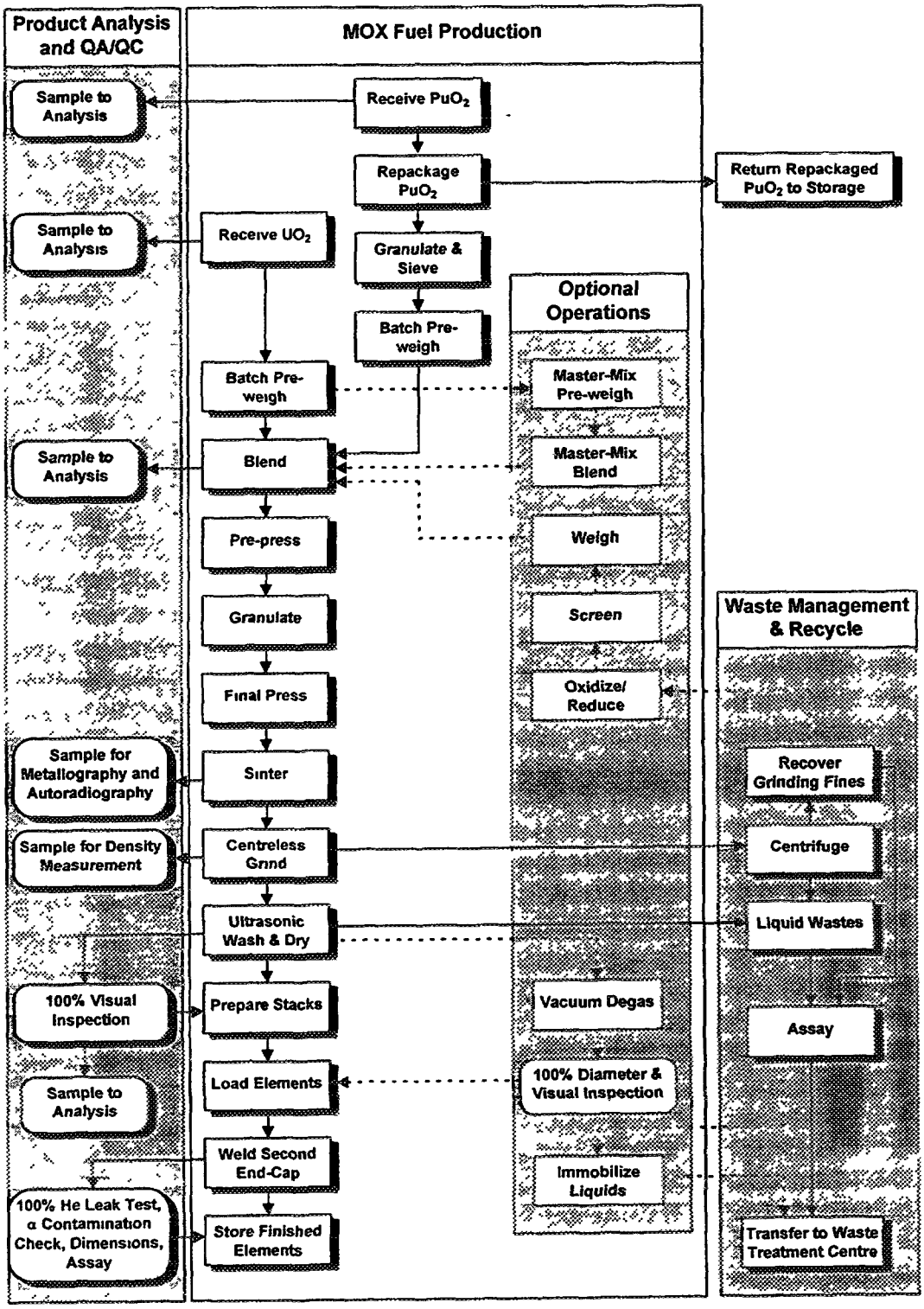

FIGURE 1. RFFL Fabrication Process Flowsheet.

TABLE I Fuel Produced in the RFFL.

Experiment	DATE	FUEL TYPE	QUANTITY
DP-12	1977-78	Natural UO_2	50, 19-element bundles
BDL-419	1979-80	(U, Pu)O_2 @ 0.5% Pu	15, 36-element bundles
BDL-422	1981-83	(Th, Pu)O_2 @ 1.75% Pu	6, 36-element bundles
BDL-430	1982	Natural ThO_2	1, 36-element bundle
WR1-1012	1982 1982	(Th, U)O_2 @ 1.8% Pu (Th, Pu)O_2 @ 2.3% Pu	2, 21-element bundles 2, 21-element bundles
WR1-1010	1982-85	(Th, Pu)O_2 @ 2.3% Pu	1332 elements
BDL-432	1986-87	(Th, U)O_2 @ 1.4% U-233	1350 elements

3. AECL's RECENT IRRADIATION EXPERIENCE (NPD-40)

3.1 The NPD-40 Experiment

The NPD-40 experiment was a demonstration-scale irradiation test involving six bundles containing (U,Pu)O_2 fuel in NPD. The objective of the experiment was to show that MOX fuel could operate successfully to extended burnup, under typical CANDU operating conditions. Subsequent to the NPD irradiation, several bundles were power-ramped in NRU.

3.2 Fuel Design and Fabrication

The six bundles were of the standard 19-element design, but contained two different types of non-standard fuel pellets from two different suppliers, and two sheath wall thicknesses. As listed in Tables II and III, three bundles had normal-(CANDU)-density hollow pellets (supplied by Aktiebolaget Atomenergi, Sweden) contained in thin-wall, collapsible Zircaloy-4 sheaths, while the other three had low-density solid pellets (supplied by BelgoNucleaire, Belgium) contained in thick-wall, free-standing Zircaloy-4 sheaths. Both pellet designs considered the effects of in-reactor operation:

- the low-density pellets had more internal voidage to accommodate pellet swelling during operation. In-service densification was accounted for by using free-standing sheaths; and
- the hollow design of the normal-density pellets served to reduce pellet thermal swelling, due to its lower average operating temperature.

The pellets were produced by both suppliers using normal fabrication methods; i.e., mixing and blending, pre-pressing, granulation, final pressing, sintering and grinding. In addition, the low-density pellets were degassed immediately prior to loading into the sheaths.

Zircaloy-4 sub-assemblies (i.e., appendaged sheaths with one endcap welded in place, and with graphite CANLUB coating on the sheath inside surface) were obtained from General Electric (GE) Canada, a commercial fuel supplier. Suitable endcaps for final-closure TIG welding were also supplied with the sub-assemblies. Glove boxes were used to load measured stacks of pellets and plenum inserts into the element sub-assemblies, and to fit the TIG endcaps. Final closure welds were then performed using a TIG welder.

Kits of 19 finished elements were sent to GE Canada for bundle assembly welding. The bundles were then returned to Chalk River Laboratories for inspection prior to irradiation in NPD.

TABLE II. Fuel Design Data for Bundles KA, KB and KC of Experiment NPD-40
(Dimensions in mm unless otherwise stated)

Bundle Design	19-Element Geometry
Overall Length	495.0
Overall Diameter	81.6
Element Design	
Overall Length	492.6
Internal Diameter	14.44
Wall Thickness	0.38
Sheathing Material	Zircaloy-4
$(U,Pu)O_2$ Weight/Element (g)	740.0
(U,Pu) Oxide Pellets	
Manufacturer	Aktiebolaget Atomenergi
Density (Mg/m^3)	10.5
Outside Diameter	14.2
Central Hole Diameter	2.54
Length	14.6
Enrichment	3.33 wt% (Pu + U) Fissile/HE
Weight $(U,Pu)O_2$/Bundle (kg)	14.06

TABLE III. Fuel Design Data for Bundles KD, KE and KF of Experiment NPD-40
(Dimensions in mm unless otherwise stated)

Bundle Design	19-Element Geometry
Overall Length	495.0
Overall Diameter	81.6
Element Design	
Overall Length	492.6
Internal Diameter	13.77
Wall Thickness	0.65
Sheathing Material	Zircaloy-4
$(U,Pu)O_2$ Weight/Element (g)	~685
(U,Pu) Oxide Pellets	
Manufacturer	Belgonucleaire
Density (Mg/m^3)	10.2
Diameter	13.67
Length	14.68
Enrichment	3.0 wt% Pu in (U + Pu)
Weight $(U,Pu)O_2$/Bundle (kg)	13.0

3.3 NPD Irradiation

Three bundles were loaded into NPD in 1973 January, and the other three were loaded in 1973 December. For most of the NPD irradiation, the bundles operated in pressurized heavy-water coolant at about 260°C and about 7 MPa pressure. Coolant was maintained at pH 10 with LiOH, and conductivity controlled to less than 1.5 mS/m.

As shown in Table IV, most bundles had an average element burnup of 14 GWd/te with the outer elements having a maximum burnup of 18 Gwd/te. (1 Gwd/te = 24 MWh/kg). The exception is bundle KB, which was operated to an average element burnup of 45 GWd/te, or an outer-element burnup of 49 Gwd/te. Figures 2-7 show the power history of each of the bundles; peak linear ratings ranged from 40 to 50 kW/m at beginning-of-life, decreasing to about 15-25 kW/m at end-of-life.

3.4 NRU Power Ramp Tests

Following the NPD irradiation, three bundles (one with normal density pellets, bundle KA, and two with low-density pellets, bundles KE and KF) were tested in NRU to observe MOX fuel performance during power ramps. After a few hours of low-power soak (about 20 kW/m linear rating), the bundles were ramped to 50 kW/m. This power ramp produced some defects in bundle KA. Bundles KE and KF survived and operated at 50 kW/m for 10 days. A further increase in power to about 70 kW/m resulted in a defect in one of the bundles, bundle KE.

3.5 Post-Irradiation Examination

All bundles were visually examined after the NPD irradiation. No peculiar findings were observed. Several bundles were measured for their element dimensional changes. In general, the elements showed slight diametral contraction from the unirradiated dimension. For the thin-wall, collapsible sheaths, measurements indicate a decrease in diameter up to a maximum of about 0.7%. For the thick-wall, free-standing sheaths, measurements also show a decrease in element diameters up to a maximum of about 0.3%.

After the power ramp in NRU, elements from bundle KA still indicated slight diametral contraction. After power ramping to 70 kW/m, outer elements of bundles KE and KF showed diametral expansions of about 1%. Inner elements of both bundles continued to show a diametral contraction.

Internal gases were extracted, measured and analyzed by mass spectrometry for all bundles except KD. As listed in Table V, the non-ramped bundles (KB and KC) had relatively low fission-gas release in the range from 2 to 4% of that produced for the outer elements. However, those elements that were ramped to powers above 60 kW/m (KE and KF) experienced high gas release in the order of 40 to 50% of that produced. Bundle KA, which was ramped to about 50 kW/m, had about 11% fission-gas release from the outer elements.

TABLE IV. NPD-40 Element Burnup (GWd/te H E.)

BUNDLE	Centre Element	Inner Element	Outer Element	Bundle Average
KA	7.1	9 2	17.5	14.2
KB	35.4	38.8	49.2	45.0
KC	7.1	9.6	17.5	14.6
KD	7.1	9.2	16.2	13.3
KE	7.1	8 8	15 4	12.9
KF	7 9	10.0	16.7	14.2

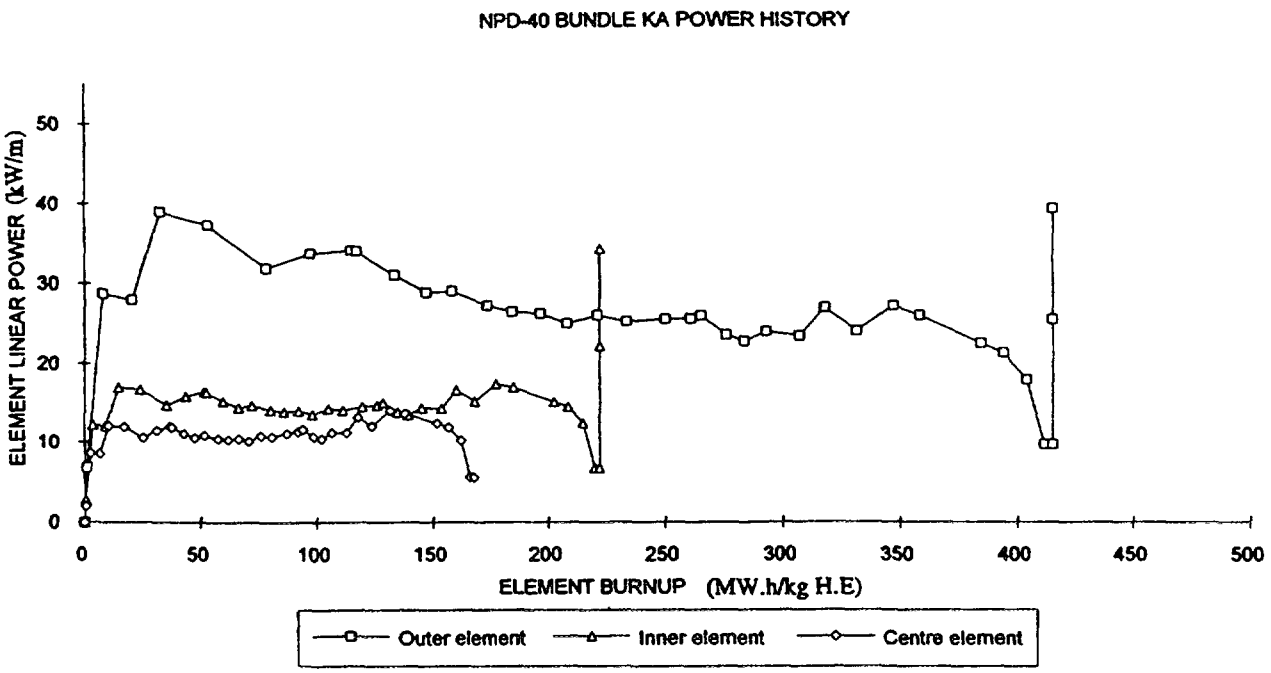

FIGURE 2. NPD-40 Bundle KA Power History.

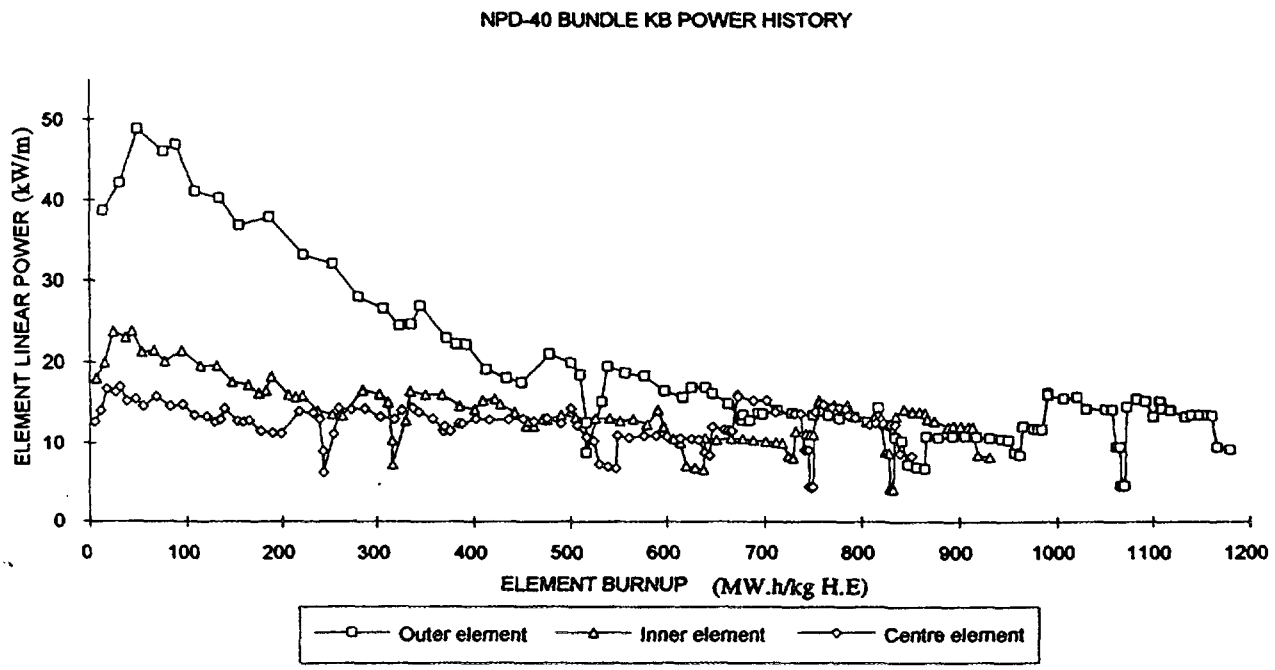

FIGURE 3. NPD-40 Bundle KB Power History.

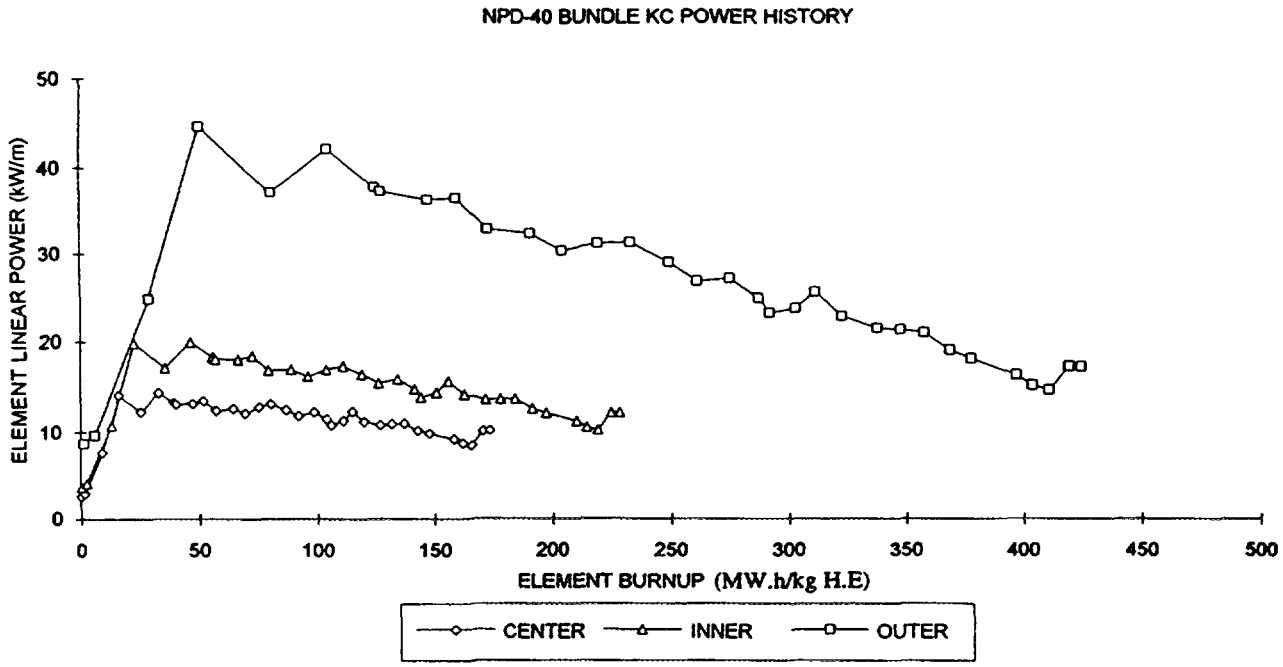

FIGURE 4. NPD-40 Bundle KC Power History.

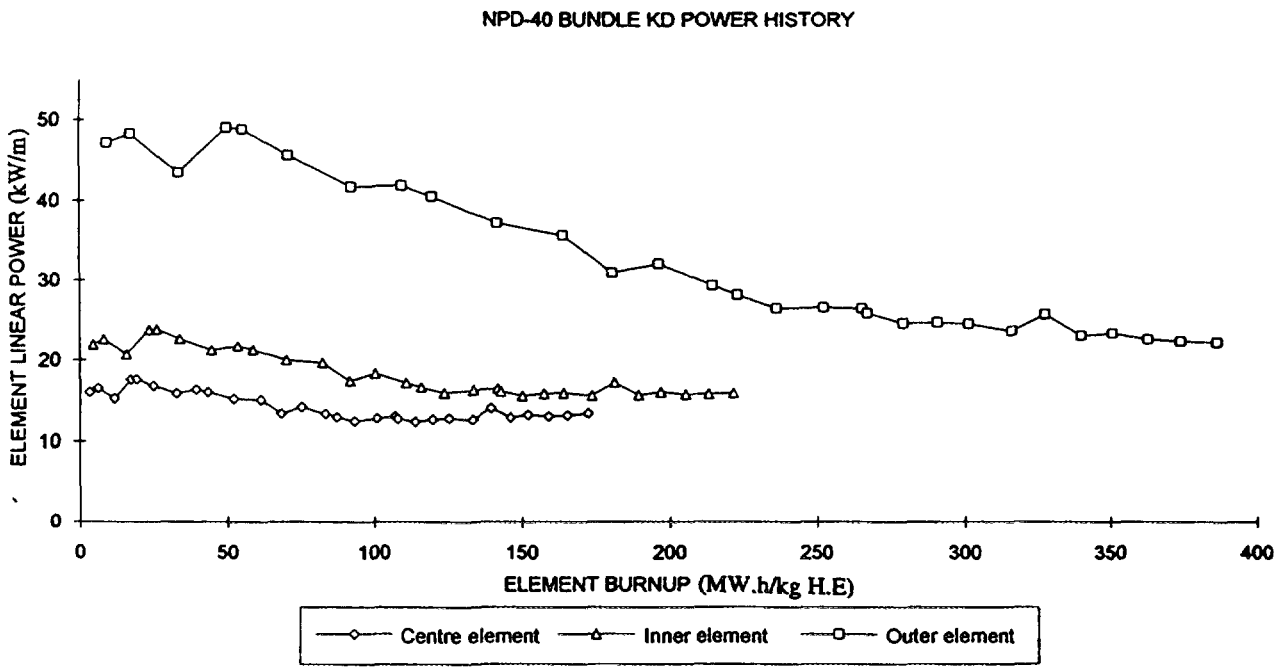

FIGURE 5. NPD-40 Bundle KD Power History.

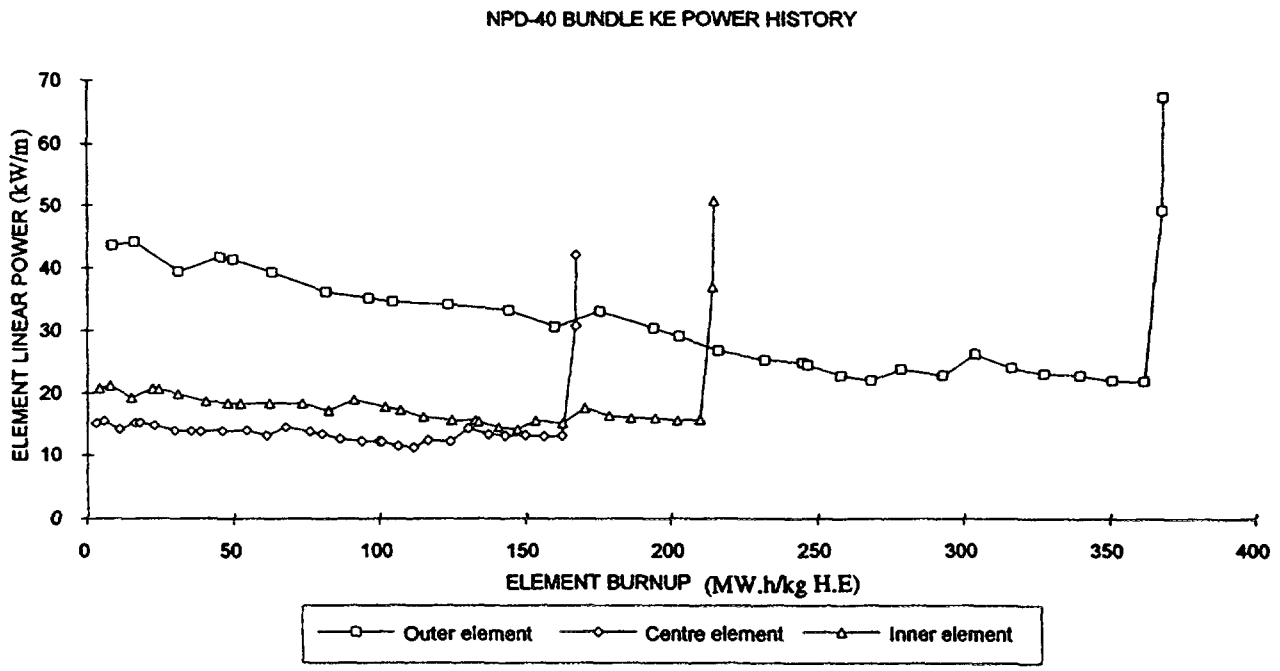

FIGURE 6. NPD-40 Bundle KE Power History.

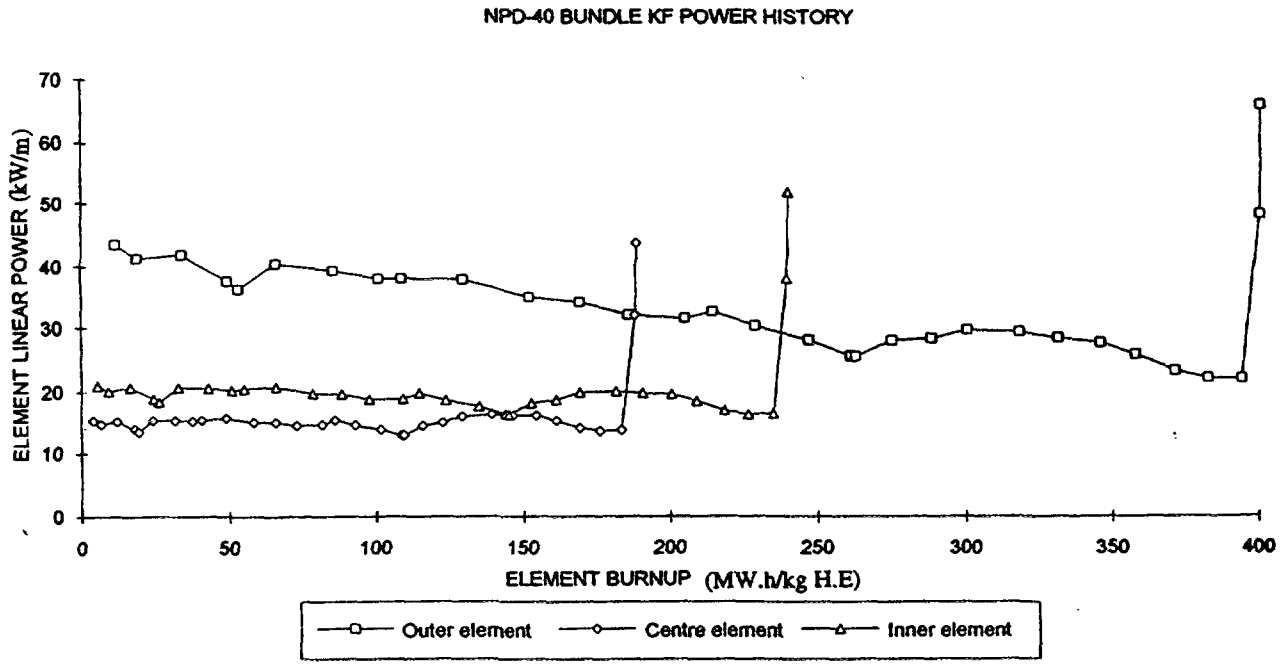

FIGURE 7. NPD-40 Bundle KF Power History.

TABLE V. Xenon Release for NPD-40 Bundles

Bundle-Element	Calculated Volume of Xenon Formed (mL)	Measured Volume of Xenon (mL)	Percentage of Xenon Released (%)
KA-01	780	86.9	11.1
KA-02	780	86.3	11.1
KA-13	401	13.3	3.3
KB-02	955	18.8	2.0
KB-04	956	38.9	4.1
KB-07	956	31.6	3.3
KB-10	954	40.6	4.3
KB-13	748	12.0	1.6
KB-16	749	9.9	1.3
KC-02	331	7.8	2.4
KC-13	178	0.3	0.2
KE-07	270	125.8	46.6
KE-08	271	129.3	47.7
KE-09	271	122.6	45.2
KE-10	271	127.6	47.0
KE-11	271	127.9	47.2
KF-02	293	142.2	48.5
KF-10	294	138.4	47.1
KF-17	173	43.4	25.1

NOTE: In the 19-element bundle geometry, elements 1-12 are located in the outer ring, elements 13-18 are in the intermediate ring, and element 19 is the centre element.

Metallographic examination showed that the microstructures had isolated regions of porosity and metallic fission-product precipitates (Fig. 8). These were attributed to the presence of high-fissile-content particles in the as-sintered pellet (Fig. 9), which resulted in zones of increased fission density and localized increase in temperature during operation. It was postulated that most of the fission-gas release was from these zones of initial high fissile Pu content.

PIE measurements indicated that elements containing low-density pellets had thicker CANLUB coatings than those containing normal density pellets (maximum thicknesses observed were 40 and 8 μm, respectively). There is no clear reason for this, although one possibility is the difference in as-fabricated coating thickness (the bundle kits were fabricated at different periods during 1972). Another possibility is the experimental error involved in determining the amount of coating retained. The thicker CANLUB coatings may have added to the benefits of low-density pellets resulting in the observed superior performance of these bundles during power-ramp tests in NRU.

3.6 Conclusions of NPD-40

This test demonstrated the excellent performance of dry-blended MOX fuel to high burnups (maximum of 49 GWd/te) at power ratings up to 45 kW/m. Dimensional changes and fission-gas release were minimal, and there were no indications of defects during the irradiation. There were no significant differences in the behaviour of the two fuel types (i.e., low-density pellets in thick-wall, free-standing sheaths and normal CANDU density annular pellets in thin-wall, collapsible sheaths).

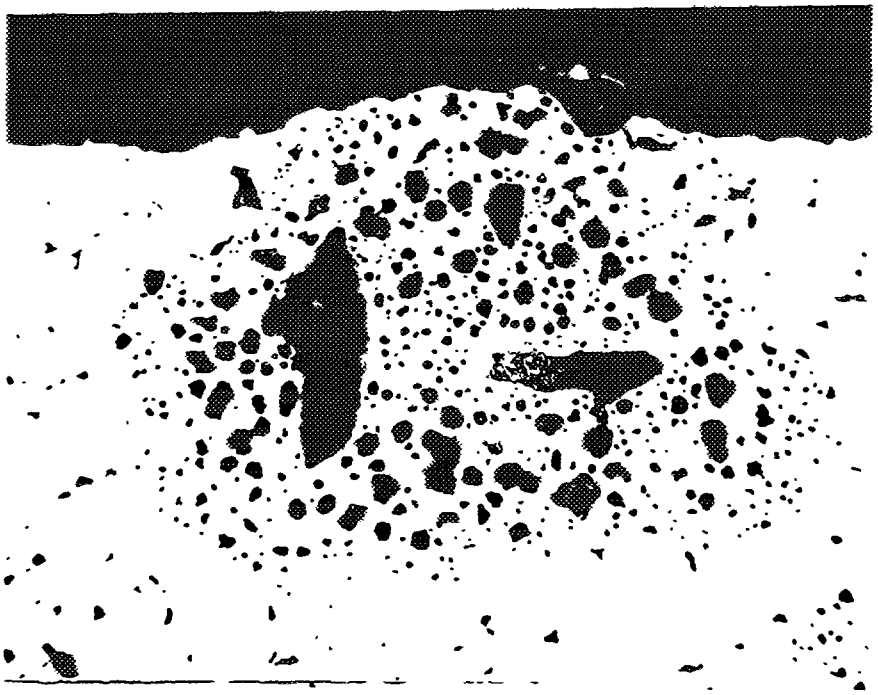

FIGURE 8. Microstructure of element KB-01 showing porous zone with metallic precipitates.

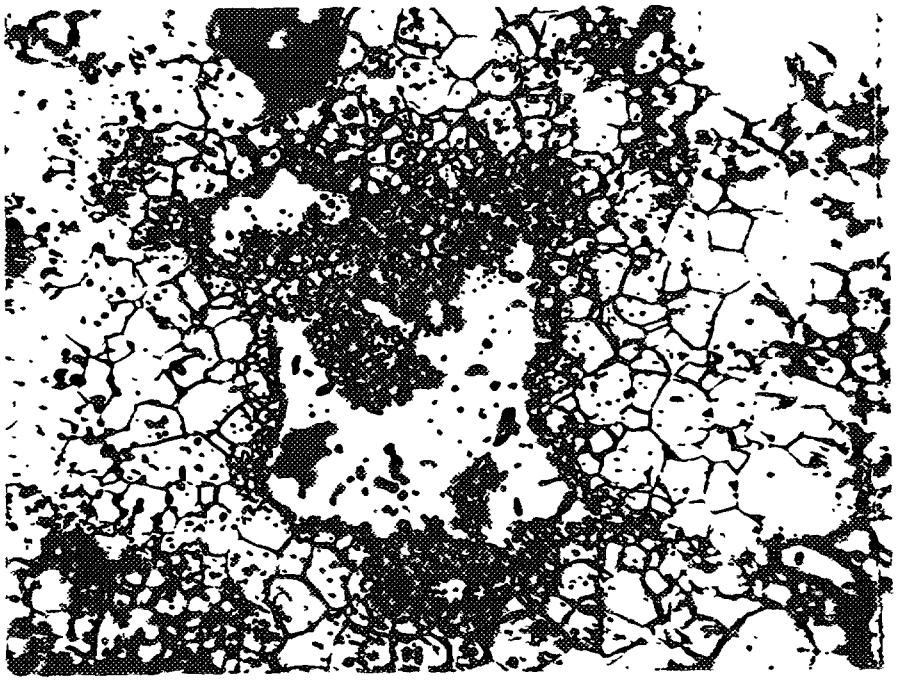

FIGURE 9. Microstructure of an archive pellet showing second-phase particles after etching.

4. CURRENT MOX FUEL IRRADIATIONS AT AECL

There are currently two MOX fuel irradiation tests in NRU: experiment numbers BDL-419 and -422. Both experiments involve demonstration of the performance of fuel bundles fabricated in the RFFL in comparison with UO_2 fuel. The bundles are of the standard 37-element Bruce design. BDL-419 contains $(U,Pu)O_2$ fuel with 0.5 wt% Pu in H.E., and BDL-422 contains $(Th,Pu)O_2$ fuel with 1.4 wt% Pu in H.E.

An initial set of six BDL-419 bundles was successfully irradiated in NRU up to burnups of 11-22 GWd/te at peak linear ratings of 35-65 kW/m. PIE of two bundles from this batch was recently completed, and documentation of the results is underway. Preliminary results indicate that the fuel performed very well, with fission-gas releases of less than 13%. A further set of six bundles from the same fabrication campaign are currently under irradiation in NRU. Outer element burnups are in the range of 7-12 GWd/te. In light of the excellent performance of the first set of bundles, the target burnup for the second set is under review, and will likely be increased to the range of 30-40 GWd/te.

BDL-422 consists of irradiation testing of six $(Th,Pu)O_2$ fuel bundles in NRU. Outer-element burnups are presently at 17 GWd/te with a target level of 42 Gwd/te.

5. SUMMARY

AECL's MOX fuel fabrication activities are conducted in the RFFL at Chalk River Laboratories. The RFFL facility is designed to produce experimental quantities of CANDU MOX fuel for reactor physics tests or demonstration irradiations. From 1979 to 1987, several MOX fuel fabrication campaigns were run in the RFFL, producing various quantities of fuel with different compositions. In late 1987, the RFFL was placed in a state of active standby, a condition where no fuel fabrication activities are conducted, but the monitoring and ventilation systems in the facility are maintained. Currently, a project to rehabilitate the RFFL and resume MOX fuel fabrication is underway. The initial campaign will consist of the production of thirty-eight 37-element $(U, Pu)O_2$ bundles containing 0.3 wt.% Pu in H.E., destined for physics tests in the zero-power ZED-2 reactor.

This paper has provided an overview of AECL's MOX fuel irradiation program. Irradiation testing and PIE of MOX fuel is continuing at AECL; it has gone from multi-element to multi-bundle demonstration testing. PIE of $(U,Pu)O_2$ bundles irradiated to burnups ranging from 18 to 49 GWd/Mg H.E. in NPD has demonstrated the excellent performance of MOX fuel to high burnup, at power ratings up to 45 kW/m, including power ramp testing. Dimensional changes and fission-gas release were minimal, and there were no indications of defects during the irradiation.

The status of current MOX fuel irradiation tests in NRU has been outlined, including the irradiation of various $(U, Pu)O_2$ and $(Th, Pu)O_2$ bundles to extended burnup.

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