

# Fuel cycle options for light water reactors and heavy water reactors

Proceedings of a Technical Committee meeting held in Victoria, Canada, 28 April – 1 May 1998



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#### FOREWORD

In the second half of the 20th century nuclear power has evolved from the research and development environment to an industry that supplies 16% of the world's electricity. By the end of 1997, over 8500 reactor-years of operating experience had been accumulated. Global environmental change, and the continuing increase in global energy supply required to provide increasing populations with an improving standard of living, make the contribution from nuclear energy even more important for the next century.

For nuclear power to achieve its full potential and make its needed contribution, it must be safe, economical, reliable and sustainable. All of these factors can be enhanced by judicious choice and development of advanced fuel cycle options.

The Technical Committee Meeting (TCM) on Fuel Cycle Options for Light Water Reactors and Heavy Water Reactors was hosted by Atomic Energy of Canada Limited (AECL) on behalf of the Canadian Government and was jointly conducted within the frame of activities of the IAEA International Working Group on Advanced Technologies for Light Water Reactors (IWG-LWR) and the IAEA International Working Group on Advanced Technologies for Heavy Water Reactors (IWG-HWR).

The TCM provided the opportunity to have in-depth discussions on important technical topics which were highlighted in the International Symposium on Nuclear Fuel Cycle and Reactor Strategies: Adjusting to New Realities, Vienna, 3–6 June 1997. The main results and conclusions of the TCM were presented as input for discussion at the first meeting of the IAEA's newly formed International Working Group on Fuel Cycle Options.

The IAEA expresses its appreciation to AECL and the Canadian Government for hosting the meeting.

The IAEA officers responsible for this publication were R.B. Lyon, B.O. Cho, J. Cleveland and V. Onoufriev, of the Department of Nuclear Energy.

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#### SUMMARY

#### Background

The IAEA convened an International Symposium on "Nuclear Fuel Cycle and Reactor Strategies: Adjusting to New Realities" in June of 1997 in Vienna. Important conclusions from Key Issue Paper 3: "Future fuel cycle and reactor strategies" were that: future fuel cycle technologies are expected to evolve from and yet remain closely related to those in use at present; some of these technologies have already been fully commercialised while others will require further development before they can be deployed on a large scale; factors influencing the choice of fuel cycle strategy include:

- economics of recycle versus once-though option
- total inventories of high level waste and spent fuel requiring permanent disposal
- environmental impact of spent fuel and uranium mining tails from each strategy
- importance of high uranium resource utilization for energy supply sustainability
- perceived need to maintain the recycle technology base for future generations
- level of actual and perceived proliferation risk arising from the total inventory of unseparated and separated plutonium in the civilian fuel cycle, and from the fuel cycle technologies themselves, and
- national technical infrastructure and policies

The use of new and advanced fuel designs capable of extended burnup and longer operating cycle times could lead to economic improvements. It will be necessary to overcome those factors that currently limit fuel burnup, including: cladding corrosion, fission gas release, fuel-cladding interactions, and fuel assembly deformation. Development of new materials will be required in some cases. The development of high burnup mixed Pu-U oxide (MOX) fuel can potentially improve fuel cycle economics because, unlike  $UO_2$  fuel, where higher enrichment implies higher cost, the cost of MOX fuel will be largely independent of burnup.

Success in maximizing the utilization of uranium will also have a significant effect on the environmental impact of nuclear power via reduction in uranium mining tails at the front-end of the cycle, and reduction in spent fuel or high level waste at the back-end.

Plutonium recycle in light water reactors (LWRs) is limited to a small number of recycles because of the build up of non-fissile (even numbered) isotopes, and similarly with uranium recycle because of the build-up of  $^{236}$ U (which increases during reenrichment of  $^{235}$ U). Recovered uranium from reprocessing can be enriched and used in LWRs, in MOX fuel with plutonium, or directly in heavy water reactors (HWRs). An alternate technology which avoids separation of U and Pu is the concept of the "Direct Use of Spent PWR Fuel In CANDU" (DUPIC), and variants thereof, in which the greater neutron efficiency of the HWR allows spent LWR fuel to be reused.

Thorium-based fuels are attractive for a number of reasons: the enhancement of fuel resources by producing <sup>233</sup>U, including the near-breeding potential in a thermal reactor; the large thorium deposits in some countries coupled with a shortage of uranium in

those countries; good in-core neutronic and physical behaviour; lower excess reactivity requirements; production of other uranium isotopes that may provide greater technical barriers to proliferation; and reduced production of Pu. Thorium has not been used more widely mainly because the ore contains no fissile isotope, and so <sup>235</sup>U or plutonium must initially be used with thorium to provide the neutrons for its conversion to <sup>233</sup>U, which is fissionable. Thorium-based cycles have been developed through the basic R&D phase in several countries, and have been demonstrated successfully in several power reactors.

#### Purpose, scope and conduct of the meeting

The technical committee meeting provided an opportunity for participants from Member States to explore fuel cycle options in further technical depth, and a forum to identify, review and exchange information on international developments. The fuel cycles under most active consideration, their development status in the context of national programmes, and their key features, issues, development needs, and expected benefits were explored with a view to their potential for enhancing the safety, economics, reliability and sustainability of nuclear power.

The scope included global and national fuel cycle strategies; and development activities for high burnup fuel, MOX fuel, slightly enriched uranium (SEU) fuel, recycled uranium (RU) fuel, thorium fuels, uranium-free fuels, LWR and HWR synergistic fuel cycles such as DUPIC and TANDEM; and the impact of the fuel cycles on uranium utilisation, fuel cycle and power generation costs, plant performance, and waste management.

The TCM was hosted by Atomic Energy of Canada Limited on behalf of the Canadian Government and was jointly conducted within the frame of activities of the IAEA International Working Group on Advanced Technologies for Light Water Reactors (IWG-LWR) and the IAEA International Working Group on Advanced Technologies for Heavy Water Reactors (IWG-HWR), and as a cooperative task between the IAEA Divisions of Nuclear Power, and Nuclear Fuel Cycle and Waste Technology.

The TCM was attended by 46 people from 12 countries, the OECD/Nuclear Energy Agency and the IAEA. 35 papers were presented. The meeting Chairman was P.G. Boczar of AECL, Canada.

#### Global and national perspectives for developing advanced fuel cycle options

The fuel cycle experience from around the world included concrete examples of successes to date. Countries are at various stages in their economic growth and their nuclear programmes. They have differing needs with respect to energy security, have differing nuclear technologies and technological capabilities, and have either Thorium or Uranium, or both in various proportions. Consequently no single fuel cycle strategy can satisfy all Member States' needs.

India has both boiling water reactors (BWRs) and HWRs. A closed fuel cycle option with recycle of uranium, plutonium and thorium is favoured. While in the long term, fast breeder reactors (FBRs) operating on the thorium cycle are planned for energy self-reliance, an

advanced HWR (AHWR) has been designed that would use the  $^{233}$ U-Th mixed oxide cycle in a near self-sustaining mode (as much  $^{233}$ U would be discharged in the spent  $^{233}$ U-Th fuel as would be required in the fresh fuel). A small number of PuO<sub>2</sub> fuel elements in the bundle would act as "driver" fuel, and provide a higher burnup capability than would otherwise be possible in a strictly self-sustaining mode.

Recycle of spent fuel is being conducted in Germany. Twelve German reactors are already licensed for the use of MOX fuel, five others have applied for MOX use. Eight reactors are currently using MOX fuel or have used it in the past. Currently most of the spent fuel elements are shipped to France and the United Kingdom for reprocessing. Some utilities now consider direct disposal of their spent fuel for economic reasons.

In the USA, the once-through LEU cycle is employed. The extended burnup programme for LWR fuel aims at doubling the current burnup in 15 years, to reduce costs, extend uranium utilization, and reduce the frequency of scheduled outages and the volume of spent nuclear fuel. The focus of the program is on developing and qualifying improved cladding materials.

In Canada, the low cost of natural uranium fuel means that any new fuel option will have to offer compelling advantages. One step in this direction is the demonstration irradiation in a power reactor of the CANFLEX advanced fuel bundle which will facilitate the implementation of advanced fuel options. The technical feasibility of fabricating MOX fuel from ex-weapons plutonium is being demonstrated in the "Parallex" program, in which CANDU MOX elements, fabricated in the U.S. and in the Russian Federation, will be irradiated in Canada. For HWRs, the high neutron efficiency, channel design, on-line refuelling, and simple fuel bundle design provide flexibility to optimise fuel cycles.

Romania has developed its fuel manufacturing capability to the stage where it has now been fully qualified, and the Cernavoda 1 core comprises more than 99% Romanian-fabricated fuel; no defect has been found in 1.5 years.

In Argentina, doubling of the discharge burnup has been achieved in the Atucha-1, pressure-vessel HWR, by replacing natural uranium with SEU, with no other changes to the core, with good operational flexibility and significant fuel cycle cost savings. Argentina has also developed a fuel design, CARA, to be used in both Atucha and Embalse reactors. A design criterion is that the CARA cost does not exceed the cost of the CANDU fuel bundle, which is half the cost for the complex Atucha fuel assemblies.

#### Fuel design, performance and testing

Fuel is being designed and tested to extend burnup, reduce defect rates, improve thermalhydraulics and heat transfer, and improve performance under accident conditions. Approaches to develop improved fuels include development of new cladding material in the Russian Federation, which allows higher burnup with reduced corrosion, development of fuel in Romania with larger grain structure that reduces fission gas release and hence pressure inside the cladding, and CANFLEX fuel development in Canada for improved thermal hydraulic performance with higher dryout powers and lower peak element ratings. A technique for measuring fissionproduct diffusion coefficients in  $UO_2$  fuel using heavy-ion implantation is being developed in Canada. There are several facilities world-wide for testing high burnup fuel under normal operating and simulated accident conditions.

#### Slightly enriched uranium (SEU) / recycled uranium (RU) / Thoria fuels

SEU/RU/Thoria fuels can be used in HWR reactors to flatten flux distributions (thereby increasing unit power), increase burnups and extend fuel resources. Recycled uranium (RU) from spent LWR fuel has about 0.9 percent <sup>235</sup>U, an enrichment ideally suited as fuel for HWRs. Use of the RU accumulating at the commercial reprocessors will depend largely on the economics of RU compared to SEU and natural uranium. Fuel management calculations have demonstrated that HWRs can use SEU/RU fuel with operation within current design envelopes, using simple fuel management schemes. Similarly, evaluations indicated that the use of a CANFLEX bundle as the carrier for RU would be compatible with current reactor designs, and operational and safety requirements.

While extensive use of thorium lies in the future, there are some current developments. A once-through thorium cycle has been evaluated in Canada, taking advantage of the flexibility of the CANFLEX bundle design, using ThO<sub>2</sub> and SEU elements. A novel Indian HWR design employs a very large fuel channel (calandria tube OD 20.7 cm), with a fuel bundle containing driver or "seed" fuel elements and pre-irradiated ThO<sub>2</sub> elements. The coolant is boiling light water, and the void reactivity coefficient is negative. Breeding of fissile material is possible, depending on the choice of seed material.

#### MOX fuel

The most significant current development in fuel cycles is the use of MOX fuel in LWR reactors, mostly in Europe. A number of countries have successfully demonstrated the use of mixed oxide  $Pu/UO_2$  (MOX) fuel in power reactors. Up to 30% MOX loading in LWRs can be considered an established technology. Higher MOX loading in LWRs in most cases will require control rod design changes to maintain current operating and safety margins. Assessments in Canada show that there is no barrier to the use of 100% MOX loading in an HWR.

The capability to achieve burnups in excess of 60 MWD/KgU will depend on development and proof testing of improved fuel elements with higher enrichment. Acceptable accident behaviour for high burnup fuel, whether  $UO_2$  or MOX, has to be demonstrated. A study comparing the economics of MOX with  $UO_2$  fuels in the USA indicated that MOX fuel cost is currently 4 to 10 times higher than uranium fuel cost; however, increased MOX production, with longer fabrication campaigns (higher facility utilisation) could bring MOX fabrication costs down.

Japanese studies have evaluated increasing the moderator-to-fuel ratio in advanced LWR MOX core designs to increase the fissile plutonium consumption rates. Critical experiments for high moderation MOX lattices have been performed. The safety implications of MOX for weapons grade Pu-disposition have been analysed in the Russian Federation. Spent MOX fuel generates more heat than  $UO_2$  which is a challenge for spent fuel storage. It is necessary to derate waste loading for dry storage. Repacking in storage pools may be appropriate but the racks may need extra neutron-poison to hold down reactivity. MOX (Pu/UO<sub>2</sub>) fuel fabricated and tested in Canada is capable of sustaining normal CANDU power densities, with burnups at least three times that of natural uranium, and with performance generally comparable to that of  $UO_2$ .

#### Inert matrix (uranium-free) fuel for actinide burning and plutonium annihilation

Consideration is being given to incineration of separated plutonium, from ex-weapon stockpiles and from reprocessed spent LWR fuel from the commercial fuel cycle, by burning in a non-fissile carrier matrix. The same concept can be applied to actinide burning as a waste management strategy. Rock-like fuels (a two phase mixture of fluorite and spinel) for carrying plutonium or actinide waste are being studied in Japan, CERMET (fuel dispersed in a metallic matrix) fuels are being studied in France and SiC is being investigated as an inert carrier matrix in Canada.

To avoid the economic penalty of a second processing and refabrication cycle a "once through" cycle is favoured with high burnups and almost complete annihilation of Pu. The fuel matrix should be as insoluble as possible to provide a technical barrier to proliferation. Studies of inert matrix fuels indicate that addition of a burnable poison (such as erbium) is necessary for reactivity control. The addition of thorium or uranium can also provide resonances for Doppler broadening, to enhance the negative temperature coefficient.

Irradiation experience is required to prove the performance of inert matrix fuels. All projects discussed have taken the first step - irradiation with ions from accelerators to simulate fission product damage, and the results to date have been excellent for silicon carbide and zirconia. Only a few countries have initiated in-reactor irradiations of inert matrix materials.

#### LWR/HWR synergistic fuel cycles

The DUPIC fuel cycle, currently under development, is a dry process for reconstituting LWR pellets into HWR pellets, without the selective removal of any isotopes. Plutonium is not separated from the spent fuel, thus providing a technical barrier to proliferation. It also offers a number of other benefits relative to the once-through cycle, including saving of uranium and a reduction in the amount of spent fuel requiring disposal. For a given amount of electricity generated, DUPIC spent fuel disposal costs are expected to be significantly lower than for either PWR or HWR spent fuel. In Canada, three DUPIC elements have been successfully fabricated for irradiation testing, and the pellets meet CANDU requirements for density and surface finish. An 'AIROX' LWR-to-LWR dry recycle process proposed in the USA is similar in concept to DUPIC.

#### Strategies and development requirements for the future

It was suggested that the nuclear industry should have as a goal, a long-term reduction in the cost of nuclear-supplied electricity of about 50% to be competitive with fossil fuels. The need for a holistic approach to optimising the entire fuel cycle, and not just one aspect, was stressed. For example, fuel could be designed to facilitate its recycle from one reactor to another. Fuel supply, fabrication, electricity generation, fuel storage, recycle, waste management and disposal should be considered in total, to simplify the fuel cycle while reducing total generation costs, increasing resource utilisation, increasing safety, reducing environmental impact, and satisfying key political objectives. Adoption of an integrated approach would also ensure that research and development is concentrated on the most important objectives.

## GLOBAL AND NATIONAL PERSPECTIVES FOR DEVELOPING ADVANCED FUEL CYCLE OPTIONS

(Session 1)

## Chairperson

**R.J. Page** United Kingdom



#### SUMMARY OF "INTERNATIONAL SYMPOSIUM ON NUCLEAR FUEL CYCLE AND REACTOR STRATEGY: ADJUSTING TO NEW REALITIES" VIENNA, JUNE 1997

N. Oi Nuclear Fuel Cycle and Waste Technology Division, International Atomic Energy Agency, Vienna



#### Abstract

The International Symposium on Nuclear Fuel Cycle and Reactor Strategy: Adjusting to New Realities was held from 3 to 6 June 1997 in Vienna, Austria. The objective of the Symposium was to prepare for decision makers and the public, a scientific assessment of the different fuel cycle and reactor strategies with particular reference to the production, use and disposition of plutonium. Six key issue papers were prepared by six groups of international experts which summarized the international common understanding of the various fuel cycle issues including those related to technology, safety, safeguards, environmental and institutional developments. This paper summarizes the major finding of the Working Groups except for Working Group 3 which will be presented in depth in a separate paper in this Technical Committee Meeting.

#### 1. Introduction

The International Symposium on Nuclear Fuel Cycle and Reactor Strategy: Adjusting to New Realities was held from 3 to 6 June 1997 in Vienna, Austria. It was organized by the International Atomic Energy Agency (IAEA) in co-operation with the European Commission, the Nuclear Energy Agency of the OECD (NEA/OECD) and Uranium Institute (UI). More than 300 participants from 44 countries and five International organizations took part.

A Steering Group of senior experts from twelve IAEA Member States and two international organizations directed the Symposium's organization over 2.5 years. Additionally, six Working Groups prepared six key issue papers with the participation of over 70 experts from 12 States — Argentine, Canada, China, France, Germany, India, Japan, Russia, South Africa, Sweden, the United Kingdom and the United States — and NEA/OECD, the European Commission, the International Energy Agency, and the UI. These papers represented the common international understanding on various aspects of nuclear fuel cycle and reactor strategy, with particular reference to the issue of plutonium, up to the year 2050, and were the result of two years of intensive work by the experts.

A Summary of the Symposium is presented here because it is thought that the outcome of the Symposium is useful in setting the stage for this Technical Committee Meeting on "Fuel Cycle Options for LWRs and HWRs".

#### 2. New realities

In about two decades since the International Fuel Cycle Evaluation (INFCE) was carried out, several new realities have emerged. The new realities are: an unexpected slow growth of nuclear energy, the escalation of back-end costs, the delay in the introduction of fast reactors and the end of the cold war. The consequence of these changes are: a surplus of uranium, the continued debate over the choice of fuel cycle (once through or closed), a surplus of separated plutonium, and plutonium in spent fuel and finally, the demilitarization of weapons plutonium and highly enriched uranium. The aim of the Symposium was to face these new realities and to come to conclusions how these new realities should be addressed.

#### 3. Global energy outlook (Key Issue Paper No. 1)

The important task of this Working Group was to examine the relationship between the expected size of the nuclear power generation and necessary fuel cycle volume up to the time frame of 2050.

- At the end of 1997, 433 nuclear power plants were operating with the total electricity output of 348 GWe. The key issue paper considered three nuclear energy scenarios (Fig. 1) with he following results in 2050: high variant 1805 GWe, medium variant 1132 GWe, and low variant 333 GWe. These scenarios were considered, based on studies by the International Institute for Applied Systems Analysis (IIASA) and the World Energy Council (WEC), and they were characterized as "contrasting but not extreme". Projected cumulative natural uranium requirements for the years 1995-2000 were calculated and compared with published levels of resources in the publication Uranium Resources, Production and Demand, commonly known as the "Red Book" (Fig. 2). The supply of uranium for nuclear power reactors will probably be sufficient to satisfy worldwide programme needs up to the year 2050. In the case of the medium variant, there may not be sufficient uranium resources to cover the years after 2050 for reactors existing then if one assumes that these reactors will have a total lifetime of up to 40, perhaps up to 60 years. Therefore, the ways and means to make better use of uranium resources, and the potential influence of steps taken, are of importance.

- A number of technical measures offer promising options for the better utilization of uranium resources. They include increasing the burnup of nuclear fuel during reactor operations, the lowering of tails assays in the depleted stream of fuel enrichment operations, and recycling plutonium. About 25% of all uranium resources can be saved by reducing the tails assay from 0.3% to 0.15%, compared with a saving of about 17% by recycling all plutonium in light-water reactors. Both options are achievable from a technical and industrial viewpoint.

- Over the long term from a worldwide perspective, strategies and technologies targeted at more efficient uses of uranium resources will probably have to be considered before 2050.

#### 4. Plutonium Management (Key Issue Paper No. 2)

- Since INFCE some 20 years ago, not very much seems to have changed as far as policy is concerned. Most countries that decided to pursue reprocessing/recyclingprogrammes have not changed their positions since then. A large and viable recycling industry has been established in Europe and is being developed in Japan.

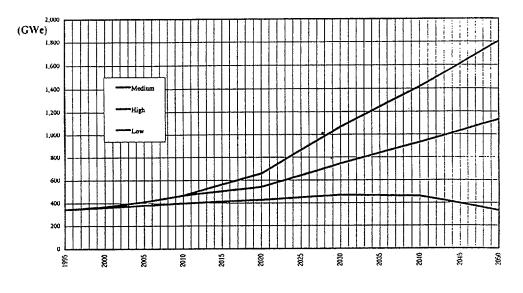


Fig. 1: World Nuclear Capacity Scenarios (IIASA/WEC)

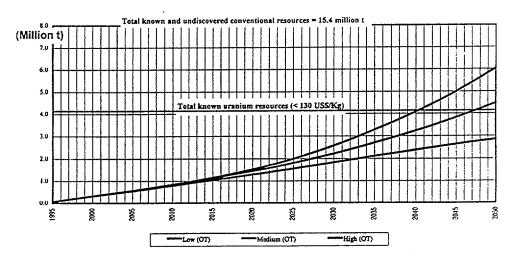


Fig. 2:

Projected Cumulative Natural Uranium Requirements up to 2050

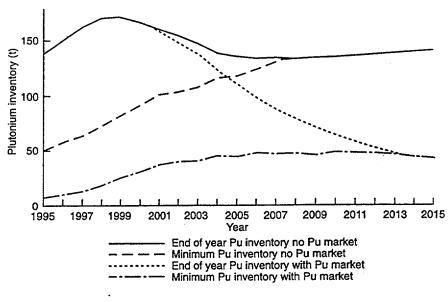


Fig. 3: Worldwide Separated Civil Plutonium Inventories

#### Table 1: Radiological Occupational Collective Doses of the Three Fuel Cycles (per 400 TW.h) excluding Radioactive Disposal

	Occupational exposure	Main contributors		
OTFC	153 man.Sv	Reactors 69%; mining/milling 29%		
мох	147 man.Sv	Reactors 72%; mining/milling 26%		
MOX-FR	139 man.Sv	Reactors 76%; mining/milling 22%		

- At the end of 1996, the inventory of separated civil plutonium amounted to about 150 tonnes, and it is expected to increase to about 170 tonnes by the end of 1999 before dropping to about 150 tonnes by the year 2015 (Fig. 3). Under free market conditions for plutonium, the inventory could be reduced to about 50 tonnes by 2013. This does not include the amounts of plutonium that Russia and the United States have in excess of their defense needs and may release into the civilian sector.

- The inventories of separated plutonium are expected to be reduced by the use of modern fuel fabrication plants for producing mixed-oxide fuel (MOX) and the licensing of light-water reactors to burn MOX fuel.

- Medium- and long-term spent fuel storage can be carried out at both "at-reactor" sites and "away-from-reactor" sites.

- Key technologies are available for the effective management of both the closed and open nuclear fuel cycles, and for the disposition of surplus military plutonium. Many of these technologies have been implemented.

# 5. Safety, Health & Environmental Implications of Different Fuel Cycles (Key Issue Paper No. 4)

- Key conclusions of this working Group which examined the health and environmental implications of the different fuel cycle options include:

- In normal operation, there are no significant differences in terms of human health and environmental safety impacts among the nuclear fuel cycle options considered. (Table 1)

- A remaining issue common to all three fuel cycles is the potential for major accidents which may have significant health and environmental consequences. The prevention of such accidents calls for a high level of vigilance and an ongoing improvement of safety.

- Long-term storage and disposal of spent fuel or radioactive waste do not raise any particular problems in terms of health. Individual exposure remains at extremely low levels as long as no intrusions into the disposal sites occur.

- Plutonium toxicity is not a major factor in the context of normal operational impacts. However, there is much misconception about this issue which has been often used as a strong argument against the fuel cycle, including reprocessing of nuclear fuel.

#### 6. Non-Proliferation and Safeguards Aspects (Key Issue Paper No. 5)

This working group considered non-proliferation and safeguards aspects related to the nuclear fuel cycle. Its key conclusions included:

- The nuclear non-proliferation regime is becoming increasingly effective. Additional demands placed upon the regime must be adequately funded by the international community.

- The nuclear non-proliferation regime needs continuous adaptation to "new realities" affecting nuclear power development. Two good examples are the IAEA's safeguards development programme through which the verification system was strengthened, and initiatives for the verification of surplus military materials transferred into the civilian sector.

- A main issue facing the nuclear non-proliferation regime over the next decades is the extent to which the IAEA will be involved in the verification of surplus military material and how this, and other demands on the safeguards systems, will be resourced. New technical and institutional approaches will be required.

- In the context of reactor and fuel-cycle choices and future technological development in the civil nuclear power sector, the nuclear non-proliferation regime should be able to provide the necessary assurances, irrespective of the nuclear technology chosen, and should not constrain future choices.

#### 7. International Cooperation (Key Issue Paper No. 6)

The sixth working group considered aspects of international cooperation. Its key conclusions included: - International co-operation has been an essential factor and a principal driving force in the development and application of nuclear power. The most distinctive feature of this co-operation the nuclear non-proliferation regime — has successfully limited the spread of nuclear weapons to a level far below those once predicted. - The supply of nuclear materials, equipment, and technology for peaceful uses by States possessing them to other States has been one of the major and most impressive successes of international co-operation.

- The arrangements and mechanisms in place for international co-operation are generally adequate to meet current and future needs. However, improvements are desirable in a number of areas, such as the disposition of surplus military plutonium, development of fast-breeder reactors, regional fuel cycle centres, international plutonium storage, and the transparency of plutonium management.

- The IAEA should explore appropriate steps to ensure the exchange of basic information on major developments, and economic and programmatic information on the fuel cycle, possibly through establishing a regular mechanism of such exchange in close cooperation with other international organizations.

#### 8. Concluding Remark

In summary, the symposium served as a valuable forum for examining the new realities and choices facing countries utilizing nuclear energy. The six key issue papers presented at the symposium summarized the common international understanding of the various fuel cycle issues, including those related to technology, safety, safeguards, environmental and institutional developments.

The symposium also served to heighten interest in continuing the dialogue at the global level, in light of the importance of issues being faced and nuclear power's established and potential role in contributing to world electricity supplies. Toward this end, the IAEA in early 1998 set up the International Working Group on Nuclear Fuel Cycle Options. Among topics that the Group will cover are the advantages and disadvantages of different fuel-cycle strategies of plutonium and waste management, which will play a key role in the future development of nuclear energy.

In the final analysis, the ongoing evolution of Agency programmes related to the nuclear fuel cycle must reflect the realities confronting the international community today, including the security and commercial impacts of ex-weapons material. Moreover, the activities will have to be geared to promoting further the reliability, safety, and economic viability of nuclear power to help interested countries meet electricity demands well into the next century.

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#### FUTURE FUEL CYCLE AND REACTOR STRATEGIES\*

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#### Abstract

Within the framework of the 1997 IAEA Symposium "Future Fuel Cycle and Reactor Strategies – Adjusting to New Realities", Working Group #3 produced a Key Issues paper addressing the title of the symposium. The scope of the Key Issues paper included those factors that are expected to remain or become important in the time period from 2015 to 2050, considering all facets of nuclear energy utilization from ore extraction to final disposal of waste products. The paper addressed the factors influencing the choice of reactor and fuel cycle. It then addressed the quantitatively largest category of reactor types expected to be important during the period; that is, thermal reactors burning uranium and plutonium fuel. The fast reactor then was discussed both as a stand-alone technology and as might be used in combination with thermal reactors. Thorium fuel use was discussed briefly. The present paper includes of a digest of the Key Issues Paper. Some comparisons are made between the directions suggested in that paper and those indicated by the Abstracts of this Technical Committee Meeting. Recommendations are made for work which might be undertaken in the short and medium time frames, to ensure that fuel cycle technologies and processes established by the year 2050 will support the continuation of nuclear energy applications in the long term.

#### 1. INTRODUCTION

Nuclear power plants and their supporting fuel cycle facilities represent a mature technology that now provides a significant proportion of the world's electricity. Studies of world energy supply indicate that there may be an increasing role for nuclear power in the 21st century, both as replacement for aging capacity in the developed nations and as new capacity to support economic growth in developing nations. The UN International Panel on Climate Change recognizes that nuclear energy could play a role in mitigating emissions of carbon dioxide. Uses of electricity may evolve during the period to include, for example, widespread use as motive power for transportation in addition to its traditional role. It also is probable that advanced technologies will increase the use of nuclear energy in non-electricity applications (e.g. district heating, desalination, process heat).

#### 2. DIGEST OF KEY ISSUES PAPER #3

This paper addressed the middle time scale of 2015 and 2050, and included the whole nuclear fuel cycle from mine to disposal. Based on a median estimate of the number of new nuclear plants which might be required world-wide up to this time, the paper considered factors which might influence the technological directions taken and then framed conclusions about the 'state of the art' within this middle time frame. A paper of this type is, of course, highly speculative. However, it is expected that the actual future development of nuclear energy will depend on three basic issues: government and public approval, economic competitiveness, and on the role which nuclear energy might be called upon to play in sustaining a healthy natural environment in the world.

<sup>\*</sup> Work performed in preparation for the IAEA International Symposium "Future Reactors and Fuel Cycles: Adjusting to New Realities" Vienna, June 1997

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The role assigned to nuclear energy will depend strongly upon its ability to compete with other energy sources (e.g. fossil fuels). In a competitive global energy market, the success of any energy source, be it nuclear or non-nuclear, will be dependent upon its ability to:

- properly utilize natural resources and national capabilities,
- maximize the economic benefits,
- minimize the overall environmental impact,
- effectively demonstrate the safety of fuel cycle facilities,
- gain government and public approval of the enterprise,
- satisfy national and international policies and goals,
- contribute to sustainable energy supply.

Decisions on the balance of new generating capacity will be influenced by the weight assigned by individual utilities and national decision-making bodies to each of these factors. For example, nations with limited natural resources are more likely to have aspirations toward energy self-sufficiency and may therefore opt for developing a home-grown nuclear program. Alternatively, energy selfsufficiency may be seen to be of lower importance in a global energy market. Economic benefits also may be profoundly affected by associated government policies such as taxation.

Evolution and increase in nuclear generating capacity suggests that the current generation of nuclear power plants and their supporting fuel cycles are economically competitive. On the other hand, the present lack of new orders in many developed nations indicates that its advantages have been eroded and are now marginal. This situation may or may not continue in the future, depending on the response of the industry.. The objective is to identify those key factors that will influence the development of nuclear reactors and fuel cycle concepts during the first half of the 21st century. Although these factors are considered here in isolation from other (non-nuclear) energy sources, performance of the non-nuclear energy options is likely to have a major influence on the size and scope of future nuclear programs. For example, as the environmental effects of fossil fuel use become better quantified and apparent, expansion of their use of fossil fuels may become impractical. But new technologies might alleviate these negative effects. Stellar performance of existing nuclear plants may be seen as essential to overcoming negative public attitudes so that future expansion of nuclear capacity can be practical. But a long period of good performance might eliminate the strong anxieties which exist today in many countries.

#### 2.1. Maximize the utilization of natural resources

Today's commodity price of uranium is low, but will rise if demand increases and cheap resources are depleted. Consequently, in addition to consideration of natural resource utilization and environmental impact, there will be economic pressure to increase the energy yield from mined uranium. This will provide motivation for:

- specifying lower tails assay in enrichment plants
- introducing higher burnup once-through cycles
- considering interim storage as a precursor to recycle instead of disposal
- recycling increased amounts of uranium and plutonium in thermal reactors
- recycling uranium and plutonium in fast reactors or fast-thermal tandem systems
- introducing concepts with higher thermal efficiency

Uranium is likely to remain the main natural resource supporting the development of nuclear power programs, given the large world-wide investment in the uranium cycle. In the longer term, however, the more widespread use of thorium may occur in some countries.

#### 2.2. Maximize economic benefits

A major influence on future development will continue to be the drive to reduce development, construction, operating, maintenance, decommissioning, and financing costs. The high cost of developing and licensing new 'first-of-kind' power plant and fuel cycle designs will continue the present trend toward increasing international collaboration and standardization. This, and the cost and uncertainty of licensing more advanced concepts, are likely to give rise to facilities based on the evolutionary development of current technology rather than to innovative new technologies. In addition, as the established parts of the nuclear fuel cycle are recognized as mature technologies, the source of funding will move from government to private sector with resulting emphasis on short-term economic planning. The requirement to reduce capital costs implies that future systems will move further toward:

- standardization of facilities and components to minimize unit costs,
- multiple-unit plants to make best use of infrastructure investment and available sites,
- choice of unit size to balance economies of scale and those of repetition,
- elimination of over-design as knowledge of true margins is refined.

In order to improve the return on capital investment and to control unit operating costs, future systems will be required to achieve high capacity factors. This will, for batch-refueled reactors, tend toward the development of long cycle times, short shut-downs, and rapid return to power. On-line refueling will be employed where appropriate with a trend toward higher fuel burnup and use of tandem cycles. The cost objective will drive the continued evolution of organizations providing fuel cycle services. Efficiency may be improved by a combination of:

- enhancement and upgrading of existing facilities to extend their capability and life,
- use of high quality components and systems in new facilities,
- rationalization of vendor base to fully realize economies of scale and new technologies,
- evolution of fuel cycle centers in technically advanced nations.

The high capital cost and long construction time of nuclear facilities coupled with uncertainties in licensing, decommissioning and waste management costs have caused difficulties in raising funds for new facilities. These considerations may lead the industry toward, for example:

- encouraging incremental investment through modular systems design, where possible,
- reducing construction and commissioning times,
- development of regional and global licensing standards and practices,
- properly quantifying and managing decommissioning and waste management liabilities.

#### 2.3. Minimize the environmental impact of fuel cycle activities

It is understood by the public that the industry must manage and reduce environmental impacts associated with fuel cycle facilities. Increasingly, this awareness will lead to the development of the next generation of nuclear fuel cycle facilities which:

- prevent waste generation, concentrating on waste control at the source,
- reduce waste streams by including high control standards in original systems design,
- intensify processes to reduce the volume of associated waste,
- use novel waste forms and new immobilization matrices and technologies,
- economically-optimum underground repositories for wastes requiring secure final disposal.

Recognizing that uranium mining is an important contributor to total radioactive materials released to the environment from existing fuel cycles, reducing the environmental impact of mining and

increasing the utilization of uranium by higher burnup and recycle can significantly reduce the overall environmental impact of nuclear power.

Programs are in place to ensure that technical issues associated with disposal of high level waste will be fully demonstrated so that facilities can be operational in the 21st century. Achievement of these goals and gaining public approval for installation of disposal facilities are tasks vital to successful continuation of the nuclear enterprise. Furthermore, if it can be shown to be beneficial to reduce the waste source term by transmutation of long lived isotopes, development of actinide-burning reactors and associated chemical partitioning plants, or appropriate accelerator technology may be introduced. The drive for reduction of long-lived isotopes also may lead to wider use of the thorium fuel cycle with its more favorable spectrum of activation products and fission products.

#### 2.4. Effectively demonstrate safety of fuel cycle facilities

Although nuclear power has an excellent safety record, the Chemobyl accident demonstrated the potential for severe and widespread (national and regional) radioactive materials contamination. As a consequence it is perceived by many people that nuclear power is unsafe. This public concern, coupled with the cost and project risk associated with the licensing of any new nuclear facility, indicates that safety issues will continue to exert a major influence on the future of nuclear power well into the 21st century. In order to reduce the magnitude of potential hazards new facilities will be designed, where practical, such that:

- processes and plants are intrinsically safe, achieved by designing out hazards rather than by adding active protective devices,
- off-site damage from potential accidents will be deemed incredible (e.g. by the use of highefficiency barriers, by enhancing core integrity, or by reducing the potential for reactor systems to damage containment systems following a severe accident).

In order to gain maximum benefit from international experience and to control the associated with safety, it is likely that trends will be towards:

- development and application of internationally accepted safety and engineering principles,
- collaboration to provide a standard range of internationally accepted designs,
- acceptance that licensing of a facility in one country provide can provide useful input to licensing processes for similar facilities in other nations,
- transfer of knowledge and experience through operator-led organizations such as WANO.

Even with these potential improvements, safety costs and potential project delays will continue to have a major influence on the implementation of nuclear programs unless and until it is perceived by both safety professionals and the general public that nuclear power is safe relative to the alternatives. Clearly, this perception will be dependent upon the safety performance of existing plants and the industry's ability to prevent any major accident during the next decades.

#### 2.5. Gain government and public approval

Public and government approval will be the dominant factor influencing the scale of nuclear power use throughout the world. An important activity for all those associated with nuclear power is to communicate clearly the true costs, benefits, and risks of nuclear power, in an open and credible manner, to the public in general and to their opinion leaders in particular.

Approval of any activity by a national government constitutes recognition that the activity is acceptably safe. This "licensing" process includes technical evaluation to assess the scope of actual risk. In addition, issuance of a license to operate includes an implicit evaluation that the activity will

benefit the nation. This evaluation is a political one and will depend in some manner on the wishes of the people. It is essential that the public and their leaders possess accurate knowledge of the benefits and risks of the activity.

#### 2.6. Satisfy national and international policies and goals

National and international policies and goals cover a range of attributes that may be considered beneficial by national governments or individual utilities, and against which the performance of different nuclear plant options will be measured. These factors include:

- non-proliferation performance and safeguards,
- security of energy supply,
- energy independence, national and regional,
- balanced energy policy,
- access to technologies and potential for capacity expansion,
- use of existing fuel stockpiles (including surplus military material) and enrichment tailings.

#### 2.7. Sustainable energy supply - an integration of factors

The various attributes and factors influencing future reactor and fuel cycles can be examined individually, they always interact. Decisions will be made based on the balance of potentially competing factors. It is apparent that the individual components of the nuclear fuel cycle and factors that influence their development should be considered holistically to arrive at a proper balance.

For the future it will be necessary to optimize fuel cycles as a whole, looking at the way in which the various components of the interact with one another. This should allow for the integration of various options for fuel supply, fabrication, generation, fuel storage, reprocessing, recycle, waste management, disposal and decommissioning in order to simplify the fuel cycle while reducing total generation costs, increasing resource utilization, increasing safety, reducing environmental impact, and satisfying key political objectives. Adoption of an integrated approach also is necessary to ensure that research and development is concentrated on the most important objectives.

#### 2.8. Conclusions of Key Issues Paper #3

- 2.8.1. Actual programs for continued development of nuclear energy use in the period from 2015-2050 are unknown and unknowable today. This paper is, therefore, speculative.
- 2.8.2. Uranium supply will be sufficient to satisfy world reactor program needs, up to 2050.
- 2.8.3 Slow evolution of present reactor types and designs will dominate in the commercial power plant market. Important reactor types in this period will be LWR and HWR.
- 2.8.4 New reactor and fuel cycle development programs will come to maturity. Development of the LMFR, HTR and other types will continue in some countries.
- 2.8.5. Out-reactor fuel cycle technology will be refined. Further developments likely will be capacity expansion to meet the needs of thermal-reactor recycle programs, reduction of cost, and reduction of environmental impact.
- 2.8.6. International cooperation in the maintenance of safety, development of international standards, and improvement of human and equipment safety will continue.
- 2.8.7. Government and public approval will determine the course of nuclear energy development.

#### 3. RELATIONSHIP BETWEEN KEY ISSUES PAPER #3 AND PAPERS GIVEN IN THIS TCM

Key Issues Paper #3 necessarily addressed only very broad subjects. In addition, its time frame began in 2015. For both of these reasons, it is necessary to place more detailed subjects covered in this TCM into context before examining what work might be appropriate in the near-term future.

Short-term commercial developments will be undertaken mainly by private industry, based on need and the desires of the customers — the electricity generation utilities. These developments will take only limited account of the larger national and international issues such as fuel supply, strategic position, and safeguards.

Longer-term developments will rely mainly on national governments which perceive a need for extended work to position their countries for the long term energy supply situation. These governments will take on the larger risks, and those successful in development will reap the benefits late in the next century.

#### 3.1. Plutonium Dispositioning

Several of the papers in this meeting deal with the incineration of separated plutonium, from ex-weapon stockpiles and from the commercial fuel cycle, by burning in a non-fissile carrier matrix. This emphasis grows naturally from the international drive to reduce world stockpiles of separated plutonium. Reactor types considered for this fuel type are the LWR and the HWR.

This development direction has been questioned on the grounds that (a) burial of plutonium is a doubtful proposition from the points of view of both conservation and security, (b) the alternative of using plutonium for electricity production is attractive in both the short-term and long-term, and (c) highly secure storage of separated plutonium is essential in any cas to retain existing and future stocks of this material. It is recognized, however, that this option may satisfy short-term objectives.

But what of the longer term? If we look to the middle of the 21st century the authors of Key Issues Paper #3 see a strong likelihood that fast breeder reactors will be required to limit the price of natural uranium, as high-grade stockpiles are diminished. When uranium prices rise significantly, plutonium will quickly become a scarce commodity because of the very high fissile inventory requirements of FBR for the first loading. Without an ample supply of plutonium, the rate of growth of FBR installed capacity will be strictly limited. It would be prudent to add this long-term consideration into the balance when evaluating methods for disposing of plutonium stocks – whether to bury, burn, or to save for a later day. The option of storing spent fuel without reprocessing also must be considered.

#### 3.2. Evolutionary changes driven by economics

In the short term, the dominant driver for change is economics. Papers in this TCM consider new fuel elements, such as CARA and CANFLEX, which are aimed largely toward cost reduction. Use of recycled uranium in CANDU reactors also falls into this category, though it also carries some opportunities for design improvement in the longer term.

#### 3.3. Steady increase of MOX recycle in LWR

The largest change occurring today is the introduction of mixed-oxide fuel into LWR plants, mostly in Europe. Several papers in this TCM consider various aspects of the use of MOX in LWR and HWR. This work shows excellent prospects from a performance point of view. Up to 30% MOX reloading in PWR can be considered an established technology. There is no barrier to use of 100% MOX loading in HWR. Higher percentage MOX loading in LWR requires additional work on technical

issues such as fission gas release at high burnup and tolerance of high-burnup fuel under accident conditions. In addition, the capability for multiple recycle in LWR is not assured; currently it is estimated that only two or three cycles will be feasible. A large amount of plutonium will remain in the fuel after these cycles. Obviously, this option is not efficient for incineration of plutonium.

There is some disagreement over the economics of the MOX recycle fuel option, mainly between nations which oppose recycle (and find it expensive) and those which favor recycle (and find it cheap). Reality will, in time, settle this argument.

#### 3.4. Increasing fuel enrichment

Fuels loading capable of higher burnup, using either uranium or MOX fuel, will depend on development and proof testing of improved fuel elements. It also will, of course, require higher fresh fuel enrichment. It is expected that the current trend toward higher enrichment will continue for LWR, reflecting the economics of batch-loaded units. Higher enrichment might be obtained through reduced tails assay as well, thereby increasing the energy yield per ton of mined uranium.

The motivation for increasing enrichment in on-power-fuelled HWR is somewhat different. Natural uranium fuel leads to an inherent reactivity limit on fuel life. Life can be increased greatly by a small amount of enrichment, thus leading to freer design options as well as to greater flexibility in fuel management. These both can be translated into better economics through higher power output.

#### 3.5. Recycled uranium use in HWR

The U235 content of uranium remaining after plutonium extraction is around 0.9 percent, which makes this material adequate as fresh fuel for CANDU. There are relatively large stores of this material at each reprocessing plant. The main economic comparison is between the cost of processing this material into fuel versus the cost of purchasing and preparing fresh slightly-enriched uranium.

Economic advantages of using slightly-enriched uranium in CANDU arise from the fact that natural uranium is slightly less than optimum for use in modern CANDU units. With 0.9% uranium it is possible to increase unit output by over 10% by radial power flattening, while increasing average discharge burnup by more than 30%.

#### 3.6. Double-burning in HWR

The term "double-burning" refers to a sequential once-through cycle in two different reactor types, with the second being capable of using discharged fuel from the first without re-enrichment. The HWR is an obvious candidate for this cycle because it can use very low-enrichment fuel.

The degree of fuel rework proposed between burn steps is varied, ranging from direct reactorto-reactor transfer to fuel reprocessing. The DUPIC cycle, currently under development, includes a dry reprocessing cycle to reform LWR pellets into HWR pellets, at the same time removing some fission products and offering a substantial degree of proliferation resistance. One paper in this TCM recommends an international program for demonstration of this technology in one of the countries participating in this project (currently Korea, Canada, and the US).

#### 4. RECOMMENDATIONS FOR ACTION FOLLOWING THIS TCM

Actions given below refer to those which seem appropriate *for application* in the long term. Many of these actions should be initiated today so that they can be mature by 2050.

Positive statements are made in the following text to avoid repetition of the multiple qualifying statements which apply to most cases. It should be recognized that virtually *all* of the quantities, with the exception of time, are quite uncertain, increasingly so for larger time intervals.

#### 4.1. Short Term

Large-scale actions in this time frame will be based mostly on economic decisions. Little need be said about them except to note that, where possible, they should avoid steps which offer purely short-term economics with demonstrable losses in the longer term. True disposal, if such exists, of the most valuable product of reactor operation – fissile isotopes – appears to be one such action to be avoided.

Nuclear plant designs were, up to the past few years, in the forefront of thermal power plant design trends. Unfortunately, slow progress of development over the past few years has meant that fossil power plant designs have caught up and passed the evolutionary development of nuclear plants. This pattern can be ascribed only partly to regulatory pressure; much of the problem can be identified with the close conservatism of both designers and their clients.

Over the next 15-20 years there is an opportunity to conduct fairly small-scale fuel development experiments in preparation for the large-scale demonstration and commercialization steps which should occur in the medium term. Such experiments will be conducted both in research reactors and as 'piggy back' experiments in power reactors.

Convincing a skeptical public that nuclear energy is, indeed, a long-term solution to the world's energy problems is a very important activity in this time frame. One important component of the convincing process must be a plan, or a set of practical scenarios, which demonstrate that fuel cycles can be put in place which could provide a large energy supply for humanity over a period of hundreds or thousands of years.

#### 4.2. Medium Term

A characteristic of the present day nuclear energy system is that both governments and private industry are unwilling to invest billions of dollars and one or more decades of development work in a new reactor concept, however promising the concept may be. Customers will not (and probably should not) buy any radically new system without clear demonstration in a large-scale prototype. Regulators will be particularly vigilant in licensing such plants. Progress will be difficult.

There is one reality of the period up to 2050 which may help this situation. Over 500 power plants will reach the end of their operational life in this period. It should be possible to make use of a few of these old units, to modify or rebuild as demonstration plants. In some cases it might be possible to obtain a license extension for a few months or years of operation.

In the medium term (2015 to 2050), the current downward pressure from competing fossil energy suppliers should lessen, either from economic dominance by the nuclear option or by full recognition of the negative environmental consequences of fossil fuel burning. (The third alternative is obvious, but bad news for the nuclear industry). Modern engineering and construction processes should lead to a steady decrease in nuclear electricity prices in the period., plus more efficient front-end processing, should result in doubling or tripling of the specific yield of electrical energy from uranium during this period. But even at the end of the period it is unlikely that more than 10% of the energy in uranium will be extracted, in the average over the world reactor population. Roughly such an increase of yield is necessary to bring phosphate ores into the "reserve" classification - a 'hurdle' which, if achieved, would greatly increase the world uranium supply. Fast reactors will, at the end of the period, be entering commercial operation in significant numbers. Later in the 21<sup>st</sup> century it will become

possible to raise the percentage of energy extracted from uranium at least to 50%, thereby jumping the economic hurdle of uranium extraction from sea water. Passing over this hurdle would make the energy supply from nuclear fission nearly infinite.

The large investment in uranium-based technologies has overshadowed the thorium alternative except in a very few countries. During this period it is possible that thorium use will increase in these countries which will become world leaders in development of this alternative, very large, energy source.

#### 4.3. Long Term

In this phase, beginning around 2050, the most economical uranium supply may be obtained from spent fuel storage vaults, reprocessing plant storage vaults, and even from enrichment plant tailings. Recycle will be a routine practice and the measure of value of any fuel cycle will be that it produces the cheapest energy, electrical or otherwise. Preparation for this phase may begin several decades before application is needed.

The key to success in this phase is extraction of a larger and larger fraction of the total fuel energy, so that the cost of low-grade ore recovery becomes less important. Low specific reprocessing cost is the first requirement, to be met by conventional or novel methods.

Recovery of energy from stored enrichment tailing stores will depend on the ability to convert fertile to fissile material in large quantities. The FBR is the most likely vehicle for this purpose; another method may be accelerator breeding. Similarly, the value of low-grade ores depends on the amount of energy which can be extracted from them as well as on the monetary effort required for recovery.

Thorium has a powerful inherent advantage over uranium in that the conversion ratio in a wellthermalized spectrum can be very near unity. Preparation for thorium use might be achieved in the midterm period through utilizing a then-existing 'excess' of fissile materials to produce uranium-233 in a once-through cycle. This new fissile isotope then might be used in a thorium stand-alone cycle when the need arises.

#### 5.0. SOME POSSIBLE WORK PROGRAMS

#### 5.1 Increase Reactor Conversion Ratio

A higher fertile conversion ratio will result in higher burnup for given enrichment, as well as a lesser need for addition of burnable poisons. Thermal reactors utilizing thorium can reach a conversion ratio near unity even with high burnup. Fast reactors utilizing metal uranium-plutonium fuel can reach a breeding ratio of about 1.4. Combining the cycles of these two reactor types would result in a system capable of extracting a very large fraction of the fuel potential energy.

#### 5.2 Increase Fuel Burnup

The obvious benefit from increasing fuel burnup is an increase in plant capacity factor. Other changes may be necessary to gain the full advantage of the change, such as more on-power maintenance and reduced maintenance schedules. For on-power-fuelled systems the main advantage comes from a reduction in the specific reprocessing cost per unit of energy produced.

#### 5.3 Decrease Reprocessing Cost

Reduction of the specific fuel reprocessing cost will be an important development to enable recycle of low-grade spent fuel (from HWR, for example). This development also will help to control the price of uranium by lowering the recycle break-even point.

A separate important development is the economic recycle of thorium-U233 fuels. There are significant differences from uranium in both the process itself and in the activity levels of the fuel constituents which must be considered.

#### 5.4 Introduce On-Power Fuelling

Batch fuelling of power reactors carries with it the disadvantage of a periodic forced shutdown for fuelling which limits the achievable capacity factor. In addition, fuel cycle flexibility is greatly improved and parasitic neutron absorption is reduced if refuelling can be carried out with the reactor at full power.

#### 6. CONCLUSION

The first justification for urgent work on advanced fuel cycles is to demonstrate that nuclear energy *can* provide a viable long-term source of energy on a large scale, for the use of mankind. The second justification for short-term fuel cycle work is part of the overall cost-reduction imperative as fossil fuel competition intensifies. The third justification is to carry out long lead-time work (firstly to identify what is needed) which will be applied some time in the next century, as cheap uranium resources are exhausted.

This paper provides only the barest skeleton of an outline of the work needed. One useful outcome of this TCM might be the initiation of a series of discussions addressing the outstanding questions for the long term, to provide guidance for short-term fuel cycle studies.



# CLOSING THE FUEL CYCLE — A SUPERIOR OPTION FOR INDIA

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#### Abstract

The closed fuel cycle option with reprocessing and recycle of uranium and plutonium (U & Pu)for power generation allows better utilization of the uranium resources. On its part, plutonium is a unique energy source. During the initial years of nuclear fuel cycle activities, reprocessing and recycle of uranium and plutonium for power generation was perceived by many countries to be among the best of long term strategies for the management of spent fuel. But, over the years, some of the countries have taken a position that once-through fuel cycle is both economical and proliferation-resistant. However, such perceptions do vary as a function of economic growth and energy security of a given country.

This paper deals with techno-economic perspectives of reprocessing and recycling in the Indian nuclear power programme. Experience of developing Mixed Oxide  $UO_2$ -PuO<sub>2</sub> (MOX) fuel and its actual use in a power reactor (BWR) is presented. The paper further deals with the use of MOX in PHWRs in the future and current thinking, in the Indian context, in respect of advanced fuel cycles for the future. From environmental safety considerations, the separation of long-lived isotopes and minor actinides from high level waste (HLW) would enhance the acceptability of reprocessing and recycle option. The separated actinides are suitable for recycling with MOX fuel. However, the advanced fuel cycles with such recycling of Uranium and transuranium elements call for additional sophisticated fuel cycle activities which are yet to be mastered.

India is interested in both uranium and thorium fuel cycles. This paper describes the current status of the Indian nuclear power scenario with reference to the program on reactors, reprocessing and radioactive waste management, plutonium recycle options, thorium-U233 fuel cycle studies and investigations on partitioning of actinides from Purex HLW as relevant to PHWR spent fuels.

#### 1. NUCLEAR ENERGY

In the coming decades, the global energy demand will be growing by leaps and bounds mainly due to the developing countries. These nations have only limited options at their disposal to meet the steep increase in energy requirement and cannot ignore the role of nuclear energy as an alternate energy source with the potential to sustain the energy demand at the projected rates. Further, from the environmental point of view, there is a global need to deploy non-fossil sources to limit the carbon dioxide liberation to the atmosphere. As of today, the proven resources of low-priced uranium are insufficient to support a long-term and meaningful contribution to global energy demand by way of the nuclear energy.

#### 2. FUEL CYCLE OPTIONS

So far as nuclear power production is concerned, there are two fuel cycle options of relevance and under consideration at the present juncture, Viz. the once -through cycle with permanent disposal of spent fuel and the closed fuel cycle with reprocessing and recycle of U and Pu. Both the options require efficient and safe waste management strategies and whichever is the option, the need for a long term geological repository cannot be eliminated, at the present stage.

In comparison to the waste from reprocessing and recycle, the disposal of spent fuel on once -through basis does not eliminate plutonium inventory and the hazards from long term perspective can only be reduced by its sustained irradiation in reactors. The reprocessing option provides a solution to radioactive waste arisings now rather than burdening future generations with this problem.

The key factor in the success of the closed fuel cycle lies in the efficient utilization of plutonium for power generation as it can increase the quantum of energy that can be derived from a given amount of uranium which varies depending on the reactor systems used. Thus closing the nuclear fuel cycle by reprocessing the spent fuel and recycle of U & Pu helps in achieving the goal of exploiting the full potential of nuclear power.

Plutonium is an unique energy source and "by allowing us to burn virtually all of the available uranium rather than just 1% as we do at present, the use of plutonium makes nuclear energy by far the largest energy resource available, indeed one that is virtually inexhaustible"[1]. Disposal of a fossil fuel after such a low level utilization of its potential as we do now with nuclear fuel would be unheard of.

Further, with the depletion of the natural uranium and fossil resources, the recycle of reprocessed uranium with an altered isotopic content of U235 may become economically viable. The redeployment of uranium as fuel from one type of reactor to another based on its depleted U235 content is an attractive proposition. The Dupic Process based on dry route and the Purex process followed by a U/Pu co-processing and precipitation route are again recycle options with minimum processing and are considered to be proliferation resistant. Over the years, the research reactor systems have spawned HTGRs and fast reactors for better utilization of uranium and plutonium. The challenges posed by fast reactors can be met by appropriate technology development in the coming years. The interim use of plutonium in LWR and PHWR as MOX fuel has been gaining acceptance.

From the safety point of view, reprocessing technology has made vast improvements by complying with national and international regulatory requirements and its annual radioactivity releases through various forms of effluents have steadily decreased over the years. As of today, these releases are very small in comparison to the present environmental burden of Pu and other radioactive elements released through atmospheric testing of nuclear weapons.

#### 3. EVOLVING PRECEPTIONS AND INDIAN OPTIONS

Over the past few decades, the operation of the uranium fueled power reactors and the various research reactor systems have led to increased fissile inventories of Pu in the spent fuel. During the first generation nuclear fuel cycle activities, reprocessing and recycle of U & Pu for power generation was perceived by many countries to be among the best of long term strategies for the management of spent fuel. For several reasons, this perception has changed gradually over the years in some of the energy surplus countries which now consider that once-through fuel option as the economical and proliferation resistant approach that should be accepted globally. However these two criteria may differ from country to country and their perceptions would also differ accordingly.

Several nuclear energy countries like France, The United Kingdom, Russia, India and Japan had used reprocessing as part of their strategy for spent fuel management whereas some countries like Canada, Sweden and USA have opted to use the uranium once-through cycle option. USA has abundant reserves of coal, natural gas and oil. Canada has abundant natural uranium reserves and has no need to reprocess and recycle Pu. Among European countries, France has pursued a very active program on reprocessing and recycle of plutonium as MOX fuel. Thus for any country, the choice of its fuel cycle options with its minor variations should rightly be governed by its own assessment of its energy requirements on a long-term perspective, the alternate energy resources available at its command with their cost and its technological infrastructure capabilities to support and sustain modern sophisticated technologies such as nuclear power and subsequent spent fuel management.

The availability of uranium resources in India is limited. Other than the constraints to be overcome in meeting the energy security due to uncertainties in fuel supplies, purely from economic considerations, uranium procurement would add substantially to the foreign exchange component of the energy bill. The Indian reprocessing and storage costs in terms of installation and operation are substantially lower in comparison with the figures reported for western countries. Scaling up the facilities would result in cost reductions.

Further in our view from a long-term standpoint, reprocessing and recycling is more eco-friendly and provides better safeguards against plutonium getting into wrong hands as compared to once-through option, because recycling consumes plutonium while once-through leaves behind huge stocks of spent fuel which contain recoverable plutonium that may prove to be a rich and easy source for Pu after several hundred years of cooling.

Thus from the Indian stand point, the reprocessing and plutonium recycle option is not only considered to be superior option but also inevitable. This perception had emerged some four decades ago and has since remained unaltered.

#### 4. THE INDIAN NUCLEAR ENERGY PROGRAMME

The Indian nuclear resources have been estimated [2] to be around 60,000 tons of U and around 360000 tons of Th. In terms of fossil fuel, this is equivalent to around 1.2 billion tons of coal equivalent through pressurized heavy water reactors (PHWR) and around 800 billion tons of coal equivalent through fast breeder reactors (FBR) and other reactor systems using thorium. Clearly, this constitutes a resource several times larger than any other resource that we have in our country for bulk electricity production. Table I. gives the profile of the energy resources in India in terms of coal equivalent.

The nuclear energy programme in India envisages three stages of implementation involving installation of uranium fueled thermal reactors in the first phase followed by utilization of plutonium in fast breeder and other types of reactors and in the third phase, utilization of reactor systems based on U233- Th cycle, which we consider to be the ideal fuel cycle of the future, from Indian context.

The first phase of our Programme is essentially based on the utilization of PHWRs for power generation with fuel reprocessing, plutonium recycle and efficient waste management as the strategies for the back end of the Fuel Cycle.

The choice of the Reprocessing and Plutonium Recycle option has endowed the program with a variety of mid-course options in both U and Th fuel cycle with Pu forming the vital link between the two.

RESOURCES	QUANTITY*
COAL	196 billion T
OIL	0.6 billion T (coal eqvt.)
GAS	540 billion $m^3$
HYDROELECTRIC (per year)	84 Gwe at 60% CF
URANIUM (in heavy water reactors)	380 Gwe-yr
URANIUM (in breeder reactors)	50, 000 Gwe-yr
THORIUM (in breeder reactors)	360, 000 T
	(> 200,000 Gwe-
	yr)

## TABLE IENERGY RESOURCES IN INDIA

#### 4.1. NUCLEAR REACTORS AND POWER GENERATION

Besides the two BWRs at Tarapur, there are several operating PHWRs with a design capacity of 220 MWe each. Some more reactors of similar type including two each of 500MWe are under different stages of planning, construction and commissioning. Under Fast Breeder Reactor (FBR) technology development programme, a 40 MW<sub>th</sub> Fast Breeder Test Reactor (FBTR) is operational at Kalpakkam and the design of a 500 MWe Prototype Fast Breeder Reactor (PFBR) is in progress. In addition to PHWRs and FBRs, it is proposed to include LWRs and Advanced Heavy Water Reactors (AHWR) in the power programme.

These activities call for extensive recycling of Pu generated from PHWR's in FBR's or in the existing PHWRs or in newly conceived reactors of the AHWR type. These concepts are evolved to maximize the use of available resources and are heavily dependent on successful reprocessing and recycle of Pu.

#### 4.2. FUEL REPROCESSING

Over the years, in tandem with the increase in spent fuel arisings from the growth of nuclear power, the reprocessing and nuclear waste management capabilities have been augmented to keep pace with the plutonium demands. There are now two reprocessing facilities to treat spent fuels from PHWRs.

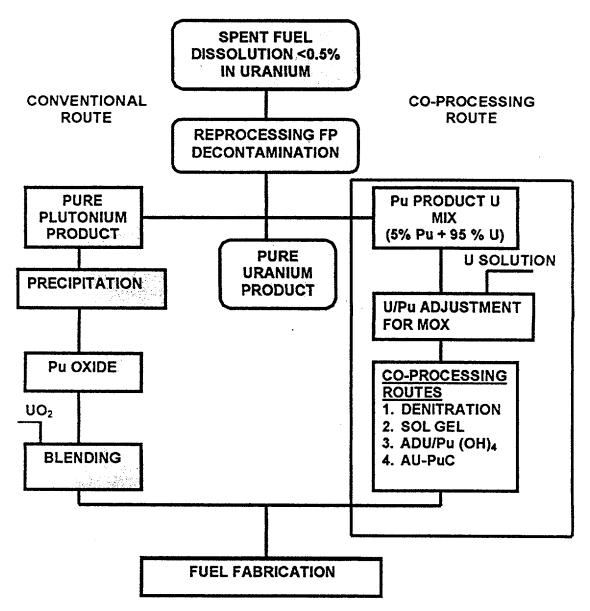


Fig. 1 Schematic Diagram of the Fuel Fabrication Route

#### 4.2.1. Recovery of U/Pu by co-processing for MOX fuel

Presently reprocessing and recycle of uranium and plutonium together without their individual separation is considered as one of the alternate approaches for closing the nuclear fuel cycle. This approach becomes attractive for reprocessing the BWR spent fuel so that the fissile values available in it can be recovered and utilized for fabrication of MOX fuels for PHWRs. From the reprocessing and MOX fuel fabrication point of view, this approach comprises the following three major tasks : 1) Co-Processing and recovery of U and Pu together in Purex process, 2) Co-conversion of U and Pu product mix into their oxides and 3) Fabrication of PHWR - MOX fuel from the mixed oxide product.

The Purex flow-sheet can be modified for reprocessing spent fuel from LWR by co-processing as shown in Fig.1. The flow sheet conditions at the partitioning stage of the process can be adjusted so that the U/Pu composition in the stripped product will be about 95%U and 5% Pu. This would form the master feed that can be diluted further with uranium to the required Pu content and processed further for conversion to MOX using any of the several possible routes..

In the wet process route, certain amount of fission product decontamination is envisaged and the process does not eliminate enriched ADU generation during LWR fuel reprocessing. A major fraction of decontaminated LEU follows the ADU route as a separate product. About 10-15% of LEU ( of the total input U) with enhanced Pu content (Pu enrichment 5%) forms the second product. Both the streams can be used for PHWR fuel fabrication.

Over the years BARC have been working on the development of several alternate routes for the conversion of U, Pu and Th products in their nitrate form to their corresponding oxides. Expertise and technological back-up for the various co-conversion process routes like Sol-gel, mixed oxalate precipitation, hydroxide precipitation, ammonium uranyl-plutonyl carbonate precipitation and direct denitration are available with us.

The Purex process, with a co-processing route for the production of Pu/U MOX in an integrated facility, may not pose major technical problems. The economic aspects and safety from radiological point of view are being examined.

#### 4.3 THORIUM/URANIUM 233 FUEL CYCLE

To meet the challenges of thorium based fuel cycle, R&D efforts are directed towards extractive metallurgy of thorium, fuel fabrication and its utilization in reactors, reprocessing of irradiated thorium for U233 recovery and studies on U233 based reactor systems. Demonstration facilities have been operated in all these domains. With U233 as fuel, India has operated three low power research reactors, PURNIMA-II & III and KAMINI, a 30KW reactor with U233-Al alloy as fuel.

Thorium in the form of oxide fuel bundles was used in each of the two units of the Kakrapar atomic power station (KAPS I & II) for the purpose of initial flux flattening. It is proposed to follow this scheme for all future PHWRs as well. Additionally, there has been a continuous programme for irradiation of thorium rods in the research reactors located at Trombay. The irradiation of thorium in the blanket region of FBTR will commence shortly and it will also help in U233 generation.

#### 4.4 FUEL FABRICATION AND EMERGING CONCEPTS IN Pu RECYCLE

Based on the plutonium-based fuel fabrication experience on pilot plant scale at Trombay, an industrial scale Advanced Fuel Fabrication Facility (AFFF) has been setup at Tarapur to meet the MOX fuel fabrication requirements. This facility will meet the fuel requirements of MOX for thermal reactors, and the initial startup requirements of Prototype Fast Breeder Reactor (PFBR). A facility to cater to the regular requirements for PFBR is being planned to be set up at Kalpakkam.

Though the FBRs are the best long term options for Pu recycle and burning, utilization of plutonium in PHWRs also offers considerable flexibility in terms of fuel cycle variations. Using U-Pu MOX in PHWR,

it is possible to get improved fuel utilization and extended burnup resulting in significant increases in the overall installed capacities.

The conceptual once through thorium fuel cycle studies reveal that Th can be used in combination with Pu in reactors to high discharge burnups. It can burn Pu to a very significant extent. Studies on various reactor concepts indicate that heavy water reactors are second only to molten salt reactor systems as a choice for thorium utilization. An attractive option would be the use of Pu as a key to initiate the thorium cycle. As part of this programme, India is working on the design of an AHWR. This reactor requires an initial inventory of uranium-233 as well as plutonium. It derives 75-80% of its power from thorium in a self-sustaining mode of U233-Th cycle. The reactor needs an initial input of Pu in the form of mixed U-Pu oxide which contributes to 20-25% of the power and the recurring need for plutonium is relatively small. U233 in the thorium is adjusted to be at the self sustaining level and a discharge burnup of 20,000MWD/T is attained using plutonium as additional make up in the form of U,Pu oxide pins. The Pu pins are placed where neutron spectrum is most advantageous to Pu and the thorium fuel remains uncontaminated by the long lived plutonium and transplutonium actinides.

Coprocessing Thorium-Uranium 233 for the AHWR, which maintains Uranium proportion unaltered, is yet another attractive option that we are examining.

#### 4.5. Pu Recycle - Technology Developments

The Indian policy has been "Reprocess and Recycle" from the very beginning. The experimental work on Pu fuel development has been initiated in the late seventies which has lead to the fabrication of mixed carbide fuel for FBTR and to the loading of MOX lead test assemblies in TAPS.

#### 4.5.1. Irradiation Experiments

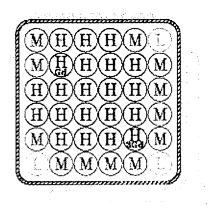
The irradiation experiments started in the late seventies with the irradiation of  $UO_2 - PuO_2$  pins in Pressurized Water Loop (PWL) in Cirus Reactor. These irradiation experiments were mainly aimed at proving MOX fuel pins of BWR design. In addition, pins with PHWR design were also irradiated in PWL. A list of important irradiation experiments are given in Table-II. The experience obtained during fabrication lead to the establishment of a flowsheet and formed the basis for the design of our Advanced Fuel Fabrication Facility (AFFF).

#### TABLE II

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

#### Fuel Composition : UO<sub>2</sub> - 4% PuO<sub>2</sub>

Sr. No.	Designation	Max.linear Rating W/cm	Bum-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.



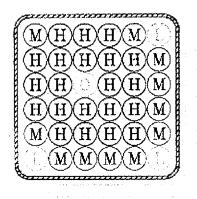


Fig. 2 (a) Standard LEU Design  $L = 1.6 \% U^{235}$   $M = 2.1 \% U^{235}$  $H = 2.6 \% U^{235}$  Fig. 2 (b) Replacement All Pu Design  $L = 0.9 \% Pu^{235}$   $M = 1.55 \% Pu^{235}$  $H = 3.25 \% Pu^{235}$ 

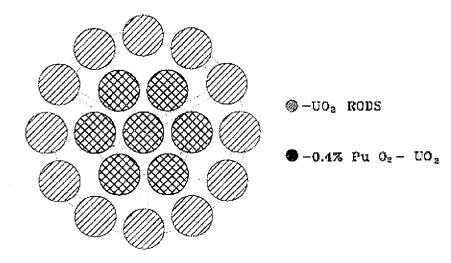


Fig. 3 MOX Fuel Bundle for PHWR

#### 4.5.2. Fast Breeder Test Reactor (FBTR) Fuel

For the sodium-cooled FBTR, built at Kalpakkam, we have used an indigenously developed mixed carbide (MC) fuel, which has a composition of  $(U_{0.3}Pu_{0.7})C$ . Initially designed for a burnup of 50,000 MWD/Te, the performance of the fuel, on recent evaluation, has been found to be quite satisfactory and an extension of burnup to 70,000 MWD/Te is now under consideration. This programme on the fast reactor is very important to us for enhancing the potential for nuclear power generation using the limited uranium resources available in our country. The confidence generated in building and operating this reactor has given us the impetus for designing a prototype 500 MWe fast breeder reactor, proposed to be taken up for construction shortly.

Fuelling Scheme No.	Inner zone Fuel/Burn-up MWD/Te	Outer zone Fuel/Burn-up MWD/Te	Average Burn-up MWD/Te	Max. Bundle Power (KW)	Max. channel power (MW)
1*	N.U./10700	N.U./6000	6600	395	2.89
2	MOX-7/14500	MOX-7/10200	10840	429.6 in MOX-7 bundle	3.14
3	N.U./11000	MOX-7/10200	10300#	422.5 in N.U. bundle	3.11
4	MOX- 7/12/17350	MOX- 7/12/13700	14300	470 in MOX-7/12 bundle	3.45

#### CORE OPTIMISATION FOR TARGET MOX REACTOR

Note : Powers are normalised to total power of 6.551 MW to coolant

\* Reference Natural UO<sub>2</sub> fuel bundles in all the channels

<sup>#</sup> Observed burn-up will be about 10,700 MWD/Te

#### 4.5.3 TAPS Fuel

A programme to partially substitute the LEU (Low Enriched Uranium) assemblies by MOX assemblies in the BWRs at Tarapur is in progress. These assemblies have been fabricated at Advanced Fuel Fabrication Facility. Figs. 2 (a) and 2 (b) show the designs of the fuel assembly; the standard LEU fuel and the substitute all-Pu assembly. At present ten assemblies have been fabricated and six are undergoing irradiation in reactors. Some of these assemblies have so far crossed a burnup of 10,000 MWD/Te and their irradiation is continuing. They have been loaded adjacent to TIP (Travelling Incore Probe) locations to facilitate continuous monitoring. Their performance has been good based on off-gas monitoring and sipping. The fraction of MOX in the core is planned to be progressively raised as experience is gained.

#### 4.5.4. PHWR Fuel

A study has been made in the use of MOX fuel in PHWRs with minimum deviation in the basic design of the fuel bundle, the reactor hardware, the control system etc. For the introduction of MOX in PHWRs, the limits on bundle power and channel power are already fixed by the present Nat.UO<sub>2</sub> design. Likewise the locations and the number of control and safety devices and fuel handling system are fixed. After analysing different cases (Table III), a MOX-7 design (Fig.3) with central seven rods containing 0.4% PuO<sub>2</sub> in nat.UO<sub>2</sub> was selected for further implementation. The selected fuelling scheme will utilise nat.UO<sub>2</sub> for the central 44 channels. The studies indicate that the core average discharge burnup will improve to 10,700 MWD/Te instead of 6,700 MWD/Te at present.

# 4.6. RADIOACTIVE WASTE MANAGEMENT AND PARTITIONING AND TRANSMUTATION OPTIONS

The Indian program on safe management of radioactive wastes envisages two distinct modes of final disposal in respect of radioactive wastes; near-surface engineered, extended storage for low and intermediate-level active wastes and deep geological disposal for high-level and alpha bearing wastes.

A waste immobilization plant for the treatment of HLW is operational at Tarapur. It is a semicontinuous pot glass process involving calcination followed by melting in the process vessels. Two more waste immobilization plants are being set up at Trombay and Kalpakkam. Use of joule heated ceramic melters is under development. A solid storage and surveillance facility (SSSF) has also been set up for interim storage of vitrified HLW.

As regards ultimate disposal, the Indian choice is focused on igneous rock formations and some selected sedimentary deposits. Investigations are in progress for evaluation of candidate sites for a repository.

From environmental safety considerations, the separation of long-lived isotopes and minor actinides (MA) from reprocessing high level waste would enhance the acceptability of reprocessing and recycle option. The separated MA are suitable for recycling with MOX fuel. However, the advanced fuel cycles with such recycling of uranium and transuranium elements call for additional sophisticated fuel cycle activities which are yet to be mastered. Fig. 4 shows the conceptual schematic diagram of the advanced closed fuel cycle.

The main objective of partitioning high level waste is that it shall lead to a safer waste, more acceptable to the public. The removal of long-lived alpha emitting actinides from these wastes under P&T option would greatly reduce their long term radiological hazards. Removal of shorter lived fission products like Sr90 can reduce the heat generation from these wastes. Further, recovery of useful nuclides from this waste will make the waste management with P&T more economical and viable.

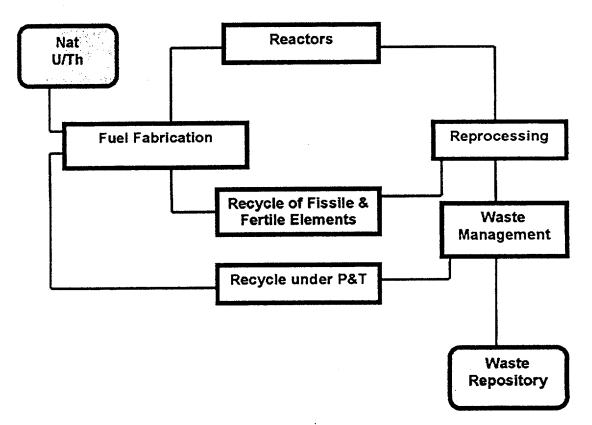


Fig.4 Schematic Diagram of Advanced Closed Fuel Cycle

From the Indian context, the present efforts are limited to the partitioning of the long lived actinides from the HLW as any reduction in the alpha burden of these wastes would render them safer with respect to long term disposal. At an appropriate time, a long term policy on the final utilization/ transmutation of the recovered actinides would be evolved, based on the available state of the art technology at that point of time.

Studies are in progress for the quantification of the PHWR spent fuel arisings, the radiological source terms of the relevant actinides and fission products in Purex HLW after reprocessing and evaluation of their hazard ranking. CMPO based solvent extraction and extraction chromatographic studies with HLW are in progress to propose suitable flow sheets for partitioning of the relevant actinides from these wastes and to reduce the alpha burden to very low levels. Other extractants are also being explored in this context.

The man-rem expenditures associated with P&T tasks should be evaluated and compared with the dose the future generation would be expected to receive in the distant future, in the event of leachates from geological repository reaching the biosphere without P&T. Such a comparison would be of help in reaching a decision regarding P&T option.

In the Th/U233 fuel cycle, the amount of transuranium nuclides generated is smaller by several orders of magnitude as compared to that arising from U235/U238 fuel cycle. In the case of LWR, the major hazard is from Am, Np, Cm isotopes and the left out Pu and U, whereas in the  $^{232}$ Th- $^{233}$ U fueled reactor, the hazard is mainly from  $^{231}$ Pa. In the very long term perspective, the presence of U233 and U234 would also need consideration as Ra226 in the case of U fuel cycle.

#### 5. CONCLUSION

Opting for a closed nuclear fuel cycle, a significant fraction of the energy output could come from the materials recycled from reprocessing. From Indian stand-point, under given limited resources of nat. U, this option is not only superior but also an inevitable one. Closed fuel cycle with Pu recovery on a 'reprocess to recycle mode' can lead to a viable, safe and eco-friendly reprocessing and waste management strategy. Development of flow-sheets for co-processing of Pu together with U, without their individual separation by Purex process for conversion to MOX in integrated facilities, appears attractive . Advancements in reactor research have spawned several new alternatives for the better utilization of Pu and U. These systems are yet to be perfected prior to commercial exploitation and public acceptability. Meanwhile, the available fissile inventory can be redeployed on an interim basis in the existing reactor systems which can lead to an enhanced energy profile. The emerging reactor concepts such as the AHWR, which integrates both U/Pu and Th/U233 fuel cycles can yield valuable information and can lead India closer to its ultimate goal, ie. the Th-U233 fuel cycle.

From environmental safety considerations, the separation of long-lived isotopes and minor actinides (MA) from reprocessing high level waste would enhance the acceptability of reprocessing and recycle option.. However, the advanced fuel cycles with such recycling of uranium and transuranium elements call for additional sophisticated fuel cycle activities which are yet to be mastered. Any strategy for sustained nuclear power generation that involves recycling of reprocessed U and Pu will have to face these challenges.

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#### FUEL CYCLE OPTIONS FOR LIGHT WATER REACTORS IN GERMANY

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#### Abstract

In Germany 19 nuclear power plants with an electrical output of 22 GWe are in operation. Annually about 450 t of spent fuel are unloaded from the reactors. Currently most of the spent fuel elements are shipped to France and the United Kingdom for reprocessing according to contracts which have been signed since the late 70es. By the amendment of the Atomic Energy Act in 1994 the previous priority for reprocessing of spent nuclear fuel was substituted by a legal equivalency of the reprocessing and direct disposal option. As a consequence some utilities take into consideration the direct disposal of their spent fuel for economical reasons.

The separated plutonium will be recycled as MOX fuel in light water reactors. About 30 tons of fissile plutonium will be available to German utilities for recycling by the year 2000. Twelve German reactors are already licensed for the use of MOX fuel, five others have applied for MOX use. Eight reactors are currently using MOX fuel or used it in the past.

The spent fuel elements which shall be disposed of without reprocessing will be stored in two interim dry storage facilities at Gorleben and Ahaus. The storage capacities are 3800 and 4200 tHM, respectively. The Gorleben salt dome is currently investigated to prove its suitability as a repository for high level radioactive waste, either in a vitrified form or as conditioned spent fuel.

The future development of the nuclear fuel cycle and radioactive waste management depends on the future role of nuclear energy in Germany. According to estimations of the German utilities no additional nuclear power plants are needed in the near future. Around the middle of the next decade it will have to be decided whether existing plants should be substituted by new ones. For the foreseeable time German utilities are interested in a highly flexible approach to the nuclear fuel cycle and waste management keeping open both spent fuel management options: the closed fuel cycle and direct disposal of spent fuel. In this context the tendency to increase the fuel burnup and to establish sufficient storage capacity for long-term interim storage is important.

# 1 Introduction

In 1997, roughly 34 % of the electricity in Germany were generated by nuclear energy. Nineteen nuclear power plants (13 PWR, 6 BWR) are in operation with an electrical output of 22,0 GWe. Nine power reactors, including six of Russian origin in the new Federal States, and several research and prototype reactors (e.g. THTR) have been shut down and are being decommissioned. One additional PWR (Mülheim-Kärlich) will probably be decommissioned too after the license has been cancelled by the German Supreme Administration Court.

Uranium enrichment is performed in the Urenco plant at Gronau with a licensed capacity of 1000 tSWU/y. One plant for the fabrication of LWR fuel assemblies is in operation at Lingen. The Hanau facilities for the fabrication of uranium oxide and MOX fuel have been shut down and the Siemens plant at Karlstein has also terminated nuclear fuel production. Other facilities designed for the fabrication of HTR and research reactor fuel were already shut down some years ago.

The Atomic Energy Act [1] was amended in 1994. One of the most important changes was that the priority for reprocessing of spent nuclear fuel has been substituted by a legal equivalency of the reprocessing and direct disposal route. As a consequence some utilities take into consideration the direct disposal of their spent fuel for economical reasons.

Handling, treatment, transport and disposal of radioactive wastes are to be licensed according to the German Radiological Protection Ordinance [2] unless these processes are covered by a licence according to the Atomic Energy Act. The waste producer is required to deliver the waste to a Federal facility for securing and disposal or to a state-run collection center (Landessammel-stelle) of the Federal States. If the activity of the waste is low enough it may be licensed to be disposed of as non-radioactive waste.

# 2 Management of spent nuclear fuel

The operator of a German nuclear power plant is obliged to present concepts for the safe management of spent fuel for six years in advance. Annually about 450 tHM of spent fuel arise from the operation of German nuclear power plants. The spent fuel elements are stored on site in the reactor pools for at least one year, and then transported to COGEMA or BNFL for reprocessing, or to AFR ("away from reactor") storage facilities.

Between 1966 and 1996 about 6990 tHM of spent fuel have been discharged from the power reactor cores. 4230 tHM have been brought to France, 480 tHM to the UK, 290 tHM to the former USSR (spent fuel from Rheinsberg and Greifswald), and 130 tHM to other facilities. 1860 tHM are currently being stored at the reactor sites or AFR storage facilities.

### 2.1 Interim storage

For the AFR storage of spent fuel there are three facilities in operation. Two storage facilities at Ahaus and Gorleben with capacities of 4200 tHM and 3800 tHM, respectively, are licensed for dry-storage casks (e.g. of type CASTOR). The storage period of casks is limited to 40 years. The Gorleben facility is authorised also for the storage of vitrified high level waste in casks. A wet storage facility (Zentrallager für abgebrannte Brennelemente, ZAB) is in operation near the Greifswald reactors with a capacity of 560 tHM. The licence expires on June 30, 2000, unless there will be an extension of that date. At the same site a dry storage facility (Zwischenlager Nord, ZLN) has been constructed. The atomic license is expected in May 1998. Apart from spent fuel elements from the Rheinsberg and Greifswald reactors it is also intended to store at the ZLN radioactive wastes from the decommissioning of these reactors.

### 2.2 Reprocessing

in 1989 the German utilities decided not to pursue the domestic reprocessing project at Wackersdorf any longer. As a consequence, new reprocessing contracts were negotiated with reprocessors in France and the UK. The old contracts - signed before 1989 - cover about 5500 tHM, the new contracts roughly 3000 tHM with options for reprocessing up to the year 2015. In accordance with these contracts the radioactive wastes from the reprocessing of German fuel will be taken back by the German utilities. The first transport of vitrified high level waste from La Hague took place last year.

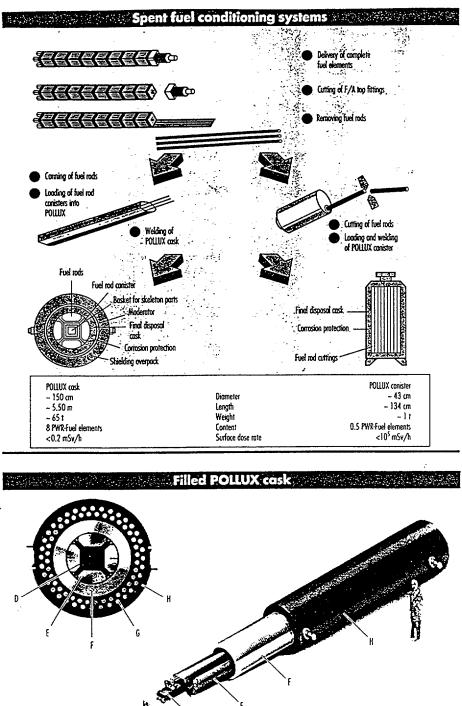


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Figure 1: Scheme of spent fuel conditioning at the PKA plant

# 2.3 Direct disposal

As an alternative option to reprocessing, a concept for the direct disposal of spent fuel assemblies has been developed. The feasibility and principal safety aspects were evaluated in comprehensive studies between 1979 and 1985 [5]. After the assessment of the results of these studies the Federal Government decided to further support the development of the technologies for direct disposal to achieve technical maturity. These investigations including R&D and the optimisation of a repository concept suitable both for direct disposal of spent fuel and the waste arising from its reprocessing were essentially finished at the end of 1995. In accordance with the concept of direct disposal a pilot conditioning plant (PKA) for spent fuel and other radioactive waste is presently under construction at Gorleben.

PKA is a multi-purpose facility designed to enable all necessary operations for spent fuel conditioning to be demonstrated on a representative scale. It is also envisaged that the procedures should be further developed and applied on an industrial scale. The pilot plant will primarily treat those fuel elements not currently envisaged for reprocessing. These include:

- LWR fuel elements with very high burnup and mixed oxides;
- High-temperature reactor fuel elements.

A technical concept for direct disposal in salt formations was developed for industrial application. It includes various steps for the conditioning of the spent fuel into a form suitable for final disposal. Depending on the type of final disposal packages required, LWR fuel rods may be packaged as a whole (POLLUX cask) or as cut-up sections of nearly one metre length (POLLUX canister). HTR fuel pebbles shall be transported pneumatically into the final disposal canisters. The POLLUX cask is based on a double containment concept, consisting of a leak-tight welded steel containment, which assures safe containment of spent fuel, protection against mechanical impacts and protection against corrosion [6]. The spent fuel conditioning systems and a scheme of the POLLUX cask are shown in Figure 1. Hot commissioning of the PKA plant is scheduled for 1999.

# 3 Plutonium utilisation

### 3.1 Current practice

The plutonium separated at La Hague and Sellafield within the existing reprocessing contracts of German utilities shall be recycled as MOX fuel in light water reactors. About 30 tons of fissile plutonium will be available to German utilities for recycling by the year 2000. Twelve German reactors (10 PWR, 2 BWR) are already licensed for the use of MOX fuel, five others have applied for MOX use (Table 1). Eight reactors are currently using MOX fuel or used it in the past. The licenses include limitations for the number or share of MOX elements being loaded into the reactor core. The values range from 9 to 50 % for the different power plants with a mean value of 30 % corresponding to about 400 tHM of MOX fuel in the cores in total. This means that about 130 tHM of MOX fuel could be burnt annually if the licensed limits were optimally used. Each core loading, however, requires an individual permission by the licensing authority.

Burnup calculations for standard fuel elements (3,6 % U-235 enrichment in UO<sub>2</sub> elements, 4,4 %  $Pu_{iss}$  in MOX elements, 40 GWd/tHM) and different MOX : UO<sub>2</sub> ratios in the core have shown that for a MOX content of 35 % there is approximately a balance between plutonium production and burning (Figure 2). If the MOX content is lower than 35 % there is a net increase of plutonium which, however, is lower than for pure UO<sub>2</sub> elements.

At the end of 1997, only 70 tHM of MOX fuel were actually in the reactor cores which is approximately one sixth of the licensed maximum inventory. A total of 225 tHM of MOX fuel elements containing about 9 tons of plutonium have been irradiated in German reactors by that

# Table 1: Licensing of MOX use in German power reactors (December 1997)

Power Plant	License G=Granted A=Applied	Max No of MOX elements per reload	Max No of MOX elements in the core	Total No of core ele- ments	Max. MOX portion (in %)	MOX al- ready used?
PWR Brokdorf	G	1)	64	193	33	yes
PWR Unterweser	G	16	64	193	33	yes
PWR Grohnde	G	16	64	193	33	yes
PWR Emsland	G	16	48	193	25	
PWR Obrigheim	G	8	28	97	29	yes
PWR Philippsburg 2	G	24	72	193	37	yes
PWR Neckarwestheim 1	G		16	177	9	yes
PWR Neckarwestheim 2	G		72	193	37	
PWR Isar 2	G	24	96	193	50	
BWR Gundremmingen B	G	68	300	784	38	
BWR Gundremmingen C	G	68	300	784	38	yes
PWR Grafenrheinfeld	G	16	64	193	33	yes
PWR Biblis A	A	24	80	193	41	
PWR Biblis B	A	24	80	193	41	
BWR Brunsbüttel	A	32	136	532	26	
BWR Krümmel	A	40	212	840	25	
BWR Isar 1	A	60	320	592	54	

<sup>1)</sup> according to self-production

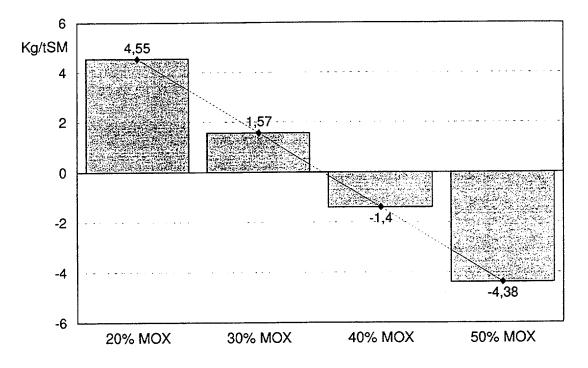


Figure 2: Plutonium balance (kg/tHM) in the reactor for different MOX contents

date. The main reasons for this relatively low recycling rate are the worldwide lack of sufficient MOX fabrication capacity on the one hand and delays in the outstanding licensing procedures on the other hand. In this context it is noteworthy that in summer 1995, after a lengthy political controversy with the licensing authority, the German utilities decided not to pursue the completion of the new Siemens MOX fabrication plant at Hanau. Contracts for the fabrication of MOX fuel assemblies since have therefore been signed with companies in Belgium, France and the UK.

### 3.2 Influence of increased burnups on the plutonium production

The increase of  $UO_2$  fuel burnups is a development which shall utilise the fuel more efficiently. Material and related potential safety aspects are the main limitations. Without any changes of the reactor concept higher burnups are achieved by a higher initial enrichment. In order to compare the effect of plutonium reduction by higher burnups, it is necessary to chose initial U-235 enrichments which lead to equal reactivities at the time of unloading. In a GRS study [4] the

Final Burnup	Initial Enrichment	Pu/GWd (grams)						
(GWd/tHM)	(wt %)	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242	Pu total	
30	2,76	4	172	79	29	17	305	
40	3,39	5	142	69	28	19	263	
50	4,06	6	123	62	26	20	237	
60	4,77	7	111	57	24	20	220	

Table 2: Grams of plutonium per GWd in spent UO<sub>2</sub> fuel elements for different burnups

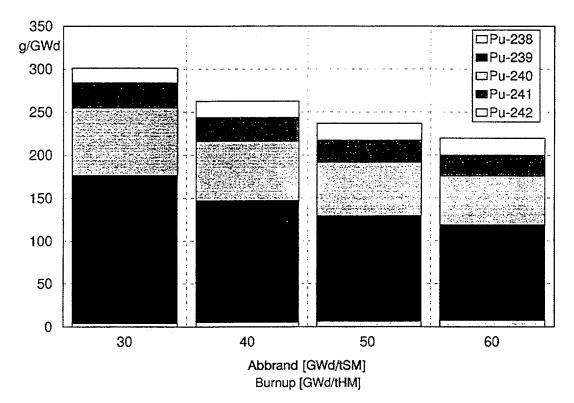


Figure 3: Plutonium production per GWd in spent UO<sub>2</sub> fuel elements for different burnups

plutonium contents in  $UO_2$  fuel elements have been calculated for different burnups and a decay time of 7 years after unloading. The results, normalized to the energy produced, are shown in Table 2 and Figure 3. It can be seen that an increase of burnup from 30 to 60 GWd/tHM reduces the specific plutonium content, normalised to the generated electricity, in the fuel element.

Most of the German power reactor strategies show a significant trend to higher burnups. According to a questionnaire of the German Federal Ministry of the Environment, Nature Conservation and Nuclear Safety (BMU) the mean burnup of the next unload varied between 34 and 47 GWd/tHM. The values of the aspired burnups varied between 37 and 50 GWd/tHM.

### 3.3 Advanced concepts for plutonium burning

Typical spent MOX fuel elements with a burnup of 40 GWd/tHM still contain about 48 kg Pu per tonne with a Pu(fiss) content of more than 50 %. This means that in principal the reprocessing and recycling of the plutonium can be performed more than one time (multiple recycling). The calculation of the plutonium in spent MOX elements shows, that there is a net decrease of the plutonium content with each recycling step which reaches a kind of saturation approximately after the third step.

Plutonium burning in LWR MOX fuel elements could be further optimised if the moderation ratio would be increased. This change of moderation is in particular necessary if the reactor core is loaded with 100 % MOX fuel. Concepts for such a reactor type have been developed. The main advantage is the high consumption of plutonium by which a significant reduction of the stocks could be achieved.

Calculations with the OREST code have shown that a thermal Thorium-Plutonium reactor could be an effective plutonium burner. In such a reactor plutonium will be burnt but not produced. The annihilation rate for plutonium per energy production unit is approximately double as high as in the MOX elements of a conventional LWR.

A similar idea is the basis for reactor concepts with inert fuel matrices. In those reactors the plutonium will also be burnt without any new production. The annihilation effect is comparable to that of the Thorium-Plutonium reactor mentioned above. A major disadvantage, however, is the positive void coefficient.

Plutonium can also be burnt in fast reactors if the breeder zone is omitted. The quantities of burnt plutonium are comparable to those in MOX elements of thermal reactors. An advantage of fast reactors is that the burning rate is nearly independent of the plutonium quality. This means that also multiply recycled plutonium can be used in the reactor [4].

# 4 Management of reprocessed uranium and depleted uranium

Reprocessed uranium (repu) is currently stored at facilities of the reprocessors. The technical feasibility of recycling in power reactors has been demonstrated with approximately 3 tons of reprocessed uranium. German utilities are planning to decide on the large-scale reuse of reprocessed uranium at the beginning of the next decade.

The depleted uranium from the enrichment process can also in principle be used for the fabrication of fuel elements. At the Gronau site about 4600 tons of uranium tails were stored in the form of UF<sub>6</sub> at the end of 1995. For long-term storage a conversion into the more stable oxide is desirable for safety reasons. The decision whether the recovered or depleted uranium will be reused or treated and disposed of as radioactive waste is left to the utilities and depends mainly on economical aspects.

# 5 Control and interim storage of radioactive wastes

The radioactive waste management concept includes conditioning, interim storage and final disposal as main steps. As far as non-heat-generating waste is concerned, a guideline [3] has been issued in 1989 to improve the surveillance of all waste management steps. Basic purpose of the guideline is to establish complete regulatory control of the relatively large volumes of this waste from their generation until final disposal. Registration and regulatory control of the waste streams and inventories are supported by a computerised system (AVK).

The operators of nuclear facilities are responsible for waste conditioning and interim storage until repositories become available. The radioactive wastes from nuclear power plants are treated and stored onsite or offsite. Two offsite facilities are in operation at Gorleben and Mitterteich. Radioactive wastes from universities, medical and industrial applications or nuclear research centres are stored at state-run collection centres operated by the Federal States or at the nuclear research centres. Radioactive wastes from the reprocessing of spent fuel in France and the UK will be redelivered to Germany and stored at interim storage sites at Gorleben and Ahaus. All categories of radioactive waste will later be disposed of in deep geological formations. The construction and operation of the repositories are legal obligations of the Federal Government. The Federal Office for Radiation Protection (BfS) is in charge of this task.

# 6 Final disposal of spent fuel elements and radioactive wastes

Research and development for radioactive waste repositories started very early in Germany. The former salt mine <u>Asse</u> is being used as an underground research facility. Between 1967 and 1978 about 125,000 containers with low level waste and 1300 containers with intermediate level waste were deposited in the mine.

At present a repository at <u>Morsleben</u> (ERAM) is in operation. In 1979, the former GDR started using the abandoned salt mine as a repository for low and intermediate level wastes. After German reunification the responsibility for the operation of ERAM was transferred to the BfS. It is planned to dispose of at least 40,000 m<sup>3</sup> of radioactive wastes until the expiration of the current license on June 30, 2000.

The abandoned <u>Konrad</u> iron ore mine is intended to be used as a repository for radioactive wastes with negligible heat generation. The licensing procedure is in an advanced state. A decision of the licensing authority is expected in 1998. After a construction phase of about five years the repository could go into operation at the turn of this century. The design capacity is 650,000 m<sup>3</sup> of radioactive waste.

The <u>Gorleben</u> salt dome is being investigated as a repository for all kinds of radioactive waste, particularly vitrified high level waste and spent fuel assemblies. The results of the aboveground exploration and test drillings into the salt dome confirmed the prospective suitability of the site as a radioactive waste repository. Currently, the underground exploration is in progress. Two shafts have been sunk for this purpose at a depth of 930 m and 840 m, respectively, and the galleries for the exploration have been excavated. A decision on the suitability of the salt dome for the construction of a repository will be made by 2005 after the underground exploration has been finished.

# 7 Perspectives of the future development of the nuclear fuel cycle

The future development of the nuclear fuel cycle and radioactive waste management depends on the future role of nuclear energy in Germany. According to estimations of the German utilities no additional nuclear power plants are needed in the near future. Around the middle of the next decade it will have to be decided whether existing plants should be substituted by new ones. The development of a joint French-German PWR will lead to a new safety standard of nuclear power generation. For the foreseeable time German utilities are interested in a highly flexible approach to the nuclear fuel cycle and waste management keeping open both routes: the closed fuel cycle and direct disposal of spent fuel. In this context the tendency to increase the fuel burnup and to establish storage capacity for long-term interim storage is important.

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#### TECHNICAL BASIS FOR THE PROPOSED HIGH EFFICIENCY NUCLEAR FUEL PROGRAM

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#### Abstract

Greenhouse gas emissions from fossil fired electricity generating stations will dramatically increase over the next 20 years. Nuclear energy is the only fully developed technology able to supply large amounts of electricity without generation of greenhouse gases. However, the problem of noncompetitive economics and public concerns about radioactive waste disposal, safety, and nuclear weapons proliferation may prevent the reemergence of nuclear power as a preferred option for new electric energy generation in the U.S. This paper discusses a new research program to help address these issues, by developing fuel designs capable of burnup values in excess of 60 MWD/kgU. The objectives of the program are to

- Improve the reliability and robustness of light water reactor fuel, thereby improving safety margins.
- Significantly increase the energy generated by each fuel loading, thereby achieving longer operating cycles, higher capacity factors, and lower cost electric power.
- Significantly reduce the volume of spent nuclear fuel discharged for disposal by allowing more energy to be extracted from each fuel element prior to discharge.
- Develop fuel that is much more proliferation resistant.

### 1. INTRODUCTION

Trends in the world's population and energy use during the past century show dramatic and relatively parallel increases in both. These trends are expected to continue in the near future (at least the next 20 years), and the total world energy consumption in 2015 will be about 54% higher than it is today, led by growing demand in Asia [1]. The demand for electricity is expected to increase more rapidly than the demand for other forms of energy throughout the world and nearly double by 2015. Coal will be used to generate much of that electricity in the developing countries. In the industrialized world, there are also dramatic structural changes underway in the electric power industry to enhance competition in the generation segment of the business. This, along with ample natural gas supplies and relatively low gas prices, has made natural gas the preferred fuel for many power producers in the U.S. and elsewhere. These developments (increasing energy demand and increasing use of natural gas and coal) are expected to <u>increase</u> the amount of carbon emitted to the atmosphere from the world's electrical power plants by about 70% over the next 20 years [1].

Nuclear energy is the only fully developed technology able to supply large amounts of electricity without generation of greenhouse gases, and therefore should be a key element in the strategy to control greenhouse gas emissions. However, several problems cloud the future of nuclear power in the U.S. and need to be addressed for nuclear power to be a preferred electric power generation option. President Clinton's Committee of Advisors on Science and Technology (PCAST) [2] recently recommended that an enhanced national R&D effort is needed to improve energy technologies. Regarding nuclear power, PCAST stated that that "the potential benefits of an expanded contribution from fission in helping address the carbon dioxide challenge warrant ...[finding] ...out whether and how improved technology could alleviate the concerns that cloud this energy option's future."

The U.S. Department of Energy (DOE) has responded to the PCAST recommendations with two new programs: the Nuclear Energy Research Initiative (NERI) and the Nuclear Energy Plant Optimization (NEPO) program. The High-Efficiency Nuclear Fuel Program was developed by DOE and submitted as part of the FY 1998 budget, but did not receive funding from Congress. The authors expect that the program will compete favorably for funding under the Department's FY 1999 NERI program. The program will be a cooperative research and development program with industry to develop improved fuels that can be operated to higher burnups with greater safety margins.

The specific goals of the High-Efficiency Nuclear Fuel Program are to: (1) develop within 7 years light water reactor (LWR) fuel designs and cladding materials that can operate satisfactorily to about 25% higher burnup; (2) develop in 15 years LWR fuels that can be used about twice as long as current fuels; and (3) demonstrate that these new fuel materials and designs are superior to current LWR fuel during normal operation and during any accident that may occur. If the program is successful, use of these fuels will enable longer plant operating cycles and improved capacity factors, which would help lower operating costs; further reduce defect rates and provide improved margin during any off-normal or accident condition; reduce the amount of spent fuel that must be handled, stored, and placed in a repository; reduce the amount of low-level waste produced by the commercial nuclear power industry; reduce worker exposures; and significantly decrease the possibility of someone using LWR spent fuel for nuclear weapons material. The purpose of this paper is to discuss LWR fuel behavior during normal and design basis accident conditions and summarize the proposed research and development program. The benefits of the proposed program are discussed elsewhere [3].

#### 2. LWR FUEL PERFORMANCE AT HIGH BURNUP

LWR fuel is currently limited to (a) about 62-MWD/kgU peak rod burnup by the USNRC because of concerns about high burnup fuel integrity, and (b) less than 5% enrichment because of the design and licensing of the fuel fabrication plants and handling and storage equipment. In addition, the control rod worths and other aspects of the core neutronics designs may limit the use of significantly higher burnup fuel. The issues which must be addressed when considering the use of LWR fuel at higher burnup include:

- Loss of cladding ductility and fracture toughness due to (a) excessive corrosion, hydrogen uptake, and zirconium-hydride formation, (b) neutron radiation damage, and (c) oxide spallation and zirconium-hydride blister formation.
- Excessive cladding growth.
- Increased fuel pellet-cladding mechanical interactions (PCMI) due to cladding creepdown, fuel swelling, and fuel-cladding diffusion bonding.
- Reduced fuel thermal conductivities and increased fuel temperatures due to plutonium and fission product buildup near the surfaces of the fuel pellets and the resulting formation of a porous rim.

- Increased fuel rod internal pressures due to the long irradiation times (more time for diffusion), larger inventory of fission products, and higher fuel temperatures.
- Runaway cladding oxidation.

Zircaloy was originally chosen for cladding the fuel in nearly all LWRs because of its low neutron cross section and relatively good corrosion resistance. However, thick oxide layers are often found on zircaloy-clad fuel rods, especially pressurized water reactor (PWR) fuel rods, irradiated to burnups of 50 to 60 MWD/kgU. A cross section of a zircaloy clad PWR fuel rod irradiated to about 60 MWD/kgU is shown in FIG. 1. The oxide layer thickness was about 150  $\mu$ m before the oxide began to spall off. The relatively lower temperatures at the locations where the oxide spalled off resulted in hydrogen diffusion down the temperature gradient and the formation of blisters of zirconium-hydride. The remainder of the cladding wall thickness contains numerous zirconium-hydride platelets. There is very little ductility or toughness left in such material.

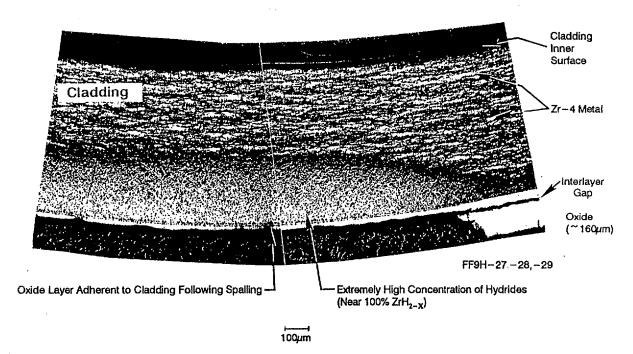


FIG. 1 Zirconium hydride concentration resulting from operation with spalled oxide.

In addition to the effects of corrosion, the ductility of zircaloy cladding is significantly reduced by neutron radiation. For example, the total plastic elongation at burst of zircaloy tubes irradiated to fast fluences above  $10 \times 10^{21}$  n/cm<sup>2</sup> (E>1Mev) is sometimes as low as 1/2 to 1% (compared to 15 to 20% for unirradiated material).- values which might result in cladding failure during modest power increases. Other problems with the use of zircaloy as cladding and structural material in LWRs include (a) excessive thimble growth resulting in bowing which restricts full insertion of control rods and (b) runaway cladding oxidation likely due, in part, to poor water chemistry.

PCMI failures have also occurred in some LWRs, especially boiling water reactors (BWRs) where control rod movement results in a significant power change in nearby fuel rods. Higher burnup will result in additional cladding creepdown, fuel swelling, and fuel-cladding diffusion bonding - phenomena which should result in more-severe PCMI during power changes.

Fortunately, the international nuclear fuel vendors have been developing and testing advanced fuel cladding and structural materials for, in some cases, over 20 years. The result

has been new products which appear to be much more resistant to corrosion and hydrogen uptake and PCMI than standard zircaloy. FIG. 2 is a plot of oxide thickness versus burnup for a variety of alternative Westinghouse fuel rod cladding materials [4]. The low-tin ZIRLO material exhibits about 1/4 the corrosion of standard zircaloy [5]. Other fuel vendors have also developed new cladding materials (zirconium alloys) which show promise for use at higher burnup [6-11]. Some of these materials not only have much better corrosion resistance, but exhibit less growth and creep. Cladding liners have also been developed for BWR fuel rods to help protect against PCMI failure.

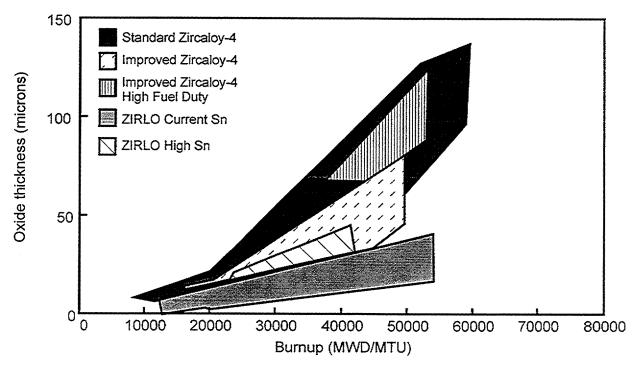


FIG. 2 Oxide thickness versus burnup for alternative Westinghouse fuel rod cladding materials [4].

There has been much less development of the LWR fuel form over the years, which is primarily pressed and sintered  $UO_2$ . There has been some minor changes in the pellet diameter-to-length ratios, the dish shapes, amount of chamfer, fuel density, and fuel grain size, primarily to minimize PCMI and fuel densification and gas release. Some LWR fuel rods have plenums on both ends of the rods, some have a top plenum only, and various end plug shapes are used. However, more work could be done to develop fuel designs which better retain the fission products within the fuel, have a more uniform rod internal pressure, and minimize fuel-cladding mechanical interactions.

#### 3. FUEL PERFORMANCE UNDER TRANSIENT CONDITIONS

If a fuel type designed for high-burnup operation is to be utilized, then the performance of that fuel under postulated accident scenarios must meet relevant regulatory criteria. Two important classes of design-basis accidents for LWRs in the U.S. are the reactivity-initiated accident (RIA) and the loss-of-coolant accident (LOCA). Recent evidence indicates that the evolution of characteristics and properties of LWR fuel and cladding materials during extended irradiation may degrade the ability of fuel rods to withstand failure under RIA and LOCA conditions; e.g., [12]. Therefore, the proposed program includes provision for assessing and demonstrating the RIA and LOCA behavior of the fuel designs developed in this program. The following sections address considerations relevant to each accident class.

#### 3.1 Reactivity-initiated accidents

Two principal regulatory criteria are used in the U.S. to assess safety under postulated RIAs [12]. To ensure core coolability after an RIA and to preclude energetic dispersal of fuel particles into the coolant, the peak fuel-rod enthalpy is limited to 1170 kJ/kg fuel (280 cal/g fuel). To allow calculations of radiological releases, other values are used to indicate cladding failure: critical heat flux values related to departure from nucleate boiling or, for low-power accidents in BWRs, 711 kJ/kg fuel (170 cal/g fuel). These regulatory criteria were established using test data from fresh fuel or from fuel with relatively low burnup (i.e.,  $\leq 5$  MWD/kgU). In Japan, the cladding failure criterion is more conservatively set at 356 kJ/kg fuel (85 cal/g fuel) for irradiated fuel, reflecting current knowledge that the RIA failure resistance of some fuel designs is significantly degraded with burnup [13].

Results of recent simulated RIA tests of higher-burnup fuel rods indicate that fuel failure is possible at fuel enthalpy values considerably lower than the U.S. Nuclear Regulatory Commission (USNRC) criteria values. Therefore, the issue continues to receive considerable international attention [14]. Initially, the consequences of low-enthalpy failures were thought to be sufficiently minimal as to pose no concern for public health and safety [12]; however, further evaluation of the observed test fuel failures has indicated that fuel dispersal from low-enthalpy failures may raise questions regarding core coolability. For example, personnel from the USNRC are now considering establishment of a new criterion of 418 kJ/kg fuel (100 cal/g fuel) as both a cladding failure threshold and a fuel dispersal threshold [15]; additional testing would be required to establish a coolability criterion above the cladding failure threshold. Ongoing test programs at the CABRI facility in France and the Nuclear Safety Research Reactor (NSRR) in Japan are intended to determine the conditions and phenomena that lead to RIA fuel failure and to assess post-failure fuel dispersal. Although the programs at each location have included tests of several rods which did not result in failure, it is instructive to review the parameters and conditions for the tests which did result in fuel failure. A summary of information from those tests is provided in Table I.

		Fuel	Cladding		Peak Fuel			
		Burnup	Oxide	Pulse Width @	Enthalpy at	Fuel		
	Fuel	MWD	Thickness	FWHM	Failure	Dispersal		
Test ID	Туре	kgU	(µm)	(ms)	(kJ/kg fuel)			
NSRR Tests [12,13,16]:								
HBO-1	$UO_2$	50	43	4.4	250	yes		
	-				(60 cal/g)			
HBO-5	$UO_2$	44	103	4.4	301	small		
	2				(72 cal/g)			
TK-2	UO <sub>2</sub>	48	23	4.4	250	small		
	4				(60 cal/g)			
CABRI Tests [17,18]:								
REP-Na1	UO <sub>2</sub>	64	80	9.5	125	yes, 6g fuel		
	-				(30 cal/g)			
REP-Na7	MOX	55	50	40	501	TBD		
					(120 cal/g)			
REP-Na8	$UO_2$	60	130	80	347	No		
	- 2				(83 cal/g)			
					<u>``</u>			

TABLE I. Summary of Information from Selected Recent RIA Tests Resulting in Failed Fuel

The conclusions emerging from the ongoing RIA testing programs in France and Japan indicate that failure of high-burnup fuel rods is induced by PCMI, exacerbated by the condition of the fuel and cladding after high-burnup, steady-state irradiation [12]. Temperaturedriven expansion of fission gas in the oxide fuel matrix is more pronounced with increasing burnup. and provides a driving force for radial expansion of the fuel as the fuel temperature increases (in addition to the intrinsic thermal expansion of the fuel). During an RIA transient, the radial expansion of the fuel places a hoop stress on the cladding. If the cladding has experienced significant degradation due to corrosion, as is often the case with zircaloy cladding materials, then the ability of the cladding to withstand the PCMI-induced stresses will be reduced. Specific cladding degradation phenomena include cladding oxidation and/or spallation, which effectively reduces the thickness of the cladding wall available to withstand the PCMI loading, and embrittlement due to hydrogen uptake and irradiation damage. Uptake of hydrogen into the cladding and movement of hydrogen down the temperature gradient to form zirconium hydride precipitates within the cladding wall is considered to be a key phenomenon leading to some of the failures listed in the table above. Development and implementation of cladding alloys that resist corrosion, and radiation embrittlement at burnup values well above 62 MWD/kgU, will be important to licensing fuel designs intended for ultra-high burnup.

The decreased grain size and locally higher fission content that forms near the periphery of a fuel pellet during prolonged irradiation (i.e., the rim effect) also facilitates fragmentation of fuel debris during a transient. Fuel grain decohesion occurs as fission gas bubbles at the grain boundaries (a characteristic of the rim structure) pressurize with increasing temperature and expand, resulting in loose particles or fragments of fuel material. Fragmented fuel debris can be released through a cladding breach into the coolant where, depending on the degree of fuel release, coolant channel blockage can be a concern. Fuel designs that avoid formation of this rim structure, or that otherwise mitigate fuel fragmentation are desirable for achieving ultra-high burnup reliability.

Although the failures of the test fuel rods listed in Table I are considered to be induced by PCMI, other phenomena can contribute to failure at different conditions. For example, if a fuel rod withstands a RIA to fuel enthalpy values higher than those indicated in the table, then failure due to internal fission gas pressurization may occur. Because fuel irradiated to high burnup retains a correspondingly higher fission gas inventory, an increase in temperature can lead to release of an amount of fission gas into the fuel rod plenum that is sufficient to balloon the cladding. Furthermore, as cladding temperatures increase during a RIA transient, the yield or ultimate strength of the cladding material decreases, perhaps to the point of failure under internal pressurization or PCMI.

RIA variables that most affect fuel failure during a RIA are cladding temperature and fuel temperature (which affects fuel expansion and/or fission gas release). The reactor parameters that control these variables are power pulse width, typically expressed as the fullwidth at half-maximum (FWHM) value of reactor power as a function of time, and energy deposition into the fuel, typically manifested as fuel enthalpy rise expressed in units of calories per gram of fuel. Time-dependent cladding and fuel temperatures are also determined by coolant flow conditions.

Three-dimensional neutronics and thermal-hydraulics calculations of the response of a LWR core to a RIA generally predict energy insertions of less than 418 kJ/kg (100 cal/g) in any fuel rod and power pulse FWHM values ranging from 70 to 100 msec, although one recent calculation resulted in 30 to 80-msec power pulses [19]. Most RIA tests of high-burnup fuel have been performed with excessively-narrow power pulses (<10 msec FWHM), although the more recent tests in CABRI have been performed with a power pulse that approximates an RIA pulse in a PWR. Narrow power pulses deposit energy into the fuel in a non-prototypic manner, exacerbating fuel-cladding mechanical interactions that may lead to failure. More specifically, narrow power pulses 1) induce fuel pellet expansion at a faster rate, leading to a higher PCMI-induced strain rate in the cladding, and 2) induce relatively higher temperatures in the rim region of the fuel pellet (due to

energy deposition occurring at a faster rate than heat transfer mechanisms can remove energy), intensifying the expansion phenomena that cause PCMI and thereby raising the peak hoop stress in the cladding. Furthermore, the peak stress on the cladding occurs before the cladding can heat up to temperatures at which the cladding will more easily yield in a ductile manner; thus, brittle failure may be artificially induced. These tests have also been performed with non-prototypic coolant conditions, using sealed capsules containing stagnant air or water coolant or using loops with flowing sodium, leading to non-prototypic pre-test temperatures and pressure. Furthermore, the fuel rods that have failed in those tests were clad with standard zircaloy alloys; these alloys exhibit much more severe oxidation and hydriding behavior (which render a fuel rod more susceptible to failure by fuel-cladding mechanical interaction) than do the improved zirconium-based alloys that are in current use in many nuclear power plants (NPPs).

The tests performed to date have been suitable for the stated objectives of those test programs, which is to study PCMI during the early stages of the transient [17], and the results reported from those test programs thus far have provided tremendous insight into the mechanism of PCMI-induced failure. However, any additional tests to be performed for determining the RIA behavior of high-burnup fuel should more closely simulate postulated RIA events, using 30 to 100-msec power pulse widths, inducing peak fuel enthalpies of 83.7 to 837 kJ/kg fuel (20 to 200 cal/g), employing prototypic coolant conditions and test rods with the improved cladding alloys in current use. A prototypic fuel temperature during a test is ensured by a prototypic power distribution through the fuel (which is determined by the duration and magnitude of the power pulse and by the neutron spectrum) and by prototypic coolant conditions. Coolant flow conditions during a test are best provided by a water loop that supplies flow to the test fuel at prototypic temperatures and flow rates.

#### 3.2 Loss-of-coolant accidents

The U.S. regulatory criteria pertaining to LOCAs are intended to ensure core coolability through the duration of and beyond the accident. Specific criteria include a limit on the maximum cladding temperature attained during a LOCA of 1204°C and a limit to the cladding oxidation corresponding to a 17% cladding wall thickness reduction. Furthermore, the core must remain coolable after the accident, which implies that failed fuel debris cannot cause an unacceptable reduction or blockage of the coolant flow.

The behavior of LWR fuel during a LOCA is influenced by somewhat different factors than those that influence LWR fuel behavior during an RIA. The sudden depressurization that occurs during the large break LOCA combined with a degraded heat transfer associated with the loss of coolant is expected to cause cladding ballooning and burst. The magnitude of the ballooning will be influenced by the absolute temperature (zircaloy is more ductile in either its alpha or beta phase and significantly less ductile when changing phases), the rod internal pressure, and the temperature distribution around and along the rod. Because of cladding creepdown, fuel swelling, and fuel-cladding diffusion bonding, high burnup tends to promote large, long balloons during a LOCA which result in more flow blockage that occurs with low burnup or fresh fuel. In addition, the choice of alloying elements can influence the cladding ballooning behavior.

The core coolability is also influenced by the cladding oxidation and embrittlement. There is some evidence that zircaloy cladding with a heavy load of hydrogen may oxidize much faster during a LOCA than normal zircaloy. Furthermore, fuel material from the outer periphery (or rim) can fragment due to decohesion induced by thermal stresses during fuel quenching, which occurs as liquid coolant is re-contacted with the fuel. As with the RIA mechanisms described, fuel fragmentation may release fuel material into the coolant, leading to a coolant flow blockage if the release is sufficiently severe. Further study of fuel and cladding properties is required to understand the implications of high-burnup irradiation on fuel behavior during LOCAs. The cladding ballooning and rupture and fuel fragmentation characteristics for high-burnup fuel with various cladding materials must be assessed to determine the conditions at which failure is expected and the consequences of failure under specified conditions. The oxidation behavior of irradiated cladding must be assessed under conditions similar to those encountered in LOCAs. Development of a modeling capability to predict fuel behavior under transients (either LOCAs or RIAs) will require more complete mechanical properties data for irradiated material at relevant stress and strain conditions.

A joint program funded by the USNRC and the Electric Power Research Institute (EPRI), with participation of the U.S. DOE, has been initiated to address the needs described above [20]. The study currently is addressing a limited number of fuel designs and cladding types with a burnup limit of 60 MWD/kgU. The program, which is being conducted in a hot cell, is comprised of two major tasks. The first task consists of engineering tests of fuel rods and cladding under LOCA conditions. These tests will determine the kinetics of oxidation of selected zircaloy cladding alloys, determine the integral LOCA behavior of fuel rod segments, and tests to determine rod stiffness and ductility for assessing resistance to seismic loading during and after a LOCA event. The second task will determine the post-irradiation mechanical properties of selected, current cladding alloys; emphasis will be placed on mechanical property testing at stress and strain conditions relevant to both LOCAs and RIAs. The program discussed below will be complementary to and coordinated with the USNRC program.

#### 4. PROGRAM DESCRIPTION AND PRODUCTS

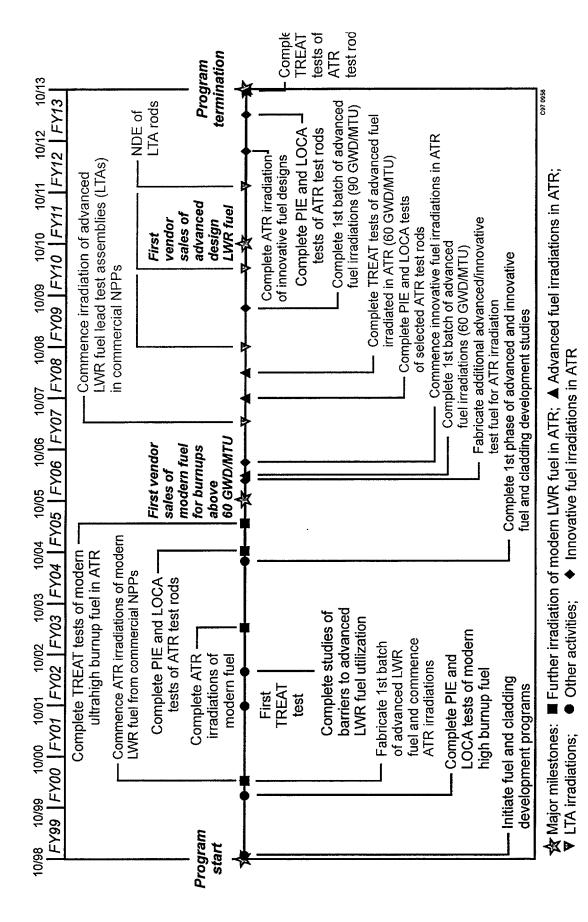
The High Efficiency Nuclear Fuel Program will include a three-part approach:

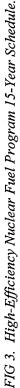
- Determine the useful life of the best fuel currently in commercial NPPs
- Develop a better fundamental understanding of the life-limiting degradation mechanisms at high burnup
- Design and test advanced and innovative LWR fuel forms.

A preliminary program schedule, which shows the relative timing of the major activities, is presented in FIG 3.

The research with the commercial spent fuel will include characterization of the condition of modern fuels that have been burned in commercial NPPs to the current USNRC limits, further irradiation of these fuels in a test reactor to ultra-high burnups, and design-basis accident testing to demonstrate that this fuel will meet USNRC licensing criteria. Further irradiation and power ramp testing of PWR fuel designed and fabricated by the U.S. fuel vendors will start 12 to 18 months into the program and will be performed primarily in the Advanced Test Reactor (ATR) in Idaho. PWR conditions can be exactly reproduced in reasonable-size loops in the ATR, and fuel previously irradiated in a commercial reactor can be safely driven to much higher burnup in the ATR and then power-ramp tested. Lead-use assembly (LUA) irradiations of BWR rods will be conducted in selected commercial NPPs, and power ramp testing of BWR fuel may also be done in the ATR. The design-basis accident testing of the ultrahigh-burnup material from the ATR and LUAs will be done in the Transient Reactor Test Facility (TREAT) in Idaho and in the Argonne and Idaho National Engineering and Environmental Laboratory (INEEL) hot cells. Both loss-of-coolant (LOCA) and reactivity-initiated accident (RIA) tests will be conducted. A complete set of results, including appropriate computer models, should be available in about 6 to 7 years.

The development of advanced fuels will be accomplished primarily by the commercial fuel vendors in collaboration with fuel experts from DOE's laboratories. This fuel will also be irradiated in the ATR. The ATR irradiations will continue for about 4.5 years and include





power ramp tests at appropriate intervals. Some of the fuel will then be removed and examined, tested in TREAT, and further examined in the Argonne and INEEL hot cells. The remainder of the advanced and innovative fuel designs will continue irradiation in the ATR to ultrahigh-burnups (about 100 MWD/kgU) and then be tested in TREAT and the hot cells. Irradiation of advanced LWR fuel LUAs in commercial NPPs will begin after about 7 or 8 years of development and testing.

In parallel with the irradiation of the advanced design fuels in the test reactors, laboratory research to study the metallurgical and environmental factors that affect the degradation (corrosion and radiation hardening) of the fuel cladding will be conducted. Analysis to evaluate fuel rod designs that better retain the fission products within the fuel, have a more uniform rod internal pressure, and minimize fuel-cladding mechanical interactions will also be conducted.

The products available for use in the commercial nuclear power industry will include:

- 1. A thorough evaluation of the useful life of the best fuel currently being sold by the participating fuel vendors at the end of 7 years. We expect that a number (maybe all) of the latest product lines can be used at burnups above the current USNRC limit of 62 MWD/kgU.
- 2. Development and initial testing of advanced ultra-high burnup fuel designs, sufficient for the vendors to start selling lead fuel assemblies at the end of 7 or 8 years.
- 3. A well-documented, physical understanding of the metallurgical and environmental factors that affect cladding (and assembly structural material) degradation. Improved fuel and fuel assembly predictive models and a solid technical basis for the sale and licensing of advanced fuels will be developed at the end of 15 years.

#### 5. TEST FACILITIES

We assume that data will be needed to license and use LWR fuel to higher burnups from the following types of tests: steady state irradiations, power ramp tests, and designbasis accident tests, primarily LOCA and RIA tests. The irradiations and tests will be designed to demonstrate reliability and, where necessary, determine failure behavior and thresholds. Hot cell examinations before and after testing will document the cladding, fuel, and bundle performance and provide data for modeling. Use of DOE's test facilities are critical to obtain timely and fully representative results, enabling the program to have its maximum benefit. The two key test facilities and the planned transient tests are discussed in the following subsections.

#### 5.1 Extended Burnup Irradiation in the Advanced Test Reactor.

Irradiation in the ATR offers the quickest means to extend burnup beyond the values allowed in a commercial reactor. The ATR core has a serpentine fuel arrangement, which provides nine flux traps. The flux traps in the corner lobes of the serpentine arrangement are almost entirely surrounded by fuel, which allows the power in a single lobe to be adjusted somewhat independently of the rest of the core. The power in a lobe at full reactor power can be maintained at levels of 17 to 60 MW. A pressurized water loop will be installed into the flux trap in the northeast lobe of the ATR, which will be designed to operate at standard PWR conditions ( $327^{\circ}$ C, 15.5 Mpa). The irradiation test space will be approximately 4 inches in diameter, sufficient to accommodate a bundle with  $32 \ 17 \times 17$  PWR-type rods (i.e., a  $6 \times 6$  array without corner positions). A proposed fuel rod arrangement for testing is shown in cross section in FIG. 4, which also shows typical linear power values (in kW/ft) for high-burnup (i.e., >60-MWD/kgU) LWR fuel rods. The proposed arrangement includes two positions for irradiation of PWR guide tubes of advanced design or material composition, which could be instrumented, if necessary, to help verify irradiation conditions in the test bundle.

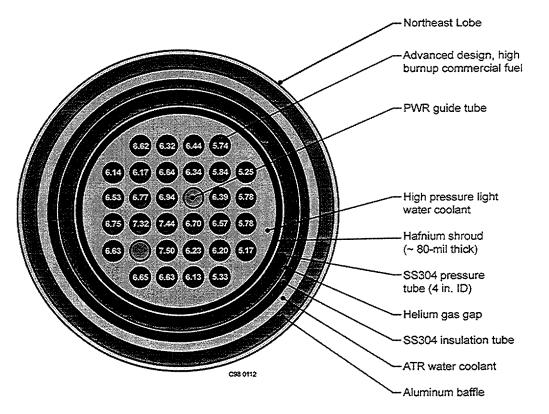


FIG. 4 ATR test bundle cross section. The powers shown were arbitrarily set at a peak of 7.5 kw/ft.

Particular attention has been given to configuring the proposed irradiation vehicle to provide conditions similar to those in a PWR. The loop will be equipped with a pressurizer and purification and makeup water systems, which will allow real-time control of water chemistry, including prototypic, time-dependent variation of boric acid, and lithium hydroxide concentrations through a cycle. The hafnium shroud shown in the cross section of FIG. 4 is included to reduce the total neutron flux in the test bundle to values typical of PWRs, thus allowing prototypic linear power values. As shown in FIG. 5, calculations thus far have shown the neutron energy spectrum in the ATR test bundle to be very similar to that of a PWR. Further work is being performed to tailor the beta and gamma flux in the ATR bundle to that of PWRs, to ensure that the corrosion behavior exhibited by the test rods is the same as expected in a PWR environment.

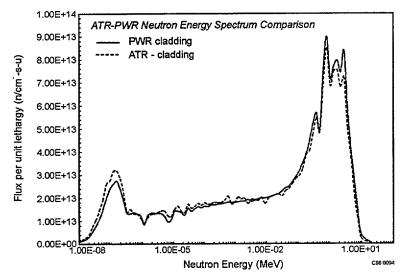


FIG. 5. Comparison of the neutron energy spectrum in typical high-power, four-loop PWR fuel cladding and in the ATR test bundle fuel cladding.

#### 5.2 Transient Tests.

It is anticipated that properly simulated RIA tests of the ultrahigh-burnup fuel developed for this program will be necessary for licensing. Because it is particularly important to perform the RIA tests at prototypic conditions, use of TREAT is proposed. A description of TREAT and a discussion of its suitability for doing this work appear elsewhere [21]. The TREAT RIA tests will be a series of 2- or 3-rod tests at increasing energy insertion until the fuel rod failure threshold is identified. We expect failure of high-burnup (60-MWD/kgU) LWR fuel, with the newer cladding materials and modest zirconium oxide layers, at energy (enthalpy) insertions above that now calculated to be possible in a LWR (i.e., at 150 to 200 cal/g fuel). It is more difficult to predict the failure thresholds for 90-MWD/kgU fuel. But, it is likely that if the new designs and materials perform as well during normal operation as we expect, the failure thresholds during an RIA may still be above what is possible in a LWR core.

The expected outcome of a LOCA strongly depends on flow blockage and cladding embrittlement. Regulatory limits include the 10 CFR 50.46 cladding embrittlement criteria and the Appendix K evaluation models (or best-estimate substitutions). There are also requirements to ensure control rod insertion. Our objective is to show that any flow blockage and cladding embrittlement during an Appendix K LOCA in a modern high-burnup core will be essentially the same as, or less severe than, in a low-burnup core. Therefore, we will conduct oxidation studies and quenching, structural response, and biaxial burst tests. This work can be conducted in hot cells and will be similar to the work the USNRC, EPRI, and DOE are currently sponsoring with older design fuel [20].

#### 6. SUMMARY

Although reemergence of nuclear power as a preferred option for new electric energy generation in the U.S. is a long-term prospect, the problems of global warming, coupled with the uncertain outlook for alternative energy sources and certain global population and energy growth, make the maintenance of the nuclear option in the United States a necessary objective. The proposed High Efficiency Nuclear Fuel Program is part of a larger effort by the DOE to address the problems associated with nuclear power (and identified by PCAST and others) of noncompetitive economics, spent fuel disposal, safety, and nuclear weapons material proliferation. Other DOE programs will address the issues associated with aging management and license renewal of the current plants, generation optimization for the current plants, advanced proliferation-resistant power technologies, etc. The High Efficiency Nuclear Fuel Program is an important part of this overall program and addresses all four of the key problem areas through the development of fuel designs capable of reliable performance to burnups in excess of 60 MWD/kgU.

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#### CANDU FUEL-CYCLE VISION

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#### Abstract

The fuel-cycle path chosen by a particular country will depend on a range of local and global factors. The CANDU<sup>®</sup> reactor provides the fuel-cycle flexibility to enable any country to optimize its fuel-cycle strategy to suit its own needs.

AECL has developed the CANFLEX<sup>®</sup> fuel bundle as the near-term carrier of advanced fuel cycles. A demonstration irradiation of 24 CANFLEX bundles in the Point Lepreau power station, and a full-scale critical heat flux (CHF) test in water are planned in 1998, before commercial implementation of CANFLEX fuelling. CANFLEX fuel provides a reduction in peak linear element ratings, and a significant enhancement in thermalhydraulic performance.

Whereas natural uranium fuel provides many advantages, the use of slightly enriched uranium (SEU) in CANDU reactors offers even lower fuel-cycle costs and other benefits, such as uprating capability through flattening the channel power distribution across the core. Recycled uranium (RU) from reprocessing spent PWR fuel is a subset of SEU that has significant economic promise. AECL views the use of SEU/RU in the CANFLEX bundle as the first logical step from natural uranium.

High neutron economy enables the use of low-fissile fuel in CANDU reactors, which opens up a spectrum of unique fuel-cycle opportunities that exploit the synergism between CANDU reactors and LWRs. At one end of this spectrum is the use of materials from conventional reprocessing: CANDU reactors can utilize the RU directly without re-enrichment, the plutonium as conventional mixed-oxide (MOX) fuel, and the actinide waste mixed with plutonium in an inert-matrix carrier. At the other end of the spectrum is the DUPIC cycle, employing only thermal-mechanical processes to convert spent LWR fuel into CANDU fuel, with no purposeful separation of isotopes from the fuel, and possessing a high degree of proliferation resistance. Between these two extremes are other advanced recycling options that offer particular advantages in exploiting the CANDU reactor's high neutron economy to reuse spent LWR fuel without the need to separate, then enrich the contained fissile material.

Thorium can provide a significant extension to uranium resources in the longer term. It is of shorter-term interest in those countries possessing extensive thorium resources, but lacking indigenous uranium reserves. The once-through thorium (OTT) cycle provides a bridge between current uranium-based fuel cycles, and a thorium fuel cycle based on recycle of <sup>233</sup>U. The optimal OTT cycle is economical today, in terms both of money and uranium resources. This cycle creates a mine of valuable <sup>233</sup>U, safeguarded in the spent fuel, for future recovery predicated by economic or resource considerations. AECL has recently devised practical OTT strategies.

#### 1. INTRODUCTION

The IAEA-sponsored International Symposium on "Nuclear Fuel Cycle and Reactor Strategies: Adjusting to New Realities" identified the factors influencing the choice of fuel-cycle strategy, and development requirements and directions [1]. The fuel-cycle path chosen by a particular country or utility will depend on many local and global factors, a few of which are short-and long-term availability,

<sup>&</sup>lt;sup>®</sup> CANDU and CANFLEX are registered trademarks of Atomic Energy of Canada Limited (AECL).

cost, security, and diversity of energy resources; the state of industrial development; availability and cost of fuel-cycle technologies both domestically and off-shore (such as enrichment, and reprocessing); backend considerations, including total inventories of spent fuel and high-level waste requiring permanent disposal, environmental impact, availability, cost, and public acceptance of permanent disposal facilities; government policy on energy and industrial development; and non-proliferation considerations. Given the historical difficulty in predicting the availability and cost of energy resources and fuel-cycle technologies, and the large uncertainties and variability in many of the factors, a superior nuclear energy strategy must include <u>fuel-cycle flexibility</u>. An inherent feature of the CANDU design is its very high degree of fuel-cycle flexibility. This enables a country, or utility, to optimize its fuel-cycle strategy based on its own unique circumstances. CANDU is an evolutionary reactor, offering a custom fuel cycle to fit local requirements.

#### 2. CANDU FEATURES FACILITATING ADVANCED FUEL CYCLES

Several key features enable the CANDU reactor to meet the energy and fuel-cycle requirements far into the future. Two of these features are the channel design of the reactor, and on-power refuelling. The fuel channels are separated by relatively large amounts of heavy water. The spectrum of neutrons entering a channel is very well thermalized, and largely independent of the fuel type. On-power refuelling provides a great deal of flexibility in fuel management. Fuelling is bi-directional, meaning that adjacent fuel channels are refuelled in opposite directions. This method of fuelling results in both a flattening of the axial flux distribution, and a symmetrical axial flux distribution. The axial power distribution along the channel is mainly determined by the variation of reactivity along the channel, which itself is determined by the fuel type (particularly the initial enrichment), the fuel-management scheme, and the location of reactivity devices in the moderator (e.g., the adjuster rods). The variation of reactivity along the channel can be controlled in the simplest instance by varying the rate of refuelling; in most cases, this provides sufficient shaping of the axial power distribution, and results in similar axial power profiles for a wide variety of fuel types. The consequence is that slightly enriched uranium (SEU), mixed-oxide (MOX), thorium, and even inert-matrix fuels (containing no fertile material) can all be utilized in existing CANDU reactors. Moreover, the axial power distribution with enriched fuels peaks towards that end of the channel in which new fuel is added, and decreases along the length of the channel. For CANDU 6 and CANDU 9 reactors, in which fuelling is in the direction of coolant flow, the peak bundle power occurs towards the coolant inlet end of the channel. This axial power distribution results in higher thermalhydraulic margins than obtained with the more symmetric axial power distribution arising from natural uranium fuel, and the declining power history with burnup facilitates good fuel performance.

Ultimately, bundles can be removed from the channel during refuelling and reshuffled, and reinserted in any order. This axial shuffling provides nearly unlimited capability for shaping the axial power distribution, if necessary. Adjuster rods are located interstitially between fuel channels, in the low-pressure moderator. They flatten the power distribution with natural uranium fuel, a function not required with enriched fuel, and provide xenon-override capability. With enriched fuel, the adjuster rods can be easily replaced, if desired, or even eliminated, providing further flexibility in accommodating advanced fuel cycles.

The fuel-management scheme can also shape the radial channel power distribution across the core. With enrichment, the extra burnup potential can be traded off for increased power in the outer channels by "flattening" the channel power distribution, obtaining more power from a given sized core. Fuelmanagement flexibility also provides many options in the transition from one fuel type to another.

High neutron economy is another feature of the CANDU reactor that is key to fuel-cycle flexibility. The ability to use low-fissile fuel makes possible a <u>unique</u> synergism with LWRs, that offers the potential of fuel recycling having a high degree of proliferation resistance, using simpler and potentially cheaper technologies than conventional reprocessing. High neutron economy also means that

about double the thermal energy can be derived from burning fissile material in a CANDU reactor compared to a PWR, regardless of whether the fuel is enriched uranium, MOX, or recycled uranium (RU). High neutron economy also results in high conversion ratios, that can approach unity with the self-sufficient equilibrium thorium cycle (meaning that as much fissile material is produced as is consumed).

Most CANDU reactors have failed-fuel detection systems; on-power refuelling enables prompt removal of any failed fuel. This reduces the risk to a utility of introducing a new fuel type. An extensive array of in-core flux detectors has always been a feature of CANDU reactors, and this ensures that the flux and power distributions are well known, regardless of the fuel type and fuel-management strategy.

Finally, the basic CANDU fuel bundle design lends itself to fuel-cycle flexibility. The fuel composition can be easily varied from ring to ring. Again, with the channel design and separation of channels from each other with large volumes of heavy water, there is a sameness in the neutron spectrum entering the fuel lattice, regardless of the details of the fuel design. Hence, new fuels can be accommodated within operating reactors without changes to the fuel bundle geometry.

#### 3. NATURAL URANIUM FUEL IN CANDU REACTORS

In considering the CANDU fuel-cycle vision, it is important to understand the benefits derived from the use of natural uranium fuel to appreciate why it remains such an attractive option for CANDU owners.

The CANDU fuel bundle is relatively small (0.5 m in length, 10 cm in diameter), and easily handled (about 20 kg). It consists of only 7 distinct components (pellets, sheath, CANLUB coating inside the sheath, spacer pads, bearing pads, end-plugs and end-plates). Hence, it is an easily manufactured product that client countries have found straightforward to localize. The use of natural uranium fuel itself simplifies manufacture, handling, as well as sourcing and diversity of fuel supply.

The uranium requirements (mined uranium required per unit of electricity generated) are about 30% lower than for a PWR reactor. The use of natural uranium generates no depleted-uranium enrichment plant tails waste – in total, a more environmentally friendly front-end of the fuel cycle.

A consequence of these factors is that fuelling costs in CANDU reactors (per unit of electricity generated) are a factor of 2 lower than for PWRs [2].

After 350 reactor-years of operation, the failure rate of CANDU fuel is very low - less than 0.1% bundle failure rate. The ability to locate the infrequent defects that do occur, and to remove the failed fuel during normal on-power refuelling operations, minimizes coolant system contamination, and the economic effect of fuel defects. Reactivity mechanisms are not part of the fuel bundle assembly, again simplifying fuel manufacture, and facilitating good fuel performance. Any dissolved neutron absorber that might be used for reactivity control is confined to the moderator, precluding the possibility of precipitation onto the fuel from the coolant, and the problems that have occurred with other reactors recently.

Nor is the lower CANDU fuel burnup a disadvantage in the back-end of the fuel cycle [3]. An extensive assessment of the Canadian concept for geological disposal has just been completed, which has confirmed its technical soundness [4]. The concept is based on deep geological disposal in an underground vault located in plutonic rock. The density of fuel emplacement in such a facility is determined primarily by the heat load of the spent fuel. The higher quantity of spent natural uranium CANDU fuel, compared to higher burnup PWR fuel, is offset by its lower heat load. The simplicity and small size and weight of the CANDU bundle also reduces the cost of the emplacement system. The overall disposal cost per unit of electricity produced is similar for spent natural uranium CANDU fuel

and spent PWR fuel. This is borne out in the OECD/NEA assessment of disposal costs [5]. Also, the size of the repository is small, considering the electricity produced.

Given all of these benefits then, any new fuel or fuel cycle will need to offer <u>compelling</u> advantages before it is introduced. The rest of this paper will identify some of these advantages.

#### 4. THE CANFLEX FUEL BUNDLE

Despite the outstanding performance of existing fuel designs, the first "compelling" product that will be introduced, at least in Canada, is the new CANFLEX fuel bundle, fuelled initially with natural uranium fuel. The CANFLEX fuel bundle has been developed by AECL since 1986, and jointly with the Korea Atomic Energy Research Institute (KAERI) since 1991 [6, 7]. It is now near commercial implementation. In 1998, a demonstration irradiation of 24 bundles will be initiated in the Point Lepreau power reactor in New Brunswick, Canada. A full-scale critical heat flux (CHF) test in water will also be completed this year, which will culminate an extensive series of qualification tests and which will establish definitively the improvement in thermalhydraulic margin over the 37-element bundle.

CANFLEX is a 43-element bundle, with 2 element sizes. The increased number of elements, and element size-grading will reduce peak linear element ratings by 20% compared with those of the 37element bundle, with performance and safety benefits. This improvement applies to natural uranium, as well as to a variety of enriched fuels and burnups. Patented CHF-enhancing appendages provide the improvement in thermalhydraulic performance.

What will make the initial introduction of CANFLEX a "compelling" product is its application in plant-life management, by maintaining thermalhydraulic performance in the face of various aging phenomena.

#### 5. SEU IN CANDU REACTORS

In many countries, including Canada and Korea, we would anticipate that after the introduction of CANFLEX bundles containing natural uranium fuel, the next step in the evolution of CANDU fuel cycles would be the introduction of SEU fuel, using the CANFLEX bundle as the carrier. The 20% lower linear element ratings in CANFLEX reduce the peak operating temperatures and hence, fission-gas release, facilitating the achievement of higher burnups. Moreover, the increased thermalhydraulic margins obtained with CANFLEX fuel provide a significant performance enhancement in addition to the other benefits of enrichment.

The inherent differences in the neutronics, and the low fabrication cost of CANDU fuel, mean that the optimal enrichment that minimizes the fuel cycle cost in CANDU reactors is much lower than in a PWR: between 0.9% and 1.2%, with most of the benefits already achieved between 0.9% and 1.0%. This lower enrichment (and burnup) avoids the life-limiting phenomena that must be addressed in high-burnup LWR fuel. Enrichments around 0.9% are below the threshold at which criticality considerations result in restrictions and complications in fuel fabrication and fuel handling. Moreover, with this level of enrichment, fuel management is extremely simple: a regular 2- or 4-bundle shift, bi-directional fuelling scheme results in excellent axial power distributions, with or without the presence of the adjuster rods. Another paper in this conference summarizes the results of time-dependent fuel management simulations [8]. It is also anticipated, that at these enrichments, the transition from a natural uranium-fuelled core to an SEU-fuelled core can be achieved in a straightforward fashion, by simply replacing natural uranium fuel with SEU during the normal course of refuelling. Operational considerations are easily met with enrichment at this level, with no changes to the reactor.

Enrichments between 0.9% and 1.2% would reduce fuel-cycle costs by 20 to 30%. This cost savings is partly due to an improvement in uranium utilization: natural uranium requirements (per unit

of electricity generated) are reduced by about 25% compared to natural uranium fuel in CANDU reactors. Moreover, with enrichments in this range, spent fuel disposal costs are reduced relative to natural uranium by as much as 30% [9].

In reactors that have surplus heat removal capability, or in which this can be provided in a costeffective manner during a planned outage, SEU can be used to uprate the reactor power without increasing the limits on maximum bundle or channel power, by flattening the channel power distribution across the core. This power uprating is done by increasing the power in the outer channels (by reducing their burnup through increasing their refuelling rate). Fuel burnup is hence traded-off against higher core power. Power uprating can provide a large economic benefit to operating plants.

In new reactors, SEU provides greater flexibility in design. Using power flattening to obtain more power from a given-sized core has an advantage in capital costs over simply adding more channels to the reactor. In the SEU-fuelled CANDU 9 reactor, using enrichment of around 0.9% to flatten the channel power distribution in the core results in ~1100 MW(e) from a 480-channel, Darlington-size core, nominally rated at 935 MW(e). SEU could also be used to increase the pressure-tube thickness to extend pressure tube lifetime, or to upgrade the primary-heat-transport system (PHTS) conditions, thereby achieving higher thermodynamic efficiency. The moderator inventory could be reduced by decreasing the moderator and reflector volumes. SEU also offers greater flexibility in fuel-bundle design, providing, for example, a means of tailoring reactivity coefficients.

Finally, the use of RU from reprocessed spent LWR fuel offers access to a potentially very economical supply of enrichment at the optimal enrichment level. Previous studies with COGEMA confirmed the suitability of this material as feedstock for CANDU fuel pellets. An earlier preliminary assessment identified the potential advantages of this material in CANDU reactors, especially compared to re-enrichment in a PWR [10]. A detailed assessment of the use of RU in CANDU reactors is currently underway as part of a collaborative program between AECL, BNFL, and KAERI. If this assessment confirms the business case for RU, then the next step will be a demonstration irradiation in a power reactor with CANFLEX fuel. In this context, RU is considered to be available on the open market, and is not linked to a utility's decision to reprocess. Other papers in this conference provide further details on the use of CANFLEX with RU [11,12].

#### 6. RECYCLE OF SELF-GENERATED PLUTONIUM

Resource and economic considerations are prime drivers in decisions on recycling. The availability and cost of fissile material (starting with natural uranium), the cost of processing (enrichment), the cost of fissile material recovery (reprocessing) and the cost of fuel fabrication are all determinants in the decision to recycle.

If one considers spent fuel as a mine of fissile material, then the spent natural uranium CANDU ore is dilute. The <sup>235</sup>U concentration in the spent fuel is at the level of depleted-uranium enrichment tails (~0.2%), so there is no economic incentive for its recovery. The fissile plutonium is also dilute, typically 2.6 g fissile Pu/initial kg U. In contrast, in spent PWR fuel, depending on the initial enrichment and discharge burnup, the <sup>235</sup>U concentration is around 9 g/initial kg U, while the concentration of fissile Pu is ~ 6 g/initial kg U. Because the cost of recovery is dependent to the concentration of the fissile material (or to the amount of material that has to be processed), then clearly spent PWR fuel will be a cheaper "mine" of fissile material than spent natural uranium CANDU fuel.

Hence, the cost of its recovery does not warrant Pu-recycle from spent natural uranium in the foreseeable future. Nor would waste disposal considerations change this conclusion, as geological disposal of spent CANDU fuel has been shown to be technically and environmentally sound, and the disposal of reprocessing wastes does not have any inherent advantages over the disposal of spent CANDU fuel [3]. Moreover, in the Canadian context, it is important to establish that from any grounds – technical, economic, social, political or environmental – there is a viable and acceptable solution to the

permanent disposal of spent fuel. This will most likely require the disposal of at least some of the stockpile of spent CANDU natural uranium fuel to establish public confidence, and to address this major issue in public acceptance with nuclear power.

#### 7. CANDU/PWR SYNERGISM

As was established in the previous section, spent PWR (LWR) fuel has a higher fissile concentration than spent natural uranium CANDU fuel. CANDU is the best reactor in which to recover the energy from the recycled material: because of its good neutron economy, up to double the thermal energy can be extracted from the recycled material, whether uranium or plutonium [13]. Moveover, the high neutron economy, coupled with fuel-cycle flexibility, enable a wide range of options to be envisioned for exploiting this fuel-cycle synergism; some of these options are unique to the CANDU reactor.

At the one end of the spectrum of recycling options is conventional reprocessing, in which the uranium (RU) and plutonium are separated from one another, and from the actinide and fission product waste. We have seen that the use of as-is unenriched RU in CANDU reactors offers many attractions. This option would be complementary to the recycle of the plutonium as MOX fuel in either PWR or CANDU reactors. The economical use of MOX fuel in CANDU reactors would favour higher burnups than are optimal for SEU fuel. (The cost of MOX fuel fabrication will be independent of the amount of plutonium contained; hence, there is an incentive to maximize the MOX burnup and minimize the number of MOX bundles fabricated.) The simple fuel design and short bundle length should be advantageous in remote fabrication. High neutron economy means that roughly double the energy can be recovered from the plutonium by recycling it in CANDU reactors rather than in PWRs.

The third product from reprocessing is the actinide and fission product waste. Nowhere else is the inherent CANDU fuel-cycle flexibility more evident than in its ability to utilize this material in existing reactors [14]. Detailed fuel management simulations have confirmed the ability to use a full core of an actinide-waste/plutonium mixture in existing fuel bundle geometries. Simple fuel management schemes would be employed; refuelling rates are easily within the capability of the current systems; bundle and channel powers are within licensing limits for natural uranium fuel; and with the very high thermal conductivity of the preferred inert-matrix carrier - SiC - fuel operating temperatures are very low, just above coolant temperatures; very low fuel temperatures, and negative void reactivity result in outstanding inherent safety features. The studies, to date, have been done using the unadjusted ratio of minor-actinides-to-plutonium from PWR fuel, and no optimization has been done of the actinide mix. The reference fuel composition has 356 g plutonium, and 44 g minor-actinides (<sup>237</sup>Np, <sup>241</sup>Am, <sup>243</sup>Am) in a 37-element bundle. With this mixture, the net destruction efficiency of the total initial actinide inventory is 60%; 90% of the initial fissile plutonium inventory is destroyed. This is a longer-term fuel-cycle option, because development of the inert-matrix fuel is required. If other fuel carriers are found to be superior to SiC, then they could just as easily be used. Another paper in this conference provides an update on AECL's reactor physics and fuel studies on inert-matrix fuels [15].

At the other end of the spectrum is a group of recycling options for which the acronym "DUPIC" has been coined: <u>Direct Use of Spent PWR Fuel In CANDU [16]</u>. These options are unique to CANDU reactors, and exploit the reactor's ability to use fuel with low fissile content. Such is the neutron economy of the CANDU reactor, that the fissile content of the spent PWR fuel can be used as-is, without enhancement. Indeed, even removal of the fission products from spent PWR fuel is not required in order to achieve an appreciable burnup.

One example of direct use would be to simply cut the PWR fuel elements into CANDU length ( $\sim$ 50 cm), straighten them, then weld new end-caps to the ends. (Optionally, the elements could be double-clad.) The smaller diameter of PWR elements would enable the use of a 48- or 61-element fuel bundle, which would significantly reduce the linear element ratings compared with those of a 37-element bundle and enhance fuel performance, and would help to accommodate the variation in fissile content

between elements. Another option is the OREOX process – a thermal-mechanical process that reduces the used PWR pellets to a powder, after the cladding has been removed. The powder would be pressed and sintered as "new" CANDU pellets, and loaded into standard sheaths that would be assembled into standard bundles. The technical feasibility of this second option is the focus of a collaborative program involving the AECL, KAERI, and the US Department of State [17]. The IAEA also participates in the safeguards aspects of this program. The recent successful fabrication of 3 DUPIC elements from spent PWR fuel, by AECL and KAERI staff, is described in another paper in this conference [18].

These DUPIC recycling options offer advantages over conventional reprocessing. They all use only dry processes: no wet chemistry is involved, and indeed, there are virtually no liquids. They are simpler than conventional reprocessing, and the expectation is that they will be cheaper. While very preliminary economic analysis suggests that this is the case [19], much more technical work will be required to define the processes before more definitive costs can be established.

A major attraction of the DUPIC fuel-cycle options is that they offer a high degree of proliferation resistance. Although no fuel or fuel cycle is proliferation-proof, several features of the DUPIC processes significantly enhance its proliferation resistance:

- The proliferation barriers that are present in spent fuel are also present in the DUPIC fuel
- There is no purposeful separation of isotopes; nor can the processes be easily tampered with to effect such a separation
- The fuel processing does not involve any we chemistry; only dry thermal-mechanical processes are employed
- With no selective separation, the plutonium concentration is dilute, making it much more difficult for the removal of a significant quantity
- All stages of the process, as well as the final DUPIC fuel bundles, are highly radioactive, which would make physical access to the material, and its removal, extremely difficult
- The high radioactivity results in an easily detected "signature" of the material, making removal easy to detect
- All processing and handling must be done in a shielded facility, again making physical entry into the facility, and removal of material extremely difficult; these measures also will result in highly automated processes and the inherent abilities to track and log in-cell operations
- The processing facility is entirely self-contained: spent PWR fuel goes in at one end, and finished DUPIC fuel bundles go out the other; there is no transport of intermediate products
- Transportation of the spent PWR fuel into the DUPIC processing facility, and of DUPIC fuel to the CANDU reactor involves highly radioactive materials

Notwithstanding these inherent, self-protecting attributes of the DUPIC process and final product, safeguards measures would be provided, and built into the design of the DUPIC facility [20, 21].

The DUPIC fuel-cycle options offer the potential of significantly reducing spent fuel quantities in a system of CANDU/PWR reactors. Another paper in this conference identifies the attractions of the DUPIC fuel cycle in the Korean context [17].

A recent study by AECL has identified significant economic benefit in the cost of geological disposal with the DUPIC fuel cycle. The heat load of the spent DUPIC fuel (after it had been irradiated in CANDU reactors), is not much different from the decay heat from the original spent PWR fuel. That means that approximately 50% more energy can be derived from the PWR fuel by burning it as DUPIC fuel in CANDU reactors, with no additional penalty in heat load. Because the density of spent fuel packing in a geological repository is determined by the decay heat, this extra energy is obtained with

virtually no increase in disposal cost. As a result, the disposal cost for DUPIC fuel (in mills/kWh) is significantly lower than for either spent PWR or CANDU fuel [9].

Finally, conventional reprocessing and DUPIC define two extremes of a spectrum of PWR spent fuel recycling opportunities with CANDU reactors. Most of the space between these extremes has not even been explored. Depending on the local and international constraints and values, the optimal recycling process might lie between these two extremes. In the TANDEM cycle, the uranium and plutonium from spent PWR fuel would be co-precipitated after removal of other actinides and fission products; this step has a higher degree of proliferation resistance than conventional reprocessing, but not as high as with the DUPIC cycle, since the chemistry could be tampered to separate plutonium. A higher degree of proliferation resistance can be achieved by leaving in the highly radioactive fission products (and removing the rare-earths that affect fuel burnup); this choice would also be a much cheaper process than conventional reprocessing, with its very high decontamination factor. Looking at the OREOX process, fuel-cycle economics could be improved by selectively removing the neutron absorbing rare-earth fission products, and hence increasing the burnup of the DUPIC fuel. Several techniques could be envisioned for achieving this.

The CANDU vision is one of an evolutionary reactor, offering a custom fuel cycle to fit local requirements. Fuel-cycle flexibility and high neutron economy open the door to unique recycling opportunities having the potential of significant cost and non-proliferation benefits. Our vision is that the CANDU reactor is an indispensable part of any LWR system employing recycling, on either a national or regional basis. Moreover, our vision is that new recycling technologies will be developed to take purposeful advantage of the unique niche that the CANDU reactor can fill in spent PWR fuel recycle using processes that are simpler and cheaper than reprocessing, and designed from the start with a high degree of proliferation-resistance. The development of such technologies will require international collaboration.

#### 8. CANDU MOX FOR PLUTONIUM DISPOSITIONING

The use of reactors for burning ex-weapons plutonium as mixed-oxide (MOX) fuel converts the plutonium to a form that provides the same proliferation barriers as in spent fuel, while deriving societal benefit through the production of electricity. The use of Canadian reactors, either on their own, or to complement MOX fuel in Russian and US reactors provides an attractive, symmetrical, simultaneous drawdown of ex-weapons plutonium in those countries. The CANDU MOX option is another compelling illustration of fuel cycle flexibility, both in the inherent features of the CANDU reactor, and in the ability to tailor the fuel design to meet specific objectives [14].

The fuel for this application would be either the current 37-element bundle for a burnup of around 10 MWd/kg HE, or the CANFLEX geometry for burnups of 17 MWd/kg HE or higher. The detailed fuel design would depend on the specific objectives of the mission, and would represent a balance between the plutonium disposition rate (e.g., the speed at which the plutonium is converted to spent fuel), the energy derived from the plutonium, the net plutonium destruction efficiency (because plutonium is produced, as well as destroyed, in-reactor), and the MOX fuel fabrication capacity required for the disposition of a given amount of plutonium. The objectives are quite different from that of conventional MOX fuel using plutonium recovered from reprocessing spent PWR fuel, in which maximization of energy recovery and burnup are the major objectives. For this application, depleted uranium is used as the matrix material throughout the bundle. In the central element, and in next ring of fuel (either 6 or 7 elements, depending on whether the bundle is 37-element or CANFLEX), dysprosium is mixed with the depleted uranium (a neutron absorber is not added to the Pu-containing elements, unlike other reactors). The addition of a neutron absorber has three effects: it increases the amount of plutonium required to achieve a given burnup (and hence increases the plutonium disposition rate at the expense of the efficiency of energy production from the plutonium); it reduces the refuelling ripple (the short-term increase in local power during refuelling); and it reduces void reactivity. In all of the fuel

designs considered, void reactivity is negative, which, while not necessary, simplifies the safety and licensing analysis by eliminating any power pulse in a loss-of-coolant accident (LOCA). The plutonium is confined to the outer two rings of the bundle, the concentration ranging from 1% to 5%, depending on the burnup.

Fuel and reactor performance for all designs considered is within the current operating and safety envelopes for natural uranium fuel. Fuel management is particularly simple: conventional bidirectional, 2-bundle shifts. A full MOX core can be accommodated with no changes to the reactor (although safe and secure storage of the fresh MOX fuel would have to be provided). The plutonium disposition rate can be increased by increasing the concentration of dysprosium or by downgrading slightly the purity of the coolant and moderator. Two of the four 825 MW(e) reactors at the Bruce A station near Kincardine, Ontario, could disposition 50 te ex-weapons plutonium in 15 to 25 years, depending on the fuel design.

The CANDU MOX option would provide the participation of a trusted third country, Canada, that can provide security and safeguards assurances in a balanced, simultaneous drawdown of both US and Russian ex-weapons plutonium. CANDU MOX is a low-cost, low-risk option, readily available in the near term, which would enable a quick start to the disposition of ex-weapons plutonium. Another paper in this conference describes a collaborative program between Canada, the United States and Russia aimed at qualifying CANDU MOX fuel for this purpose [22].

#### 9. MOX FUEL EXPERIENCE IN CANDU REACTORS

AECL has more than 30 years of experience in research and development on Pu-containing MOX fuels [23]. Research activities include development of MOX fuel fabrication technology, measurement of physical properties, production of prototype fuel, experimental irradiations and post-irradiation examinations (PIE), and reactor physics measurements in the zero-power ZED-2 reactor.

The Recycle Fuel Fabrication Laboratory (RFFL) at the Chalk River Laboratories is designed to produce experimental quantities of MOX for reactor physics tests or demonstration irradiations. Following an extensive commissioning campaign using natural  $UO_2$ , a number of MOX fuel fabrication campaigns were completed in the RFFL from 1979 to 1987, producing various quantities of fuel with different compositions. After a stand-by period of about 8 years, a project to rehabilitate the RFFL and bring it back into production was completed in June 1996. MOX operations were resumed in the facility with the production of thirty-seven 37-element (U,Pu)O<sub>2</sub> bundles destined for void reactivity measurements. This campaign was completed in March 1997 [24].

Fuel performance with CANDU MOX fuel has been found to be generally equivalent to that of  $UO_2$ . Another paper in this conference describes a recent post-irradiation examination (PIE) of 4 MOX fuel bundles irradiated at high power in the NRU research reactor to a range of burnups [25].

#### **10. THORIUM FUEL CYCLES IN CANDU REACTORS**

There are several characteristics of thorium fuel and fuel cycles that make them of interest in the overall CANDU fuel-cycle vision [26]. The primary driving force is the long-term extension of nuclear fuel resources. The abundance of thorium in the earth's crust is about 3 times that of uranium. Although thorium does not contain a fissile component, <sup>233</sup>U is produced in-reactor through neutron capture in <sup>232</sup>Th, and subsequent beta-decay of <sup>233</sup>Th and <sup>233</sup>Pa. The concentration of <sup>233</sup>U in the spent fuel is about 5 times that of  $^{239}$ Pu in spent natural uranium UO<sub>2</sub> fuel. This isotope of uranium is a very valuable fissile material because of the high number of neutrons produced per neutron absorbed ( $\eta$ ) in a thermal neutron spectrum. Recycling the <sup>233</sup>U can reduce mined uranium requirements by up to 90% [27]. Complete independence from uranium is theoretically possible with the self-sufficient thorium fuel cycle, which in equilibrium, produces as much <sup>233</sup>U as is consumed. Hence, a single reactor technology

can provide both short-term and long-term assurance of fuel supply. Alternatively, high conversion-ratio CANDU reactors utilizing thorium would be synergistic with more expensive, fast-breeder reactors (FBRs), supplying the initial fissile material. Also, because commercial thorium fuel recycling facilities have not been built, there is an opportunity to develop a new, proliferation-resistant technology for recycling.

While the full exploitation of the energy potential of thorium requires recycling, which will not be economically justified for many years, the allure of using thorium in CANDU reactors is that benefit can be derived from this fuel today, in existing reactors, at fuel-cycle costs that are comparable with the already low cost of natural uranium fuelling; and with improved uranium utilization compared to natural uranium fuel. A strategic mine of <sup>233</sup>U can be produced that is safeguarded in the spent fuel, and available for future recovery and recycle when predicated by economic, technical, and strategic considerations. This possibility will be of particular interest in those countries having abundant thorium reserves, but lacking in uranium.

This bridge between the thorium recycle options of the future, and current uranium-based fuel cycles is the once-through thorium (OTT) cycle in CANDU reactors. Our analysis indicates that the optimal OTT cycle is economical today, both in terms of money and in terms of uranium resources. Two general approaches have been devised for OTT cycles in CANDU reactors. The first is a "mixed-core" approach, in which a large number of channels fuelled with "driver" fuel would provide the external source of neutrons for a fewer number of channels fuelled with ThO<sub>2</sub>. This is the conventional OTT, and theoretically, values of enrichments, burnups, and relative feed rates can be chosen that make this fuel cycle competitive (both in terms of resource utilization and in economics) compared not only with natural uranium, but also with SEU fuel [28, 29]. On-power refuelling enables the ThO<sub>2</sub> fuel to remain in the core much longer than the driver fuel. With the large disparity between the properties of the "driver" fuel and the ThO<sub>2</sub> channels, fuel management would be particularly challenging.

A "mixed-fuel bundle" approach is an alternative strategy that has recently been devised by AECL, which provides a practical means of utilizing thorium in operating CANDU reactors. Although the uranium utilization is not quite as good as in the "mixed-core" approach, this strategy has many benefits: uranium resource utilization is better than with natural uranium fuel, and fuel-cycle costs are comparable; fuel management is particularly simple; refuelling rates (in bundles per day) are a third of those with natural uranium; excellent axial power distributions are obtained, with or without adjuster rods; maximum bundle and channel powers are lower than with natural uranium fuel; and void reactivity is reduced. The "mixed-fuel bundle" contains ThO<sub>2</sub> in the central 8 elements of a CANFLEX bundle, and SEU in the outer 2 rings of elements. The disadvantage compared to the "mixed-core" approach is that separate dwell times cannot be achieved for the ThO<sub>2</sub> and the driver fuel because they are part of the same bundle. However, even with a modest bundle-average burnup of about 22 MWd/kg HE, the ThO<sub>2</sub> elements experience sufficient irradiation that they contribute positively to the overall uranium utilization. (While the overall uranium utilization is better than for natural uranium fuel, it is not quite as good as for SEU alone.) Details of these OTT fuel management studies are published for the first time in another paper in this conference [30].

Although the primary focus for interest in thorium fuel cycles in CANDU reactors is uranium resource extension, there are other benefits. The thermal conductivity of  $ThO_2$  (thoria) is about 50% higher than that of  $UO_2$  over a large temperature range, and its melting temperature is 340°C higher than that of  $UO_2$  [31, 32]. As a consequence, fuel operating temperatures will be lower, and all thermally activated processes, such as creep and fission gas diffusion will be reduced. Fission-gas release from  $ThO_2$  fabricated with proper control of microstructure will be lower than for  $UO_2$  operating under similar ratings. Thoria is chemically very stable and does not oxidize, a benefit for normal operation, postulated accidents, and waste management – both interim storage, and for geological disposal [33].

Thorium-232 produces fewer minor actinides than does <sup>238</sup>U. The resultant lower radiotoxicity of spent thorium fuel is sometimes claimed to be a benefit in waste management. However, in the Canadian

concept for engineered geological disposal, the actinides contained in used fuel are not a significant contributor to radiological risk [34, 35], and this benefit is judged to be small.

In AECL's fuel-cycle vision, thorium fuel cycles ensure long-term nuclear fuel supply using a single reactor type. In general, such cycles would not be employed until shortages of uranium resulted in significant increases in uranium price. Countries with abundant thorium reserves may elect to deploy the OTT fuel cycle earlier in CANDU reactors, to acquire experience in thorium fuel-cycle technology, and to build a strategic resource of <sup>233</sup>U safeguarded in the spent fuel, without committing to its future recovery. This would provide a low-cost insurance policy against future shortages of uranium.

#### **11. DEVELOPMENT REQUIREMENTS FOR FUTURE FUEL CYCLES**

Finally, underpinning these specific fuel-cycle developments are generic advancements in fuel design and performance that can be applied to any of these advanced fuels, including SEU, MOX or thorium. These include a generic high-burnup element design, an advanced CANLUB coating, enhanced thermalhydraulic performance, tailored reactivity coefficients, and low-temperature fuels (such as inert-matrix, or graphite-disk fuel). As well, advanced characterization techniques will help to elucidate the relationship between fuel properties and fuel performance for advanced fuels. Innovative techniques currently under development in AECL include the use of advanced techniques for measuring the diffusion coefficient of fission gases [36], and methods to accurately determine plutonium distribution in MOX fuel.

#### 12. SUMMARY: CANDU FUEL-CYCLE VISION

No single fuel-cycle path is appropriate for all countries. Many local and global factors will affect the best strategy for an individual country. Fuel-cycle <u>flexibility</u> is an important factor in an everchanging, and unpredictable world. CANDU is an evolutionary reactor, offering a custom fuel cycle to fit local requirements. Its unsurpassed fuel-cycle flexibility can accommodate the widest range of fuel-cycle options in existing CANDU stations.

In the near-term, CANFLEX bundles will be deployed with natural uranium fuel, to benefit from its enhanced thermalhydraulic performance. The use of SEU in CANFLEX bundles would be the next logical step. If the business case confirms the compelling benefits of RU, then that will be the preferred source of enrichment.

Our vision is that the CANDU reactor will be an indispensable part of any LWR system employing recycling, on either a national or regional basis. Initially, that synergism may be based on conventional reprocessing, with RU, and perhaps MOX, being recycled in CANDU reactors. Some countries will see RU-use in CANDU reactors, and eventually actinide-burning as complementary to MOX fuel in their PWRs. In some regions, there will be a strong incentive to develop advanced recycling technologies, such as DUPIC, that can take purposeful advantage of the CANDU niche in recycling spent PWR fuel using processes that are simpler and cheaper than conventional reprocessing and that are designed from the start with a high degree of proliferation resistance. The development of such technologies will require international collaboration, and international organizations, such as the IAEA, can play a role in their development.

The OTT cycle will be employed first by countries having extensive thorium reserves, but lacking indigenous uranium. Only when uranium prices are very high will thorium fuel cycles involving <sup>233</sup>U recycle be introduced. In the long term, the CANDU reactor is synergistic FBRs, with a few expensive FBRs supplying the fissile requirements of cheaper, high conversion-ratio CANDU reactors, operating on the thorium cycle.

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### **ROMANIAN NUCLEAR FUEL PROGRAM**



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### Abstract

The paper presents and comments the policy adopted in Romania for the production of CANDU-6 nuclear fuel before and after 1990.

The CANDU-6 nuclear fuel manufacturing started in Romania in December 1983. Neither AECL nor any Canadian nuclear fuel manufacturer were involved in the Romanian industrial nuclear fuel production before 1990.

After January 1990, the new created Romanian Electricity Authority (RENEL) assumed the responsibility for the Romanian Nuclear Power Program.

It was RENEL's decision to stop, in June 1990, the nuclear fuel production at the Institute for Nuclear Power Reactors (IRNE) Pitesti. This decision was justified by the Canadian specialists team findings, revealed during a general, but well enough technically founded analysis performed at IRNE in the spring of 1990.

All fuel manufactured before June 1990 was quarantined as it was considered of suspect quality. By that time more than 31,000 fuel bundles had already been manufactured. This fuel was stored for subsequent assessment. The paper explains the reasons which provoked this decision.

The paper also presents the strategy adopted by RENEL after 1990 regarding the Romanian Nuclear Fuel Program.

After a complex program done by Romanian and Canadian partners, in November 1994, AECL issued a temporary certification for the Romanian nuclear fuel plant. During the demonstration manufacturing run, as an essential milestone for the qualification of the Romanian fuel supplier for CANDU-6 reactors, 202 fuel bundles were produced. Of these fuel bundles, 66 were part of the Cernavoda NGS Unit 1 first fuel load (the balance was supplied by Zircatec Precision Industries Inc. - ZPI).

The industrial nuclear fuel fabrication re-started in Romania in January 1995 under AECL's periodical monitoring.

In December 1995, AECL issued a permanent certificate, stating the Romanian nuclear fuel plant as a qualified and authorised CANDU-6 fuel supplier.

The re-loading of the Cernavoda NGS Unit 1 started in the middle of January 1997 with fuel produced by the Romanian fuel plant.

The quality evaluation of the "pre-1990" fuel started in April 1996 and was performed by the Nuclear Fuel Plant (FCN) Pitesti, under the supervision of the Nuclear Power Group (GEN) - a distinct department of RENEL.

The paper presents the involvement of Romania in the activities related to the Advanced CANDU Fuel Cycle.

The future prospect and trend of the Romanian Nuclear Fuel Program are also presented in this paper.

#### 1. INTRODUCTION

This paper presents the two major objectives of the Romanian Nuclear Fuel Program:

- manufacturing of the standard CANDU 6 fuel;
- implementing in the future of the most appropriate for Romania of Advanced CANDU Fuel Cycle.

The first objective mentioned above of the Romanian Nuclear Fuel Program has been presented at the 5<sup>th</sup> International CANDU Fuel Conference [1].

Romania's option for the CANDU type reactors was made in the mid 60's and the first commercial discussions between the Romanian authorities and AECL began in the late 60's.

At that time, the main reasons for this choice of the Romanian authorities were related to the nuclear safety issues, the outstanding performance demonstrated by the CANDU type reactors and to the strategic advantage of the CANDU system that the nuclear fuel can be supplied locally. These reasons are still valid today.

In the 1970's Romania suffered severe floods and a serious earthquake and these facts combined with other economical shortages delayed the negotiations with the Canadian partners. In 1978 a contract for the CANDU-6 system license was signed between the Romanian authorities and AECL. From that moment on, Romania became the only Eastern and Central European country developing its nuclear power program based on the Western technology, and the first European country using the CANDU system. This situation is valid still today.

The paper presents the policy which governed the Romanian Nuclear Fuel Program before 1990. For the long term, this policy proved to be, inappropriate in several essential aspects, as it will be demonstrated below.

Several important decisions regarding the nuclear fuel production in Romania before 1990 did not prove to be well enough founded technically and economically while some proved even wrong. The political ambitions of the former Romanian nuclear authorities to develop the CANDU-6 fuel manufacturing without any Canadian support, and the approval for the start of the large scale fuel production without any AECL involvement in the qualification of the Romanian nuclear fuel plant proved, on the long term, to be a mistake which generated significant economical losses.

After 1990, the new created Romanian Electricity Authority (RENEL) assumed the responsibility for the Nuclear Power Program. As for the Nuclear Fuel Program, RENEL elaborated a realistic strategy having three major objectives:

- The upgrading, with the Canadian support, of the new organised Nuclear Fuel Plant (FCN) as a distinct subsidiary of RENEL, in order to qualify the Romanian manufacturer as a recognised CANDU-6 nuclear fuel supplier according to the Canadian Z 299.2 standard;

- The re-start of the nuclear fuel production, after the qualification of the plant, in order to meet the Cernavoda NGS Unit 1 needs;

- The quality evaluation of the fuel produced before 1990 in order to decide upon the best possible technical and economical solution for its recovery.

The paper presents the way this policy was implemented and the results it led to.

The paper presents also the activities performed in Romania in order to meet the second major objective, above mentioned, of the Romanian Nuclear Fuel Program.

The future prospects and trend of the Romanian Nuclear Fuel Program are also presented.

The paper ends with several general conclusions to be drawn out from the Romanian experience regarding the CANDU fuel manufacturing.

# 2. STANDARD CANDU-6 FUEL MANUFACTURING IN ROMANIA

In 1978 a contract for the CANDU-6 system licence was signed between the Romanian Authorities and AECL. From that moment on, Romania became the only Eastern and Central European

country developing its nuclear power program based on the Western technology, and the first European country using the CANDU system. This situation is valid still today.

#### 2.1 Romanian nuclear fuel program before 1990

It is essential to notice that as far as the nuclear fuel is concerned, the contract signed in 1978 between the Romanian Authorities and AECL had only provided the transfer to the Romanian Party of an information package that included only the Technical Specifications applicable to the CANDU-6 fuel and the Fuel Design Manual for the Cernavoda station. The contract had no provisions regarding the fuel manufacturing technology transfer and no kind of co-operation with an experienced Canadian fuel manufacturer. At that time, the former Romanian nuclear authorities believed that it was possible to successfully develop the fuel manufacturing technology without any Canadian support. On long term, this approach proved to be a mistake which generated significant economical losses, in spite of the important and dedicated efforts made by the Romanian specialists involved in the nuclear fuel program. From the beginning, our specialists opposed the approach mentioned above, but the political ambition of the former Romanian nuclear authorities prevailed. As a result, in fact, Romania had to "re-invent" the CANDU fuel manufacturing technology and this goal had to be fulfilled under the conditions of a total isolation of our country imposed by political reasons, from the world nuclear fuel industry.

The history of the nuclear fuel fabrication in Romania before 1990 is described in detail in a paper presented previously at an international CANDU fuel conference [2]. The main activities related to the development of the CANDU-6 nuclear fuel manufacturing were carried out at the former Institute for Nuclear Power Reactors (IRNE) in Pitesti.

With a few, but essential exceptions, the nuclear fuel fabrication in Romania went through a logical sequence of steps:

- Laboratory research and studies;

- Development of the our own fuel design;

- Development of a Quality Assurance (QA) system, including an Inspection and Testing Plan (ITP);

- Development of the technological processes for the nuclear fuel manufacturing;

- Designing and fabrication of the manufacturing equipment;

- Procurement from foreign suppliers of some special manufacturing equipment;

- Commissioning of a fuel pilot plant;

- Irradiation of fuel elements in research reactors, both in Romania and in foreign facilities (MZFR in Germany, BR2 in Belgium and NRU in Canada);

- Out of pile tests for fuel bundles produced in the pilot plant, tests performed in the high pressure and temperature loop built at IRNE.

The results obtained were encouraging. No abnormal results were recorded during this (though limited) testing program, both in irradiation and in out of pile tests.

To fulfil these steps, mentioned above, many financial and technical efforts were made at IRNE Pitesti. It is enough to mention here the procurement of a 14 MW Material Testing Reactor (TRIGA - type) - in operation from 1979, hot cells facilities - in operation from 1984, high pressure and temperature loop and analysis and control laboratories.

Regarding the activities described above, the following remarks should be emphasised:

- The irradiation tests consisted mainly in the well known type-tests for CANDU fuel (overpower test and power ramp test) for fuel elements. This kind of tests are aiming, mainly, at checking the fuel element design and cannot in any case be considered as a substitute for the qualification of the fuel manufacturing plant;

- No fuel bundles were tested in power reactors;

- The compatibility testing program on the fuelling machine head was not performed before 1990.

MONTH	YEAR							
	1983	1984	1985	1986	1987	1988	1989	1990
January	10	48	133	262	551	417	287 <sup>1</sup>	400
February	2	56	42	441	531	583	507	210
March	0	96	260	393	375	700	766	410
April	0	80	32	450	675	442	720	450
May	0	120	106	465	534	585	843	456
June	0	98	290	389	609	800	605	48
July	0	112	242	355	565	650	122	0
August	0	188	242	402	560	607	660	0
September	0	206	240	404	561	600	1033	0
October	0	100	253	200	597	509	809	0
November	5	176	205	347	569	208	852	0
December	65	325	464	405	485	970	134	0
Total production/year	82	1605	2509	4513	6612	7071	7338	1974
Total in stock/year	65	1577	2505	4509	6605	7071	7338	1974
Total production = $31,7$	Total production = 31,704 fuel bundles							

Total production in stock = 31,644 fuel bundles<sup>2</sup>

# <sup>(1)</sup> Depleted fuel bundles

 $\binom{2}{2}$  The difference between the total fabricated fuel and the stocked fuel bundles was used for out of pile tests

It is evident that looking back at the chain of the activities performed in order to prepare the large scale fuel production, the following links were missing:

- The fuel plant was not qualified by AECL, the design authority for CANDU system;

- The lack of co-operation with an experienced Canadian nuclear fuel manufacturer;

- The limited (if any) feed-back in the fuel production coming from the fuel behaviour showed during irradiation tests.

In spite of this evidence, the former Romanian nuclear authority (the State Committee for Nuclear Energy - abolished in January 1990) decided, without any involvement neither by AECL nor by an experienced Canadian manufacturer, to approve the start of the industrial production for CANDU-6 nuclear fuel at IRNE Pitesti. On this basis, which later did not prove to be enough founded, the industrial production of CANDU-6 fuel began at IRNE Pitesti in December 1983. Before June 1990, intensive production of CANDU-6 fuel was maintained (see Table 1). It should be noticed that the fuel production dynamics had no logical linkage with the real status of the progress in the construction of the power reactors at the Cernavoda site.

Looking back in the past, we can conclude that the policy followed in Romania regarding the Nuclear Fuel Program was not appropriate and not well enough founded technically and economically.

However, we should emphasise that in spite of this policy, impressive results were obtained and, besides, the Nuclear Fuel Program developed in Romania before 1990 was an excellent frame to grow an important number of specialists in the CANDU fuel nuclear technology. These positive aspects paid back, to a great extent, the efforts made in Romania before 1990.

Unfortunately, as it will be present below (and in detail presented in a paper at the 5<sup>th</sup> CANDU Fuel Conference [3], the quality of the fuel produced before June 1990 (more than 31,000 fuel bundles) was not good enough concerning the quality and that is why this fuel cannot be used as such in reactor.

#### 2.2. Romanian nuclear fuel program after 1990.

In January 1990 the former Romanian nuclear authority - the State Committee for Nuclear Energy was abolished and the new created Romanian Electricity Authority (RENEL) assumed the responsibility for all the energy national program (electricity and heat production). The Nuclear Power Group (GEN), as a department of RENEL, assumed the responsibility for the Romanian Nuclear Power Program.

Regarding the Nuclear Fuel Program, the first action decided by GEN was to request that AECL perform an evaluation of the fuel fabrication at the Romanian plant. The approach was focused on the technology, quality assurance system and the quality of the fuel produced before 1990. The evaluation was performed by AECL and Zircatec Precision Industries Inc.(ZPI). The findings of the Canadian specialists were presented in detail in [4], [5].

We present here the main conclusions of the Canadian specialists after their assessment performed at the Romanian fuel plant:

- Romanian fuel plant achieved impressive progress in developing the facility, training personnel and implementing manufacturing processes and inspection methods. The review found many positive features which indicate that some parts of the fuel are of good quality [4], [5];

- There were several negative findings:
- \* The Quality Assurance (QA) system in use at the Romanian plant was not adequate for nuclear fuel manufacturing [4];
- \* In Canada, Product Specifications combine the requirements of the Technical Specifications with essential information on manufacturing processes and quality assurance, and specify the levels of conformance with Technical Specifications requirements. At the Romanian fuel plant, Product Specifications did not exist before 1990;
- \* Conformance to the stated requirements was unacceptable, and in one important case (fuel element closure welds) the requirement was inappropriate. This leads to low quality of the fuel;
- \* The manufacturing processes and their control were inadequate in the assemble fuel element operation, and suspect in the braze operation;

- As regarding the fuel produced before 1990 the Canadian experts concluded that the fuel already fabricated should be considered as being of suspect quality for the in reactor use [4], [5].

On this basis and after a realistic assessment of the situation, it was the RENEL-GEN's decision to stop, in June 1990, the nuclear fuel production at IRNE Pitesti. All the nuclear fuel manufactured before June 1990 was quarantined as it was suspect for use in power reactor. As mentioned before, more than 31,000 fuel bundles had already been manufactured at that time. The already fabricated nuclear fuel was stored at Pitesti under International Atomic Energy Agency (IAEA) safeguards.

The Nuclear Power Group elaborated a strategy to be followed for the Romanian Nuclear Fuel Program. Three major objectives have been decided:

- The upgrading, with Canadian support, of the Romanian nuclear fuel plant in order to qualify it as a recognised CANDU-6 fuel supplier according to the Canadian Z 299.2 standard;

- The re-start of the nuclear fuel production of the plant, in order to meet, on a realistic basis, the Cernavoda NGS Unit 1 needs;

- The evaluation of the "pre-1990" fuel quality, aiming at identifying the best possible way for its recovery, both from technical and economical point of view.

To achieve these goals it was essential that, in the new contract signed in August 1991 between RENEL and AECL - Ansaldo Consortium (AAC) for the completion of the Cernavoda NGS Unit 1, all the necessary conditions were assured, including the nuclear fuel program. This contract included the assignment for AAC to provide assistance for the qualification of the Romanian nuclear fuel manufacturer.

Since February 1992 the nuclear fuel manufacturing has been organised as an independent RENEL's subsidiary, named Nuclear Fuel Plant (FCN).

Arrangements between GEN and AAC ended up in November 1992 with a contract for the qualification of FCN as a recognised CANDU-6 fuel supplier, giving to AECL the co-ordinating role. ZPI was selected to provide technical assistance and to supply equipment for several processes. The qualification program started in December 1993, after the Export Permit issuing by the Canadian Government.

The qualification of the plant was completed in July 1994 and a demonstration run was performed during October 1994 by producing 202 CANDU-6 fuel bundles and 66 of these fuel bundles were part of the Cernavoda NGS Unit 1 first fuel load (the balance was supplied from Canada by ZPI).

In November 1994, AECL issued a temporary certification for FCN.

The philosophy and detailed steps of the FCN qualification are described in two papers presented previously at CANDU fuel conference [6],[7].

Industrial nuclear fuel production was resumed in January 1995 under periodical AECL's monitoring.

In December 1995, AECL issued a permanent certificate, stating FCN as a qualified and authorised CANDU-6 nuclear fuel supplier. The plant capacity is of 23 bundles per day.

The evolution of the FCN fuel bundles production after the plant qualification is presented in the Table 2.

The Cernavoda NGS Unit 1 started its commercial operation on 1996 December 2. The reloading of the reactor started in the middle of January 1997 with fuel produced by FCN after 1995. Till now, 6508 bundles produced by FCN were already loaded in the Cernavoda NGS Unit 1 without any problem (no defect of the Romanian fuel was detected till now).

On the basis of the new status of the plant, Nuclear Power Group (GEN) requested FCN to perform detailed evaluation of the stock fuel quality. Once a very comprehensive feasibility study performed by the fuel plant specialists was accepted, GEN decided that this evaluation program should be implemented by FCN. The evaluation program for the assessment of the stock fuel quality started in April 1996.

# TABLE 2

YEAR	QUARTER	PRODUCTION	CUMULATIVE PRODUCTION
1994	4	202	202
1995	1	230	432
	2	316	748
	3	601	1349
	4	531	1880
1996	1	257	2137
	2	775	2912
	3	90	3002
	4	453	3455
1997	1	1088	4543
	2	1604	6147
	3	1236	7383
	4	1470	8853

The strategy adopted by FCN for the quality evaluation of the fuel produced before 1990, the results obtained and the best possible technical and economical recovery solutions are presented in detail in [3].

In December 1996, RENEL-GEN requested that AECL, as design authority for the CANDU system, perform a qualified appraisal of the evaluation program developed and implemented by FCN. The task was performed by Dr.Roman Sejnoha and the detailed findings, conclusions and recommendations are summarised in [8]. The main conclusions in [8] are that the evaluation strategy was well conceived, the effective work was well done and the data were collected and recorded in a proper manner. This assessment report also confirms the FCN solution for the best possible recovery of the stock fuel (already applied by now).

The results obtained by FCN during the quality evaluation of the stock fuel show, without any doubt, that this fuel is not suitable for in reactor use. Only the uranium contained in the stock fuel can be recovered either as pellets, as they are, or by recycling the unacceptable pellets.

By performing the complex program for the quality evaluation of the stock fuel, in a very satisfactory manner, it was demonstrated that the excellent co-operation with AECL and Zircatec, brought FCN capability to a level beyond the manufacturing activities. It should also be emphasised that the quality evaluation of the fuel produced before June 1990 was performed in parallel with the normal fuel manufacturing activity.

#### 2.3. The future of the Nuclear Fuel Program in Romania

Today, Romania has a fully integrated nuclear supply industry for its CANDU-6 type reactor. The nuclear fuel production is an essential activity for the Romanian Nuclear Power Program.

After the FCN qualification was done with AECL and ZPI support, the nuclear fuel production at the Romanian plant was very satisfactory. High quality CANDU-6 fuel was produced, appropriate relationships between the fuel plant, its suppliers and the Customer (Cernavoda NGS Unit 1) were established and the FCN personnel gained more experience and confidence in their capability.

For the future the following objectives are essential for the nuclear fuel manufacturing:

- Preserve and improve the actual FCN capability to produce high quality fuel. This means not only to keep the plant equipment in good shape or to procure more competitive equipment where necessary, but also to preserve and increase the personnel skill and responsibility that is laid upon them;

- Maintain a permanent contact with AECL, as the design authority, and with experienced CANDU-6 fuel manufacturers from abroad in order to be ready at any time to implement all the improvements which will certainly appear in the future;

- Decrease the fuel bundle cost especially by looking for better economical contracts with the fuel plant suppliers of  $U0_2$ , Zy-4 tubes and Zy-4 sheets and rods. Technology improvements, which can be developed at FCN, could also contribute to achieve this goal;

- Consider and analyse the possibilities and the advantages for both parties to establish a jointventure with a foreign experienced CANDU-6 fuel manufacturer. This could help to increase the FCN position especially in the world CANDU-6 market;

- On the short term, four to five years, FCN has to complete the recovery of the fuel produced before June 1990, on the basis of the solutions established after the quality evaluation of this fuel, evaluation which now is, practically, completed.

The nuclear fuel production at FCN should be in a strict correlation with the Romanian market demand (and possible with the international market) in order to avoid the old policy of producing fuel on stock.

With respect to the Romanian nuclear fuel market it is easy to predict the future needs. The new Romanian political authorities, in place after the November 1996 election, issued, at the beginning of this year, a Governmental decision defining the completion of the Cernavoda NGS Unit 2 as a national priority for the next five years. As a result, for this period of time, FCN has two major tasks:

- Meet the nuclear fuel needs of the Cernavoda NGS Unit 1;

- Prepare the increase of the production capacity in order to meet, starting not later than 2001, the needs for two CANDU-6 Units.

Now, RENEL is subject to a restructuring process, but regardless of the final decisions about the way this will be pursued, the future of the FCN is solid and safe. It is sure that in the future, the status of FCN will consolidate and the nuclear fuel plant will gain more flexibility and independence.

# 3.ADVANCED CANDU FUEL CYCLE- AN IMPORTANT DIRECTION OF THE ROMANIAN NUCLEAR FUEL PROGRAM

Even if from the beginning the Romanian Nuclear Fuel Program was focused on the development of the technologies for producing standard CANDU- 6 fuel to be used in the classic CANDU- 6 Cycle, in Romania, at IRNE, a special attention was paid to the Advanced Fuel Cycle for CANDU reactors.

Starting with the late 70', Romania was deeply involved in the field of the analysis related to the Advanced CANDU Fuel Cycle.

From the beginning, IRNE actively participated in the International Nuclear Fuel Cycle Evaluation - INFCE having several contracts with IAEA referring to this subject.

An important step was to develop a complex computer code - ANFEB (Analysis of Nuclear Fuel Element Behaviour) presented in its final version in [9] able to simulate the irradiation behaviour, at high burnup, of fuel elements having various characteristics such as:

-graphite discs between the fuel pellets;

-pellets with central hole;

-thicker graphite layer between pellets and sheath;

or combinations of these variants.

A special module STEBITD (Stead State Bidimensional Temperature Distribution) [10] has been developed and implemented in the ANFEB code in order to calculate the temperature in fuel elements with graphite discs between pellets. It is very interesting to find out that the results we obtained in 1978 are, practically, similar with what was reported in [11], 17 years later.

The Romanian contribution to the IAEA's INFCE Program included many studies [12-20] performed under contracts between IRNE and IAEA.

The results obtained represent an important part of the reference [9].

It is worth mentioning the activities performed at IRNE in order to develop the manufacturing technologies required by the Advanced CANDU Fuel Cycle (e.g.: "free flowing" powder) [21].

We can affirm that from its beginning the Romanian Nuclear Fuel Program has had as an important component the theoretical and practical activities related to the Advanced CANDU Fuel Cycle and this remained so after 1990 as well.

The Romanian specialists analysed all the options for the Advanced CANDU Fuel Cycle except for the DUPIC option.

These activities are also performed today, limited only by the financial aspect, as you will see from the presentation at this Technical Committee Meeting of the three papers written by my colleagues from INR (Institute for Nuclear Research)[22-24].

This papers has been selected in order to show the present Romanian directions related to the Advanced CANDU Fuel Cycle:

- Computer code development;
- CANFLEX type fuel design;
- CANFLEX type fuel bundles testing in the high temperature and pressure loop at INR (Institute for Nuclear Research);
- Reactor physics calculation for SEU (Slight Enriched Uranium) option;
- Technologies to produce fuel powder able to be suitable for high burnup.

Since January 1998, the FCN(the Romanian Nuclear Fuel Plant) has been directly involved(through ZPI- Zircatec Precision Industries Inc.) in the very promising program aiming at using the Russian weapons plutonium in the Canadian CANDU reactors [25-27]. FCN has already supplied to ZIP components such as:Zy-4 CANDU fuel sheaths-brazed appendages and CANLUB graphite coated, Zy-4 spacers, Beryllium coated, etc., to be used in this MOX project.

In the future we are decided to continue in our involvement in the activities related to the Advanced CANDU Fuel Cycle and to increase as much as possible the international co-operation with all the partners who will be interested.

#### 4. CONCLUSIONS

Romania opted for the CANDU system in the mid 60's and this option proved, on long term, to be an excellent choice.

In the 80's the Governmental program for nuclear energy was quite impressive and too ambitious, in some aspects even unrealistic. It included nuclear fuel fabrication for all five CANDU-6 reactors to be constructed at Cernavoda. However, the bilateral arrangements for the nuclear fuel manufacturing technology transfer from Canada to Romania were inconsistent with the Romanian objectives. As it is well known, the nuclear fuel manufacturing technology was developed before 1990 without any Canadian support.

In most respects, the manufacturing technology reached before 1990 an impressive level. However, as presented in this paper, several essential aspects related to the CANDU-6 fuel manufacturing were not properly solved.

The decision to start the large scale nuclear fuel fabrication in December1983 without any involvement neither by AECL nor by an experienced Canadian fuel manufacturer, proved to be a mistake which generated significant economical losses.

The nuclear fuel production in Romania before June 1990 had no logical linkage with the real status for the construction progress of the power reactors at the Cernavoda site.

Even if the policy of the Romanian Nuclear Fuel Program before 1990 was inappropriate, a remarkable experience was gained and this positive experience, paid back, to a great extent, after 1990. It is worth mentioning here that because of this experience gained before 1990, the cost paid for the qualification, by AECL and Zircatec, of FCN as a recognised CANDU-6 fuel supplier, was rather modicum.

After 1990, the new Romanian nuclear power authority, RENEL-GEN, elaborated a realistic Nuclear Fuel Program. This program went through the Romanian nuclear fuel plant qualification with the Canadian (AECL and ZPI) support, re-starting in January 1995 of the industrial nuclear fuel production, quality evaluation of the fuel produced before 1990 and starting of the recovery of this fuel.

This new policy already produced good results.

FCN is now a qualified CANDU-6 fuel supplier and by now about 99% from the fuel core of the Cernavoda NGS Unit 1 is coming from FCN production.

The future of the Romanian Nuclear Fuel Program is bright and has solid basis.

Romania was, is and will be deeply involved in the Advanced CANDU Fuel Cycle activities.

We are decided to increase the international co-operation in this field with the partners who will be interested.

As a general conclusion we can stress that it is obvious that without a political support no national nuclear program is possible, but too much political involvement in the technical aspects is not beneficial at all.

It is not a wise policy to develop all the things using only your own efforts without any international co-operation.

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# ACTIVITIES IN ARGENTINA RELATED TO THE USE OF SLIGHTLY ENRICHED URANIUM IN HEAVY WATER REACTOR NPPs

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#### Argentina

#### Abstract

An overview of activities related to the use of Slightly Enriched Uranium (SEU) fuel in HWR type NPPs, currently under execution in Argentina, is presented. The activities here described cover certain *R&D lines* as well as the main aspects of the *Project "Transition from full Natural-U to full SEU core in Atucha-1 NPP"*. Concerning the R&D lines, a summary is given on investigations related to reduction of void-coefficient using SEU fuel assemblies, annular pellet SEU fuel for bundle power flattening, etc. The main aspects of the above mentioned Project are outlined. At present, Atucha-1 core is approaching a 40. % of core load with SEU fuel, while the target of full SEU fuel core should be reached in 2-3 years. The expected exit burnup for such a core, namely 11000 MWD/tnU, is already currently obtained for SEU fuel in the present mixed core, while an increase in exit burnup of Natural-U fuel has also been obtained in very good agreement with reactor physics calculations. The comprehensive safety analysis carried out for each phase of the Project showed very moderated changes in plant behaviour under the set of postulated accidents and abnormal transients. A recent development, namely the CARA Project, aimed at unifying manufacturing of fuel assemblies for both operating NPPs in Argentina is presented in an accompanying paper.

# **1. INTRODUCTION**

The two NPPs currently under operation in Argentina are both of the HWR type, with on-power refueling and designed for Natural-U fuel in the form of UO2 pellets located in annular fuel assemblies with 37 rods. However, while the 648 MWe Embalse NPP is of the CANDU-6 type using 0.5 m long fuel bundles loaded in sets of 12 bundles within 380 horizontal pressure-tube channels, the 360 MWe Atucha-1 NPP has a pressure-vessel reactor with 252 vertical fuel channels, each one accommodating one 6.5 m long fuel assembly.

A third NPP, Atucha-2, still under construction with nearly 80. % completion, is of Siemens design as Atucha-1 and has a very similar conceptual design. Capacity is 745 MWe and the pressure-vessel reactor has 451 vertical fuel channels.

Natural-U fuel exit-burnup in Atucha-1 (6000 MWD/tnU) is lower than the equivalent for Embalse (7100 MWD/tnU), and at the same time the manufacturing cost is much higher for Atucha-1 fuel than that for Embalse, mainly due to the length and complexity of Atucha-1 fuel assemblies (See Figure 1). These factors led to a quite high impact of fuel cost on the total operating cost of Atucha-1 NPP. As a result, different lines of activities, developments and projects were initiated in Argentina since several years ago in order to try to lower that impact of Atucha-1 fuel.

Such developments and engineering activities are at present effectively giving their results through the execution of a transition in Atucha-1 reactor from a full Natural-U fuel core (as designed) to a new full 0.85 % Enriched - Uranium (SEU) fuel core, with practically no major modification to the plant hardware and no perturbation to plant availability.

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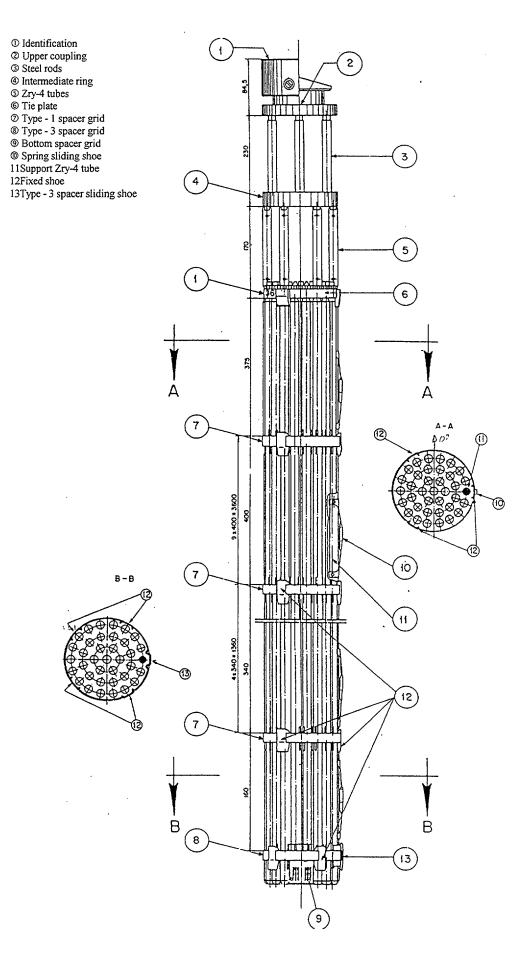


FIG. 1. General view of Atucha-1 fuel assembly.

Meanwhile, the various development lines under way could lead in the future to a possible introduction of SEU fuel also in the CANDU-type Embalse NPP. These developments include, in particular, the CARA Project aimed at unifying manufacturing of fuel assemblies for both operating NPPs in Argentina, which is presented in an accompanying paper.

#### 2. DEVELOPMENTS AROUND THE USE OF SEU FUEL

In this Section certain R&D lines related to the use of SEU fuel in Argentine NPPs are outlined. Due to the possibility of a direct transition in a HWR from a Natural-U to a SEU fuel core, the selected and standard value of U enrichment has been up to now restricted to 0.85 %. Higher values have also been studied but only in exploratory studies oriented to long term schemes.

#### 2.1 Economic Studies.

Table 1.

A wide variety of economic studies have been executed in order to evaluate different aspects. As an example, a particular study is described in this paragraph, summarizing the work presented in Ref. [1].

# Studies concerning the impact of SEU fuel as a First Core for Plant Start-up

Due to on-power refueling together with operation flexibility, switching from Natural-U to SEU fuel in HWR-type NPPs has always been considered as a transition; moreover as a very smooth transition with no specific plant shut-down for this change of fuel. This is effectively the case of operating HWR NPPs but, what is the actual situation for a new plant concerning that transition versus a direct use of SEU fuel from the very beginning ?

A new plant like, for instance, Atucha-2 could in principle be started up either with a Natural-U or with a SEU fuel core. However, the startup core, which is normally fully or mainly composed of fresh fuel assemblies, is very far from the subsequent equilibrium core which is composed of high burnup fuel assemblies. This very special situation of a HWR startup core requires a significant use of neutron absorbers in order to compensate for the strong reactivity excess : usually a liquid poison in the moderator and / or a partial load of Depleted-U fuel assemblies. This situation is unique of the startup core since, later, the equilibrium core is progressively attained. This is very different from the case of LWRs with no on-power refueling and operating under burnup/refueling cycles.

The use of SEU in the startup core of a HWR makes the above mentioned unique situation still more intense, leading to a longer transition to the equilibrium core while at the same time exit burnup is smaller. The final and global result is a practically insensitive value of the levelized KWh cost during the plant lifetime, to the use of a Natural-U or a SEU startup core. Using quite conservative assumptions and considering a full-time Natural-U core as the reference case, the levelized KWh cost shows the following reductions for Atucha-2 using a 0.85 % enrichment for SEU fuel.

CASE	KWH COST SAVING (%)
1. REFERENCE (Full-time NatU core)	
2. Full-time SEU core	27.
3. Startup core : Natural-U fuel Equilibrium : SEU fuel	25.

# 2.2 Reactor physics investigations

Different studies have been undertaken regarding the potential of SEU fuel to improve neutronic safety parameters, to improve operation performance (especially exit burnup, fuel consumption and final fuel cost) and operation flexibility (especially power limitations and in-core fuel movements). Two examples are summarized below.

# Reduction of void reactivity coefficient

A study on the reduction of the void reactivity coefficient is summarized in this paragraph taken from Ref. [2 and 3].

The normally positive void reactivity coefficient of HWRs using standard Natural-U or SEU fuel assemblies can be reduced or even turned negative through an appropriate fuel composition distribution within the annular fuel assembly, using a strong absorber like Dysprosium in the inner rod rings and SEU fuel in the outer rod rings [4, 5]. It has been established [2] that for CANDU-type fuel bundles, the combination of Depleted-U with 1. % Dy in the inner rings and 2. % Enriched-U in the two outermost rings has an almost equivalent behaviour to that of a "Uniform" 37-rod 0.85 % SEU fuel bundle in reactivity evolution with burnup.

Calling "Low Void Reactivity Bundle" (LVRB) to the former bundle and "Uniformly Enriched Fuel Bundle" (UEFB) to the last one, it is observed a higher reactivity for LVRB at very low burnup and a lower reactivity at very high burnup. However, between approx. 2000 and 12000 MWD/tnU, that is most of the usefull region, the bundle reactivity is almost identical for LVRB and UEFB.

Nevertheless, the "Low Void Reactivity Bundle" (LVRB) raises a new problem, namely, inner (radial) power peaking, whose factor changes from approx. 1.1 for the "Uniformly Enriched Fuel Bundle" to approx. 1.4 for the LVRB.

#### **Reduction of Power Peaking**

A study presented in Ref. [6] is summarized in this paragraph.

The above mentioned inner power peaking can be flattened through the use, in the outermost rod rings, of holed or annular pellets with an appropriate inner diameter. This concept can be applied either to Candu-type or Atucha-type fuel assemblies, having in both cases similar effects.

The following Table 2 shows a comparison on the Power Peaking Factor (PPF) for Embalse NPP reactor cell in the hypothesis of different fuel types. The minimum and maximum values observed for the whole burnup range of interest are quoted. The last case correspond to the use of 11.% voided pellets in the outermost rod ring, instead of normal pellets.

Table 2.

FUEL TYPE	POWER PEAKING FACTOR (Minimum - Maximum Value)			
Natural-U / standard	1.115	1.13		
0.85 % Enriched-U / standard	1.10	1.145		
0.85 % Enriched-U / 11.% voided outer pellets	1.045	1.08		

For the same three cases, calculations carried out for Atucha-1 fuel cell showed similar effects. The *maximum values* of PPF are respectively : 1.10, 1.11 and 1.04.

Concerning the exit burnup losses due to the use of holed pellets in the outermost rod ring in a Embalse-type fuel bundle, they are very small : around 250 MWD/tnU over a value of approx. 12500 MWD/tnU for Uniform 0.85 % Enriched-U fuel bundle. A very similar situation holds for Atucha-1 fuel : exit burnup losses would be around 160 MWD/tnU over a value of approx. 11000 MWD/tnU for Uniform 0.85 % Enriched-U fuel bundle.

#### Potential for a combined application

Both for Atucha and Candu types of fuel assemblies the annular configuration of fuel rods is : 1 central rod + 3 rod rings, with respectively 6, 12 and 18 rods.

This number of rings (3) is not very high for satisfactorily accommodating at the same time the LVRB concept (with a central absorber region and an external fissile region) and the holed pellet concept in the outermost or external region. Emphasis in power flattening, through a lower fissile load in the outermost ring would lead to decrease the capacity of the LVRB concept to lower the positive void reactivity coefficient.

The CARA fuel assembly design aimed at being used in both types of plants, and having 60 fuel rods instead of 37, distributed in a higher number of rings, brings much more space for an optimum combination of the LVRB concept and the holed pellet concept.

# 3. PROJECT "USE OF SEU IN ATUCHA-1 NPP" (1)

#### 3.1 Main features of Atucha-1 Core

The above mentioned 252 vertical fuel channels of Atucha-1 core form a 27.2 cm-pitch trangular lattice, inside the moderator tank. This tank is within the reactor pressure vessel, while the annulus between them forms the coolant down-comer similar to that of a PWR. Each channel is essentially an approx. 2 mm.-thick Zry-4 tube separating the inner D2O coolant (at 280.0C average temperature) and the outer D2O moderator (at 190.0C average temperature). Both fluid systems are connected outside the core and kept at the same pressure (approx. 115 ata); the channel tubes assure a colder moderator separated from the coolant in the core region.

In order to establish a quite uniform exit coolant temperature among the 252 fuel channels at the upper plenum, these channels are lumped into 8 different "Hydraulic Zones" (HZs), each HZ having a different coolant flow selected for approximately matching the radial power profile. The central HZ corresponding to the most rated fuel channels has no flow restrictors, while the 7 outer HZs have 7 different types of throttles located at the entrance of each channel (See Figure 2).

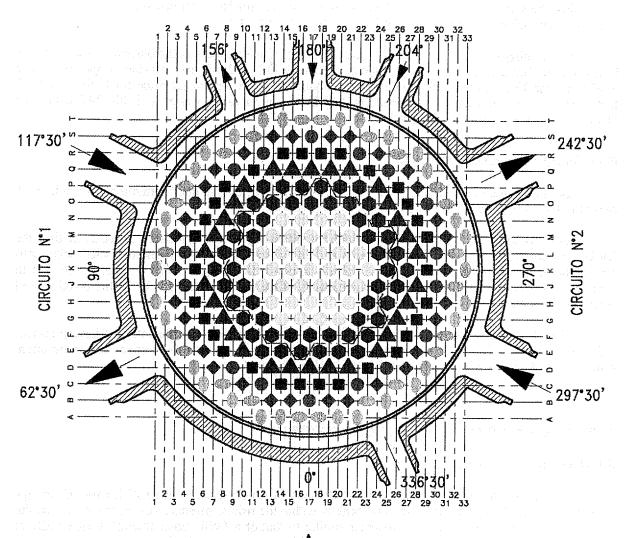
Up to the beginning of 1995, only fresh Natural-U Fuel Assemblies (NU FAs) were loaded and shuffled following a 3-position in-core path. Such a scheme defined three "Burnup Regions" (BRs). Numbering the 3 BRs from the core center to the periphery, they were :

- BR 1 : Central BR, with intermediate burnup FAs.
- BR 2 : Intermediate BR, entrance region (fresh FA loading) with low burnup FAs.
- BR 3: Outer BR, exit region (FA unloading) with high burnup FAs.

Every FA followed the path :

Fresh FA >> BR 2 (core load) >> BR 1 >> BR 3 (core unload) >> Fuel Pool

<sup>(1)</sup> The author is Technical Reviewer of the Project.



# ENTRADA MAQ. CARGA

TIPO DE ESTRAN_ GULAMIENTO	DIAMETRO EQUIVALENTE mm.	N"DE E.C.	CAUDAL min. kg/seg.(1)	CAUDAL min. kg/seg.(2)	PLANO M20
1	18,6	30	8,54	4,14	405-1
2	21,0	24	11,86	5,43	405-1
3	25,0	30	15,40	7,30	405-1
4	28,4	18	19,14	9,04	405-1
5	34,2	30	23,74	10,96	405-2
6	39,0	30	28,67	13,43	405-3
7	44,0	54	29,41	13,84	405-4
8		37	32,90	16,08	

FIG. 2. View of hydraulic zones in Atucha-1 core.

90

# 3.2 Organizational aspects

The Project "Use of SEU in Atucha-1 NPP" is at present carried out by the Utility, Nucleoelectrica Argentina S.A. (NASA), through its Engineering Div., responsible for the Project, and with a relevant participation of Atucha-1 NPP staff.

Other sectors of NASA as well as of the National Atomic Energy Commission (CNEA) provide technical support and execute engineering tasks in specific areas. Manufacturing of Atucha-1 SEU FAs is carried out (as for all other Argentine NPP FAs) by the local manufacturer CONUAR.

# 3.3 Phases of the Project

This is a Project formally divided into Phases with the purpose of initial 0.85 % Enr.-U FA incore irradiation and essay, followed by a smooth transition from the full NU FA core to a full SEU FA core. Each Phase has two parts :

- 1) Engineering tasks and elaboration of licensing documentation.
- 2) SEU FA Irradiation Program and evaluation.

At the onset of the Project in 1993, a rather quite number of Phases were foreseen, reflecting a limited knowledge on how SEU FA would work in this HWR. Main concerns were with :

- \* Atucha-1 FA behavior under a very high burnup (changing exit burnup from 6000 to 11000 MWD/tnU)
- \* Reactor control system behavior under much higher positive reactivity insertion during FA loading.
- \* Adequate selection of core channels for fresh SEU FA loading, taking into account quite thin channel power and local power margins even for fresh NU FAs.
- \* Adequate selection of in-core movement positions for irradiated SEU FA, taking into account channel power and local power limits, as well as Pellet-Cladding Interaction prevention criteria (PCI criteria).
- \* Overall core behavior, core parameter changes and core response in front of postulated accidents and abnormal transients, under the presence of SEU fuel in the core.

During 1993 and 1994 the engineering tasks of the First Phase were carried out [7] and the first SEU FA was loaded in January 1995. This was defined as follows :

<u>Phase 1:</u> Initial SEU FA irradiation and essaies. The SEU FA load is limited to a maximum of 12 over 252 FAs in the core. Minimum perturbation of core parameters.

On the basis of both engineering studies and early evaluation of results of SEU FA irradiation during Phase 1, the initial scheme of many Phases was lumped, for the rest of the Project, into only two more Phases, as follows :

- <u>Phase 2:</u> Massive SEU FA irradiation. The SEU FA load was limited to a maximum of 60 and later extended to 99 over 252 FAs in the core.
- *Phase 3*: Approach to the Full (0.85 % enriched) SEU FA core.

# 3.4 Summary of Phase 1

A presentation of main results of this Phase was given in [8].

The main outstanding aspects may be summarized as follows :

- All SEU FAs were loaded at 6 selected channels having exit-coolant-temperature thermocouple measurements but located at the outermost channels of a high coolant flow HZ in order to have the maximum possible channel power margin.
- The in-core SEU FA movement scheme was the same as that for NU FAs, though with much higher residence times in each BR and higher transition burnups.
- Excellent behavior of Atucha-1 FAs containing SEU fuel and approaching the target exit burnup for the final core. Most of the 18 SEU FAs irradiated in this Phase 1 reached exit burnups of around 10000 MWD/tnU.
- A special study was done for providing practical and more flexible PCI criteria and a new specification for in-core fuel handling involving SEU FAs was elaborated [7] and applied [8].
- A comprehensive experimental verification of predicted / calculated parameters was executed. This involved especially regulation rod bank movements and coolant exit temperature at fresh SEU FA channels, both during SEU FA loading.

# 3.5 Summary of Phase 2

The engineering activities, including all the safety studies and licensing requirements are presented in [9].

The most significant aspect of Phase 2, due to its complexity, was the determination of an appropriate in-core refuelling scheme involving a rapid replacement of fresh NU FAs to be loaded by SEU FAs. This was solved through the use of a two-way scheme for SEU and a one-way scheme for NU FA movements within the core.

Phase 2 Irradiation Program was initiated just after the planned annual outage of the plant at mid 1996. It was originally elaborated for reaching a maximum of 60 SEU FAs loaded in the core, nearly at the end 1997. However, due to planification reasons it was decided to consider an extension of the Phase up to a total load of 99 SEU FAs. For that, a more specific study [10] was elaborated on the basis of Ref. [9].

Meanwhile, this Irradiation Program has up to now proceeded in the same excellent way as Phase 1. No SEU FA failure attributed to the use of SEU fuel has been observed; FA failure rate remained very low as before the initiation of the SEU Irradiation Program. Core's operation parameters were hardly modified as expected and the safety aspects were adequately covered as mentioned below. The target average exit burnup for the equilibrium full SEU core, namely 11000 MWD/tnU, is already achieved for SEU FAs unloaded during this Phase 2, while a progressive increase in that parameter for NU FAs is also observed.

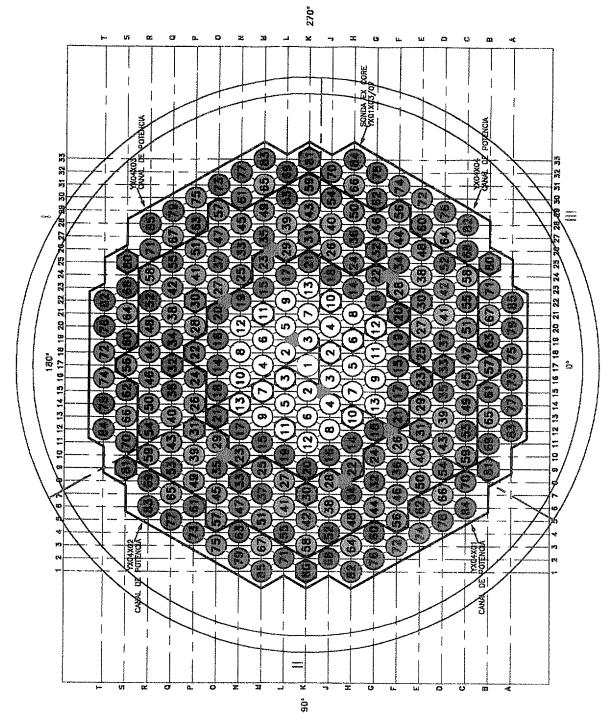
It is expected to reach the 99 SEU FA load at mid 1998, just before the planned annual outage. After that, Phase 3 would be initiated in order to accomplish the full load with SEU fuel.

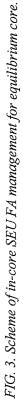
# 3.6 Some relevant aspects of the future equilibrium full SEU core

Main aspects of this future core were already calculated and analyzed. Some of these elements are included in Refs. [8,9,10].

In-core fuel management should be based in a basically 3 BR scheme quite similar to that employed with the former full NU core, using two ways (See Figure 3).

Radial power flattening associated to the use of SEU fuel, would bring larger power margins for the 2 innermost HZ (having the most rated FAs), providing more flexibility for FA shuffling and general reactor operation.





Way 2	CAMINO 2	NGRESO (5)	(9) OLISANALIO	🛞 EGRESO (7)		
Way 1	Advert	(1) INGRESO (1)	(z) UISHYUL	(E) OLISNVEI	(A) FGRESO (A)	

The predicted average exit burnup is 11100 MWD/tnU, representing a saving of approx. 46. % as compared to the former full NU core on total irradiated FAs.

Other similar or significant savings are also expected in terms of use of the fuel loading machine, fuel pool capacity needs, etc., especially savings in the current and levelized fuel costs in the total generation cost, savings situated between 30. and 35. %.

## 3.7 Safety aspects

Activities in this area encompasses the following for each Phase of the Project, according to the maximum load of SEU fuel for that phase :

- \* Determination of new reactor-physics core parameters
- \* Determination of operation systems changes under normal conditions
- \* Determination of changes in radioactive inventories and releases, operational exposure and dosis to the public through different paths and conditions under conservative assumptions.
- \* Simulation and analysis of the set of accidents and abnormal transients postulated for the previous full NU core.
- \* Identification, simulation and analysis of new postulated accidents and abnormal transients associated to the use of SEU FAs.

The most relevant fact to be outlined is the small impact of the replacement of NU by SEU as fuel on all the reactor-physics safety parameters. Such changes though generally in the less-safe sense, are so small that lead to quite insensitive results on the postulated accidents. As an example for Phase 2 the following changes are observed against the former full NU core [8,9]:

Table 3.

REACTOR PARAMETER	CHANGE (%)	
Effective delay neutron fraction $\beta$	- 1.3	<u> </u>
Average prompt neutron half-life $\Lambda$	- 1.3	
Fuel temperature reactivity-coefficient	- 3.6	
Coolant void reactivity	- 0.2	
Boron reactivity	- 1.0	

#### 3.8 Longer term prospects

The future equilibrium 0.85 % Enriched-U core could be in principle the last stage of fuel strategy por Atucha-1 NPP, since the most significant economic savings could be reached just from the point of view of FA's operating costs.

However, another aspect could then become relevant : the necessity of a new fuel pool for FA irradiated FA storage up to the end of plant lifetime, since the present capacity using this type of SEU FAs is not enough.

One interesting alternative to avoid the expensive construction of a new fuel pool in the plant can be a future transition from 0.85 % Enrich.-U to around 0.95 to 1.0 % Enrich.-U, just after achievement of the full 0.85 % Enrich.-U load in the core. In that case the new gain in exit burnup will reduce the total accumulated irradiated FA inventory towards the end of plant lifetime avoiding the need of a new fuel pool, bringing at the same time an additional saving in the direct FA consumption cost contribution to the total operating cost.

#### ACKNOWLEDGEMENTS

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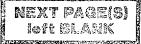
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# FUEL DESIGN, PERFORMANCE AND TESTING

(Session 2)

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# RIA TESTING CAPABILITY OF THE TRANSIENT REACTOR TEST FACILITY

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#### Abstract

The advent of high-burnup fuel implementation in LWRs has generated international interest in high-burnup LWR fuel performance. Recent testing under simulated RIA conditions has demonstrated that certain fuel designs fail at peak fuel enthalpy values that are below existing regulatory criteria. Because many of these tests were performed with nonprototypically aggressive test conditions (i.e., with power pulse widths less than 10 msec FWHM and with non-protoypic coolant configurations), the results (although very informative) do not indisputably identify failure thresholds and fuel behavior. The capability of the TREAT facility to perform simulated RIA tests with prototypic test conditions is currently being evaluated by ANL personnel. TREAT was designed to accommodate test loops and vehicles installed for inpile transient testing. During 40 years of TREAT operation and fuel testing and evaluation, experimenters have been able to demonstrate and determine the transient behavior of several types of fuel under a variety of test conditions. This experience led to an evolution of test methodology and techniques which can be employed to assess RIA behavior of LWR fuel. A pressurized water loop that will accommodate RIA testing of LWR and CANDU-type fuel has completed conceptual design. Preliminary calculations of transient characteristics and energy deposition into test rods during hypothetical TREAT RIA tests indicate that with the installation of a pressurized water loop, the facility is quite capable of performing prototypic RIA testing. Typical test scenarios indicate that a simulated RIA with a 72 msec FWHM pulse width and energy deposition of 1200 kJ/kg (290 cal/gm) is possible. Further control system enhancements would expand the capability to pulse widths as narrow as 40 msec.

#### 1. INTRODUCTION

Continued interest in extending burnup limits of LWR fuel to accommodate longer fuel-cycle lengths has led regulatory agencies and industry to evaluate the safety and reliability of LWR fuel at burnup values near and above 60 MWD/kgU. The degradation of fuel and cladding properties during irradiation has implications for fuel transient performance as well as steady-state performance [1,2]. LWR licensing in the US requires analysis against a small number of design-basis accidents. One class of such accidents is the reactivity-initiated accident (RIA), the most severe of which for LWRs results from a rod-drop or rod-ejection event. US Nuclear Regulatory Commission (USNRC) regulatory criteria [3] require that during such an accident the peak calculated fuel rod enthalpy not exceed 1170 kJ/kg fuel (280 cal/g fuel) to ensure core coolability. Furthermore, accidents for which fuel enthalpy is expected to reach threshold values at which failure is assumed must be analyzed to show that subsequent releases are within US Federal Code (10 CFR Part 100) limits. To allow calculations of radiological releases, other values are used to indicate cladding failure: critical heat flux values related to departure from nucleate boiling or, for low-power accidents in BWRs, 711 kJ/kg fuel (170 cal/g fuel). Recent simulated RIA tests of high burnup fuel (i.e., fuel with peak burnup around 55 MWD/kgU) have induced failures of irradiated LWR fuel rods at fuel enthalpies well below the threshold values [1,4,5], and therefore, the issue continues to receive considerable international attention [6]. Ongoing evaluation of the observed test fuel failures has indicated that fuel dispersal from low-enthalpy failures may raise questions regarding core coolability. As a result, personnel from the USNRC are now considering establishment of new, more conservative criteria intended to avoid cladding failure altogether [7]. It appears that additional testing (of some type) would be required to establish a coolability criterion above the cladding failure threshold. Such testing will be particularly important for efforts to develop fuel that can be licensed for irradiation well beyond current burnup limits (62 MWD/kgU in the US), as has been recently proposed [2].

In support of the development of the proposed High Efficiency Nuclear Fuel Program [2], personnel at Argonne National Laboratory (ANL) have been evaluating the capability of the Transient Reactor Test (TREAT) Facility to test LWR fuels under simulated RIA conditions. This effort has been comprised of three major tasks: 1) assessment of the requirements and need for RIA testing of LWR fuel, 2) conceptual design of a pressurized water test loop for the TREAT reactor, and 3) preliminary calculations of the ability of the TREAT reactor to provide the test conditions necessary for simulated RIA tests. This paper provides a description of the progress made to date with various elements of this effort. As background information, a description of the TREAT facility and the test programs and methods associated with its operations is also provided.

#### 2. TREAT FACILITY BACKGROUND

# 2.1 TREAT facility description

The TREAT facility is located at the Argonne National Laboratory-West site at the Idaho National Engineering and Environmental Laboratory, which is located outside of Idaho Falls. The TREAT reactor is an air-cooled, thermal, heterogeneous test facility designed to evaluate reactor fuels and structural materials under conditions simulating various types of transient overpower and undercooling situations During its 40-year history, the reactor has generated several thousand reactor transients, and hundreds of experiments have been performed for a wide variety of reactor development programs and fuel types.

The TREAT reactor core consists of a 19x19 array of fuel and reflector assemblies, which are each 10-cm (4-in.) square and 2.4-m (8-ft) long (Figure 1). Surrounding the array is a permanent graphite reflector 0.6-m (2-ft) thick, around which is a 1.5-m (5-ft) concrete biological shield. The assemblies contain a 1.2-m (4-ft) active fuel region with 0.6-m (2-ft) reflector regions above and below. Fuel assemblies are removed from the core array to accommodate emplacement of experiment hardware or loops. The TREAT reactor fuel is a dilute mixture of fine particles of highly-enriched  $UO_2$  dispersed in graphite and carbon, in which the U-235 is approximately 0.2 wt.% of the total mixture. The graphite and carbon fuel matrix absorbs heat rapidly, providing an essentially instantaneous large, negative temperature coefficient of reactivity. The core is capable of a 2.5-GJ maximum energy pulse, with a maximum power level of about 19 GW for temperature-limited transients. The core is air cooled and designed to remove the heat generated during steady-state operation or following transient operation. Throughout the duration of the reactor operating history, 6,640 reactor startups and 2,884 transient irradiations were completed generating a total of 2,600,000 MJ of reactor energy.

The TREAT reactor is capable of generating a wide variety of transient shapes. Transients can be terminated by the natural temperature response of the core, which can provide a power pulse with a FWHM of hundreds of milliseconds, or reactor power can be clipped by insertion of the transient control rods or the shutdown rods. The Automatic Reactor Control System (ARCS) provides for open and closed loop computer control. Open control provides transients which are initiated by a step insertion of reactivity and are terminated when a predetermined reactor period, power, energy release, or time interval has been realized. Closed control provides transients which follow a predetermined time and power or period profile and are terminated when a predetermined experiment parameter, reactor parameter or time interval has

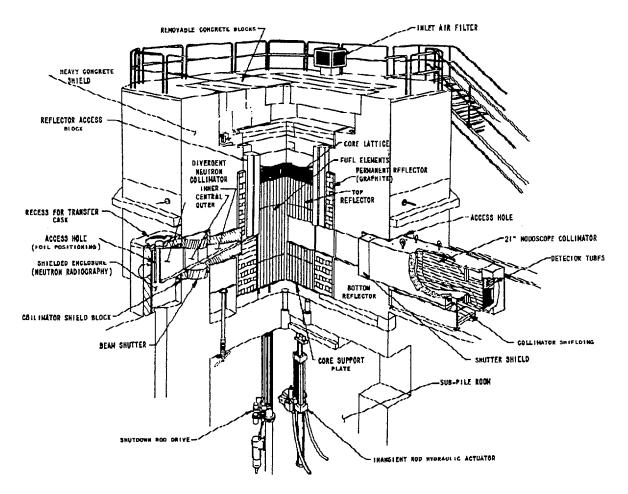


FIG.1. Cut-away view of the TREAT reactor showing major features.

been realized. The ARCS subsystems acquire reactor power, period, and transient control rod information, process the data, and issue transient rod position commands to generate specified reactor power-time profiles. The ARCS digital controllers and transient rod drive system are designed for the fast time response and speed required for production of transients with fractions of a second duration. The system is equally capable of producing transients lasting for tens of minutes.

Major upgrades have since been made to the TREAT facility to enhance its capability. These were most-notably (a) a sophisticated computer control system and transient simulator, (b) a computerized, real-time feedback system allowing communication between experiment vehicle instruments and the reactor control program to permit real-time automatic tailoring of the reactor power according to sensed events or conditions occurring within the experiment vehicle, (c) expansion of the building size and crane capacity, (d) reduction of constraints on allowable experiment vehicle size accommodated within the reactor, (e) an increase in sensing and recording capability of the hodoscope and enlargement of the hodoscope collimator, and (f) reconfiguration of the core to reduce undesired dependencies of the neutron-flux spatial distributions. The primary diagnostic systems available include a 360-channel (10x36 array) hodoscope, a fission product detection system, and pre-test and post-test neutron radiography.

# 2.2 TREAT testing experience and methods

Early TREAT tests studied the behavior of short fuel samples tested in dry capsules, in stagnant-coolant capsules, or in transparent assemblies that permitted viewing through the side of the capsule by high-speed optical cameras. Beginning in the 1960s and extending into the 1990s, flowing-sodium loops of various designs were used in the reactor for

testing oxide, carbide, and metallic fuel samples as large as seven-rod bundles of full-length, highburnup fuel rods. Also during that time, two capsule tests were performed on mixtures of uranium oxide fuel and steel to study transition-phase phenomena during hypothetical core disruption accidents in liquid-metal-cooled reactors. In addition, a series of source-term experiments were performed on irradiated light-water-reactor (LWR) oxide fuel in which fission product collection was of paramount importance. These test programs supported the existing LWR industry; the planned liquid metal fast breeder reactor (LMFBR) program, which focused on the Experimental Breeder Reactor-II, the Fast Flux Test Facility, and the Clinch River Breeder Reactor); and the liquid-metal-cooled reactor (LMR) program, which focused on the Integral Fast Reactor (IFR) concept.

Depending upon the goals of the experiments, various experiment "vehicles" have been designed and used to provide the proper thermal-hydraulic environment and containment for the fuel samples. Most of these were assemblies that fit within the TREAT core and were designed to be brought to, and removed from, the TREAT facility as a "package," being assembled and disassembled generally at the nearby Hot Fuel Examination Facility. These assemblies included low-pressure assemblies without coolant medium (some with windows allowing high-speed-photography by ex-core cameras), high-pressure stagnant-coolant (water or sodium) capsules, a high-pressure blow-down type sodium loop (with sodium tanks located atop the reactor), and high-pressure flowing-sodium "package" loops. These vehicles were designed to accommodate fuel-rod segments, single intact fuel rods, or arrays of up to seven fuel rods. Other vehicles, which were designed in considerable detail but never completed due to program redirection, included a large "package" steam loop designed for bundles of up to 37 full-length LMFBR fuel rods, a large "package" steam loop designed for an array of heavy-water-reactor (HWR) production-reactor fuel assembly components, and a large gas-cooled loop (most of which was designed to be located ex-reactor) for gas-cooled fast reactor (GCFR) safety tests.

A common objective of the various experiments and experiment series that were performed in TREAT was to provide information about the response of reactor fuel rods to accident transients and, in many cases, to also provide data on fuel-coolant interactions and on the motions of fuel, cladding, and coolant following cladding failure. The central importance of fuel motion in many of the tests led, in the 1960s, to the development of a fast-neutronhodoscope, a neutron collimating and detection system for fuel-motion diagnostics that was subsequently used as a primary test diagnostic system in most of the tests performed since that time, and which has been significantly upgraded from time to time.

The development and utilization of the various experiment vehicles noted above was motivated by the changing experiment needs as different types of nuclear reactor and reactor fuel issues arose. During the first few years of operation, a large number of fuel-rod meltdown experiments (85 during the first two years) were performed on unirradiated and irradiated fuel, most of them in dry capsules. These were largely phenomenological experiments in that they were used to identify and study the basic physical processes occurring during rapid overheating of the fuel-rod samples, rather than simulating specific accident conditions. Although sodium-bonded uranium-alloy fuel was the predominant type tested, various gas-bonded ceramic fuels, including uranium oxide, uranium sulfide, uranium carbide, thorium-based oxide, and mixed-oxide fuels, in various cladding materials, were also studied. By the mid-1960s, integral experiments in flowing-coolant loops were being conducted.

Tests supporting severe-accident safety evaluations on oxide-fuel rods under the US LMFBR program during the 1970s and the US/UK PFR-TREAT program during the 1980s included those on fuel-coolant interactions; fuel breakup and dispersal mechanisms; failure threshold and post-failure fuel dynamics under transient overpower (TOP) conditions; coolant boiling, voiding, and fuel-rod melt relocation during loss-of-flow (LOF) and during LOF-driven-TOP conditions; and transition-phase behavior under decay-heating conditions. Also during the 1980s, several experiments were performed on LWR fuel rods to determine fission-product release and transport characteristics, and a series of TOP experiments were performed on IFR metallic-fuel rods to measure pre-failure fuel elongation, cladding failure characteristics, and early post-failure fuel motion. As the sophistication of the tests increased through the years from relatively-simple "phenomenological" tests to the later "integral" tests that investigated interactions of multiple phenomena, improved analytic capabilities were developed and applied for use in experiment planning and post-test analysis. These included thermal-hydraulic models and codes to describe the conditions in the experiment vehicle and neutronics models to describe the neutronic interactions among the experiment fuel sample, experiment vehicle, and reactor core. Excellent knowledge of the initial and transient heating and cooling conditions during each test (which served as input to the detailed experiment predictive computations) was obtained through a variety of pretest steady-state and transient irradiations of neutronic calibration samples, as well as an in situ heat-balance transient performed just prior to the actual test. The accuracy of 5 to 10% achieved in the determination of the fission-heating and coolant flow rate measurements in the later loop experiments was due in large part to the diversity of calibrations employed.

#### 3. RIA TESTING OF LWR FUEL

The variables that most affect fuel failure during an RIA are cladding temperature and fuel expansion and/or fission gas release; e.g., [1,4]. (The degree of cladding oxidation and hydriding induced by normal, steady-state operation prior to the RIA also affect cladding properties during the event.) The reactor parameters that control these variables are power pulse width, typically expressed as the full-width at half-maximum (FWHM) value of reactor power as a function of time, and energy deposition into the fuel, typically manifested as fuel enthalpy rise expressed in units of calories per gram of fuel.

Three-dimensional neutronics and thermal-hydraulics calculations of the response of a LWR core to an RIA generally predict energy insertions of less than 418 kJ/kg (100cal/g) in any fuel rod and power pulse FWHM values ranging from 70 to 100 msec, although one recent calculation resulted in 30 to 80-msec power pulses [8]. Most RIA tests of high-burnup fuel have been performed with excessively-narrow power pulses (<10 msec FWHM), although the more recent tests in CABRI have been performed with a power pulse that approximates an RIA pulse in a PWR. Narrow power pulses induce fuel pellet expansion at a faster rate, leading to a higher PCMI-induced strain rate in the cladding, and induce relatively higher temperatures in the rim region of the fuel pellet (due to energy deposition occurring at a faster rate than heat transfer mechanisms can remove energy), intensifying the expansion phenomena that cause PCMI and thereby raising the peak hoop stress in the cladding. Furthermore, the peak stress on the cladding occurs before the cladding can heat up to temperatures at which the cladding will more easily yield in a ductile manner; thus, brittle failure may be artificially induced. These tests have also been performed with non-prototypic coolant conditions, using sealed capsules containing stagnant air or water coolant or using loops with flowing sodium, leading to non-prototypic heat transfer through the test.

The tests performed to date have been suitable for the stated objectives of those test programs, which is to study PCMI during the early stages of the transient [4], and the results reported from those test programs thus far have provided tremendous insight into the mechanism of PCMI-induced failure. However, any additional tests to be performed for determining the RIA behavior of high-burnup fuel should more closely simulate postulated RIA events, using 30 to 100-msec power pulse widths, inducing peak fuel enthalpies of 83.7 to 837 kJ/kg (20 to 200 cal/g), employing prototypic coolant conditions and test rods with the improved cladding alloys in current use. A prototypic fuel temperature during a test is ensured by a prototypic power distribution through the fuel (which is determined by the duration and magnitude of the power pulse and by the neutron spectrum) and by prototypic coolant conditions. Coolant flow conditions during a test are best provided by a water loop that supplies flow to the test fuel at prototypic temperatures and flow rates.

## 4. CONCEPTUAL DESIGN OF A PRESSURIZED WATER LOOP

A high-pressure, circulating water loop is being designed for TREAT for testing irradiated fuel rods under thermal-hydraulic conditions (inlet water pressure, temperature, and flow rate) prototypic of pressurized water reactor (PWR) and boiling water reactor (BWR) conditions. Modification of the concept would allow testing of CANDU-type fuel elements. Basic features of the loop, which is at the end of the conceptual design phase, are shown in Figures2 and 3. A U-shaped extension (in-pile assembly) in the reactor core region contains the test train which houses the test fuel rods (up to three PWR rods or two BWR rods). Within the test train, each rod will be located within an individual flowtube, and separate thermal-hydraulic conditions will be provided for each rod. The in-pile assembly is designed to be assembled with test train and fuel rods at the nearby Hot Fuel Examination Facility (HFEF), brought to TREAT for the RIA test, and then returned to HFEF for disassembly and post-test inspection. (The design of the in-pile assembly takes advantage of key design features of existing loops used in the liquid-metal reactor program, allowing utilization of existing transfer casks and hot-cell interface hardware and operational procedures.) The primary containment wall of the in-pile assembly is constructed of Zircaloy to minimize neutron attenuation and thus increase the fission power that can be generated in the test fuel. An example cross section of the in-core assembly through the fueled region is shown in Figure 4. Filters will be used for control of fission products in tests

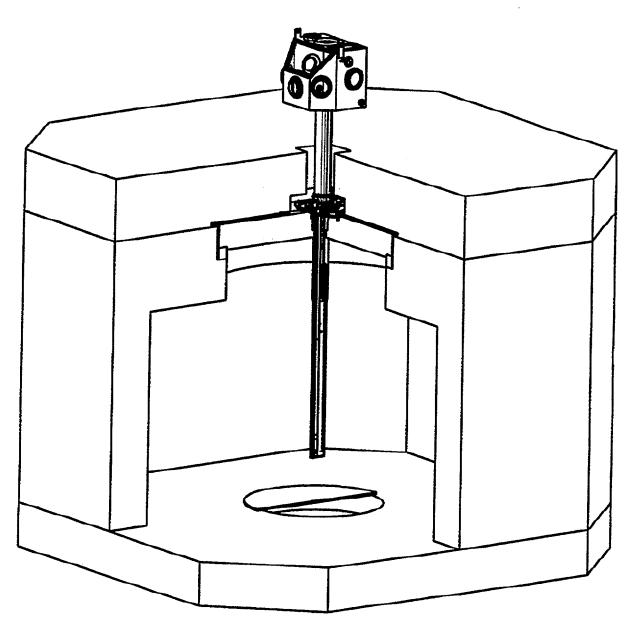


FIG. 2. Sketch showing conceptual pressurized water loop inserted into TREAT reactor (only the reactor's concrete biological shielding is shown).

involving breaching of the fuel rod cladding. The test train will be designed to be removable from the in-pile assembly post-test and to be reused after tests in which the sample fuel rods remain intact. The containment structure of the in-pile assembly which houses the test train will also be reusable, even after tests with failed fuel rods. The in-pile assembly is sized to be accommodated in the neutron radiography facilities at TREAT and at HFEF.

The main, ex-reactor portion of the loop, constructed of stainless steel, remains in place from test to test. Its fundamental components are water holding tanks, pump, water heater, and cesium filter/trap, all sized to be handled by existing casks. Much of the loop support equipment will be located on top of the TREAT reactor. A secondary containment around the entire system provides required protection against potential release of radioactive material. Instrumentation is provided in the test fuel region and elsewhere in the loop to measure temperatures, flow rates, pressures, and other parameters of interest.

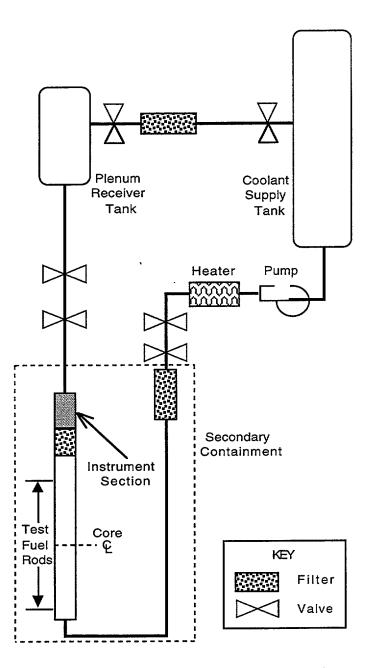


FIG. 3. Schematic representation of major components of conceptual pressurized water loop.

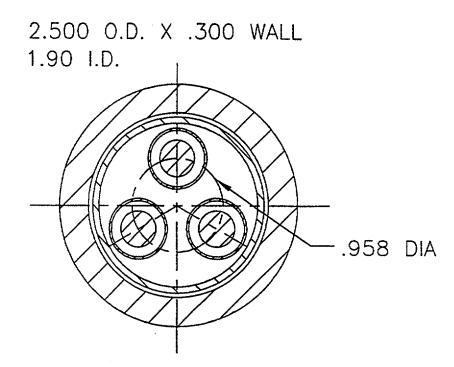


FIG.4. Cross section of test fuel region of conceptual pressurized water loop showing arrangement of three PWR rods.

### 5. EVALUATION OF HYPOTHETICAL TREAT RIA TESTS

Proper assessment of the ability of TREAT perform RIA tests requires an indication of the amount of energy the reactor can deposit into a test fuel rod and, in particular, peak fuel enthalpy increase induced in the test fuel. Calculation of the time-dependent thermal response of the fuel rod is therefore important to determine if a hypothetical simulated RIA test will achieve the desired test conditions. Efforts currently underway at ANL will establish the capability to calculate, using modern methods, the time-dependent energy deposition and time-dependent temperature response of a test pin, which will allow calculation of peak fuel enthalpy increase and cladding temperature. Because this work is not complete, the current evaluation is limited to preliminary calculations of energy deposited into test rods during hypothetical transients. This section summarizes the work performed thus far.

### 5.1 Predicted LWR fuel test parameters

The TREAT reactor transient parameters (i.e., control rod motion and positions) required to generate power pulses of various duration (i.e., 60 to 100 msec) were determined using the reactor simulator associated with the ARCS. The simulator employs a point kinetics core model, which has been demonstrated to reliably predict integral core response. A PWR RIA transient having a full-width at half-maximum (FWHM) of less than 100 msec can be simulated by clipping a maximum reactivity, temperature-limited transient to minimize the pulse width (which would also limit the total energy). As an example, a transient of this type having a FWHM of 72 msec would be limited to a maximum pulse energy of approximately 1400 MJ (Figure 5). If desired, the RIA pulse can be preceded by a pre-transient irradiation to induce a desired pre-test fuel temperature profile, or can be followed by a low-power, post-transient irradiation to simulate decay heat; such pre-transient or post-transient operation would alter the characteristics of the example transient illustrated in Figure 5.

Using the transient parameters determined with the simulator, a series of one-dimensional transport calculations using the ANISN code and a 50-group neutron cross section set were performed to determine the test-fuel power coupling. The calculations were used in conjunction with radiochemical data collected during a test series conducted in the TREAT reactor during late 1992 and early 1993. (Even though that particular test series was conducted in

support of LMR fuel testing, the core loading and test vehicle are similar to the expected LWR core loading and test vehicle.) Test-fuel power coupling is defined as the ratio of test-fuel power to reactor power. For PWR test fuel having a beginning-of-life enrichment of 4.95% and irradiated to a burnup of 78,000 MWD/kgU, the power coupling was calculated to be 0.88 J/g fuel per MJ of reactor energy for a PWR RIA transient utilizing the LWR test loop and vehicle described in the preceding section. The total energy which could be deposited in the test fuel during the 1400 MJ transients presented in Figure 5 would be 1200 kJ/kg (290 cal/gm). Using the transient control rods, the pulses can be tailored to almost any size. For example, a transient with a power pulse FWHM of about 57 msec would deposit approximately 412 kJ/kg (100 cal/g) in the PWR test fuel. These energy deposition values compare favorably with the energy deposition values of French and Japanese RIA tests that were able to induce peak fuel enthalpies up to 836kJ/kg (200 cal/g).

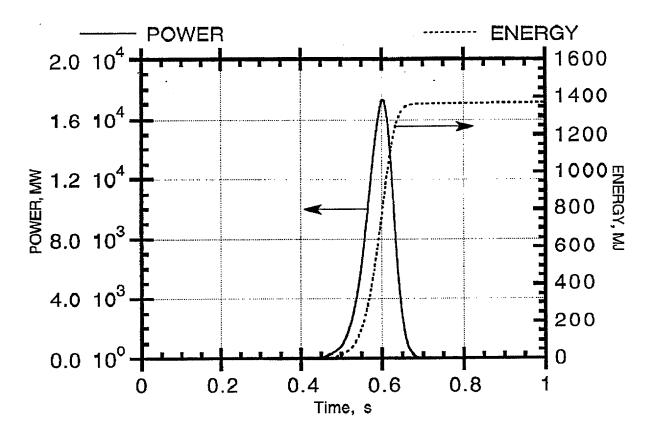


FIG. 5. Plot of TREAT reactor power and energy for hypothetical RIA-type transient resulting in 1400-MJ pulse with a 72-msec FWHM capable of depositing 1200 kJ of energy per kg of fuel (290 cal/g).

Testing of BWR test fuel exposed to extended levels of burnup will be similar to the PWR results discussed earlier. Since the fissile content of the pins is very low, there is very little self shielding. Therefore, the increased size of the BWR pins will not significantly affect the energy deposition.

#### 5.2 Predicted CANDU-type fuel test parameters

The conceptual LWR test loop and vehicle and the TREAT reactor could be used to test a wide variety of fuel types. For example, an unirradiated naturally enriched uranium oxide CANDU type fuel pin was calculated to have a power coupling of 0.50 kJ/kg fuel per MJ of reactor energy in the LWR test loop and vehicle. A CANDU RIA transient having a FWHM of 1 to 2 seconds could generate up to 2 GJ of reactor energy and 1000 kJ/kg (240 cal/g) of test fuel energy. Similar energy depositions could be expected for LOCA-type transients.

### 5.3 Potential improvements in TREAT RIA capabilities

Several concepts designed to provide significantly narrower RIA pulses are currently being investigated. One concept which shows promise is a He-3 system designed to insert significant quantities of negative reactivity into the core very rapidly. By making use of existing control rod penetrations, the system could be installed in the core without requiring modification of the facility. The transient simulations discussed in Section 5.1 were repeated assuming emplacement a He-3 system capable of inserting an additional 5% negative reactivity in 5 msec. The calculated FWHM values ranged from 46 to 60 msec (Figure 6).

In addition to a He-3 system, the pulse widths can be further reduced by making changes to the ARCS software and reducing the minimum allowable reactor period. It is anticipated that the combined effect these changes would allow RIA FWHM values of approximately 40 msec.

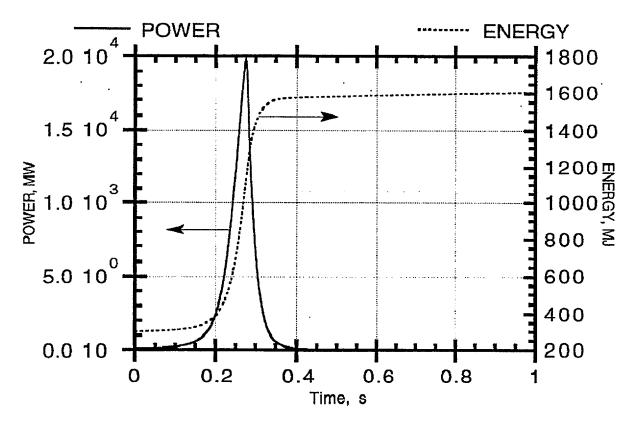


FIG. 6. Plot of TREAT reactor power and energy for hypothetical RIA-type transient, including a 300 MJ pre-transient irradiation, resulting in a 1600-MJ pulse with a 48-msec FWHM. The use of a He-3 shutdown system was assumed for this simulation.

# 6. SUMMARY

The advent of high-burnup fuel implementation in LWRs has generated international interest in high-burnup LWR fuel performance. Recent testing under simulated RIA conditions has demonstrated that certain fuel designs fail at peak fuel enthalpy values that are below existing regulatory criteria. Because many of these tests were performed with nonprototypically aggressive test conditions, the results (although very informative) do not indisputably identify failure thresholds and fuel behavior. The capability of the TREAT facility to perform simulated RIA tests with prototypic test conditions is currently being evaluated by ANL personnel. TREAT was designed to accommodate test loops and vehicles installed for inpile transient testing. During 40 years of TREAT operation and fuel testing and evaluation, experimenters have been able to demonstrate and determine the transient behavior of several types of fuel under a variety of test conditions. This experience led to an evolution of test methodology and techniques which can be employed to assess RIA behavior of LWR fuel. A pressurized water loop that will accommodate RIA testing of LWR and CANDU-type fuel has completed conceptual design. Preliminary calculations of transient characteristics and energy deposition into test rods during hypothetical TREAT RIA tests indicate that with the installation of a pressurized water loop, the facility is quite capable of performing prototypic RIA testing. Further control system enhancements would expand the capability to pulse widths as narrow as 40 msec.

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# A NEW TECHNIQUE TO MEASURE FISSION-PRODUCT DIFFUSION COEFFICIENTS IN UO $_2$ FUEL

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## Abstract

This paper describes a new out-reactor technique for the measurement of fission-product diffusion rates in UO<sub>2</sub>. The technique accurately simulates in-reactor fission-fragment effects: athermal diffusion that is due to localized mixing in the fission track, radiation-enhanced diffusion that is due to point-defect creation by fission fragments, and bubble re-solution. The technique utilizes heavy-ion accelerators – low energy (40 keV to 1 MeV) for fission-product implantation, high energy (72 MeV) to create fission-fragment damage effects, and secondary ion mass spectrometry (SIMS) for measuring the depth profile of the implanted species. Preliminary results are presented from annealing tests (not in the 72 MeV ion flux) at 1465°C and 1650°C at low and high concentrations of fission products.

## 1. INTRODUCTION

Release of fission gas and other volatiles from fuel during irradiation is a critical performance factor because it can lead to sheath defects, and it contributes to the inventory available for immediate release during off-normal conditions and spent-fuel storage. One of the tasks of fuel-performance codes used throughout the nuclear industry is to predict the amount of gas released under various normal and off-normal operating conditions. After over 30 years, considerable work still is being applied to improving these computer codes. For example, the IAEA has sponsored a program over the last few years comparing fuel-performance codes to assess how they perform in a set of blind tests [1].

There are several steps between the formation of fission gas and its release to the fuel-sheath gap. The first is the diffusion of the gas atoms after their formation to grain boundaries where they coalesce into bubbles. This step can be a thermally activated process – normal diffusion by jumping from one lattice site to an adjacent one or from one interstitial site to an adjacent one (or by a more complicated neutral tri-vacancy mechanism as some propose [2]). But there is also a non-thermally activated process (termed *athermal* diffusion) that leads to the movement of gas atoms to grain boundaries – the passage of fission fragments through the matrix causes rearrangement of atoms in the *fission track*, and the result is similar to the random walk of diffusion. The passage of the fission fragments can be likened to a brief local heating or melting in the fission track (approximately 8  $\mu$ m long). This is the important process at low temperatures where thermally activated diffusion is relatively slow.

In the process of diffusing from within the grains to grain boundaries, there is a complication – coalescence of gas atoms at defects in the matrix can produce tiny gas bubbles. These bubbles act as traps, effectively preventing the gas atoms from re-entering the fuel matrix. The bubbles migrate more slowly than the individual gas atoms, and thereby reduce the flux of gas atoms reaching the grain boundaries. Again, the fission fragments assist: as they pass through bubbles, they 'knock' gas atoms back into the  $UO_2$  matrix, and can even totally eliminate small bubbles. This is termed *re-solution*. Taking into account the delay of gas atoms in these micro-bubbles, fuel-performance codes use an *effective* diffusion coefficient for the overall rate at which gas atoms move through grains. The diffusion rate strictly within the  $UO_2$  matrix between bubbles, without considering trapping at bubbles, is termed *intrinsic* diffusion.

Measurements of diffusion rates of fission gas in nuclear fuels are difficult. First, the techniques usually rely on simple measurements of the rate at which the diffusing species are evolved from the samples. This is inaccurate – the best techniques for diffusion measurements, in general, measure profiles of the diffusion species (or of one particular isotope of that species) within the sample after diffusive spreading of a concentrated source, such as a deposited layer, has occurred. Also, in measuring the gas evolved from samples, all processes that contribute to movement of gas to the fuel surface are included, not just diffusion to the grain boundaries. While total gas evolution is an end-result that codes need to predict properly, this technique is not conducive to understanding or modelling individual processes so that the predictive capabilities being developed are applicable outside the specific conditions of the tests being done. Another reason for the difficulty of diffusion measurements is that out-reactor techniques do not include the effects of fission fragments — the athermal diffusion and re-solution of bubbles, described above. In-reactor techniques, on the other hand, are difficult, expensive, and test parameters are not easily controlled or measured.

Dozens of papers describe diffusion of fission gases in  $UO_2$  – see, for example, review articles by Catlow [2], Matzke [3], and Lawrence [4]. There is a large spread in reported diffusion rates; the new method described herein has the potential to be the most accurate and to be able to measure the effects of influences such as impurities, fission products and stoichiometry changes.

This technique is an out-reactor technique that gives in-reactor-equivalent results. That is, the effects of fission fragments are properly simulated. The method determines both intrinsic and effective diffusion rates, and is not based on measuring the rate or amount of gas evolution from test samples. Rather, it is a technique based on profiling of the diffusing species after spreading of a concentrated layer has occurred.

# 2. METHOD

The basics of the method are (subsections provide details):

- Preparing fuel samples
- Implanting a stable isotope of a fission product into a thin layer near the surface of the samples. A small fluence<sup>1</sup> means that the concentrations are low enough that bubbles have a low probability of formation. Larger fluences will favor the formation of bubbles. Measurements from the former provide intrinsic diffusion rates; measurements from the latter provide effective diffusion rates.
- Diffusive spreading of this implanted layer either by heating or by a second process akin to athermal diffusion the implanted sample surface is bombarded by iodine ions of energy 72 MeV, typical of fission-fragment energies. Because this energy is much higher than that used in the implantation step, these ions pass completely through the implanted layer and cause diffusion almost exactly as fission fragments do.
- Measurement of the concentration of the implanted ions, as a function of depth, before and after diffusion broadening, by secondary ion mass spectrometry (SIMS).

# 2.1. Sample Preparation

One surface of each sample of sintered UO<sub>2</sub>, simulated extended burnup fuel (SIMFUEL) and single crystal UO<sub>2</sub> was mechanically polished and annealed at 1500°C in flowing Ar-4%H<sub>2</sub>. The anneal removes the effects of polishing damage near the surface.

<sup>&</sup>lt;sup>1</sup> The term *fluence* is used to indicate the total number of implanted atoms (or ions) per unit area of sample in the beam.

# 2.2. Implantation

Samples were implanted with iodine or xenon using an accelerator at an ion-implantation energy of approximately 1 MeV. This gave a mean range of about 150 nm. Other samples were implanted with krypton, rubidium or iodine at about 40 keV, which gave a mean range of 10 to 20 nm. The implantation fluence was varied to achieve a wide range in concentrations of the implanted species. The lowest fluence used was  $1 \times 10^{13}$  ions/cm<sup>2</sup>, and the highest was  $1 \times 10^{16}$  ions/cm<sup>2</sup>. New samples are being prepared, some with europium implantation, to fluences even lower than those previously used to ensure that bubbles do not form.

# 2.3. Diffusive spreading of the implanted layer

As indicated previously, two methods of broadening the implanted layer have been used: thermal anneals and re-bombardment with high-energy ions to simulate fission fragments. For the latter, iodine at 72 MeV was used because this represents a typical fission-fragment element at a typical fission-fragment energy. These ions have a range of about 6  $\mu$ m, and thus pass through the implanted layer and cause localized diffusion within the volume of their 'tracks'. The only differences to in-reactor fission-fragment fragment effects are

- (a) all the ions move essentially in the same direction, compared with all directions for fission fragments;
- (b) all accelerator ions are iodine, whereas, in-reactor, there are many fission-fragment elements; and
- (c) the energies of the ions as they pass through the implanted layer all have approximately the same energy, whereas fission-fragments in-reactor would have a complete distribution of energies representative of fission fragments at all points along their paths.

Points (a) and (b) will not likely have much effect on diffusion; in point (b) because ions slowing to rest from 72 MeV occur mainly by electronic interactions between the ion and the matrix, this process is independent of ion species. Nuclear interactions occur only at the end of the implantation track because the ion is nearly at rest. However, point (c) could potentially have an effect on diffusion rate - this could be tested and corrected for by using different energies or by using a range of energies in one test. This has not yet been done. Also, all the accelerator tests have been at ambient temperature (<150°C) to effectively eliminate thermal diffusion and observe effects that are totally attributable to the accelerator bombardment. Tests at elevated temperatures should be performed because the fission fragments will also affect the diffusion rates through much of the thermal regime. This effect occurs because the fission fragments induce point defects, additional to those that are naturally present. In other words, the concentration of point defects that contribute to thermal diffusion is not the thermodynamic equilibrium concentration but a higher concentration that is due to those created by the passage of fission fragments through the fuel matrix. This is termed radiation-enhanced diffusion (RED); only at sufficiently high temperatures, above about 1400 to 2000 K, depending on many factors such as the fuel power, is the equilibrium concentration of point defects sufficiently high that the extra defects that are due to fission fragments are largely inconsequential.

# 2.4. SIMS measurements of the distributions

A CAMECA IMS 6f SIMS is being used to profile the iodine distributions. This instrument is extremely sensitive to iodine – concentrations ranging from less than 10<sup>14</sup> atoms/cm<sup>3</sup> to greater than 10<sup>20</sup> atoms/cm<sup>3</sup> have been measured. To calibrate the SIMS sputtering rate, depths of all craters were measured with a stylus profilometer. Results of these depth measurements are given in the next section. Figure 1 shows an example of the distribution directly after implantation of a low fluence of iodine, and after a thermal anneal at 1650°C for 2 h, showing the broadening that has occurred. In contrast, Figure 2 shows the results after a similar heat treatment for a sample with a high-fluence implant. Little broadening of the distribution has occurred. The implanted iodine has formed bubbles, which, without

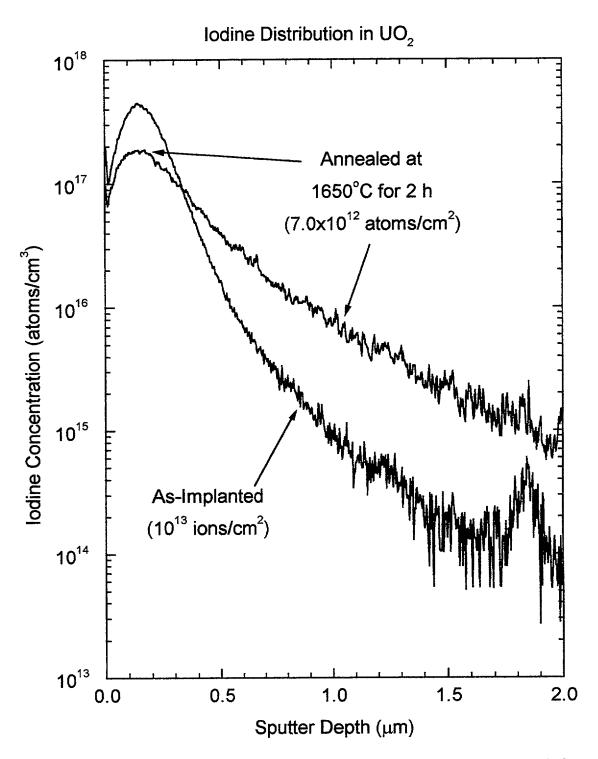


FIGURE 1 SIMS depth profile of a low fluence of iodine implanted at 1 MeV before and after an anneal at 1650°C for 2 h.

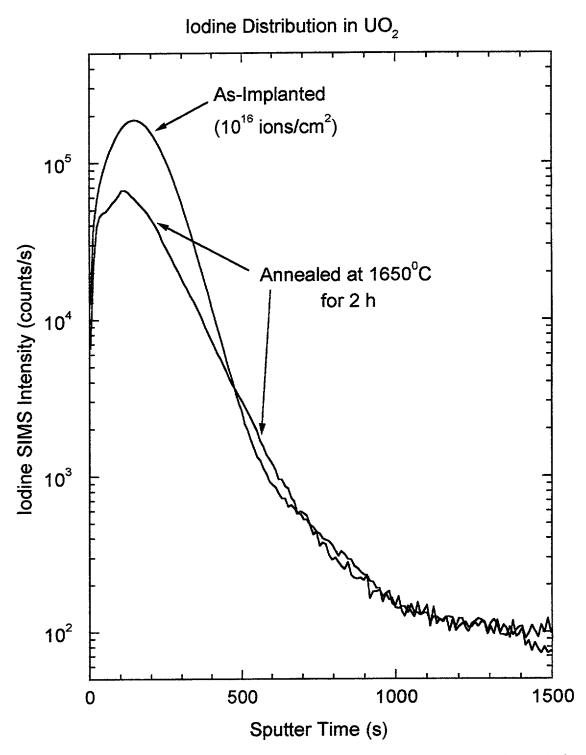


FIGURE 2 SIMS depth profile of a high fluence of iodine implanted at 1 MeV before and after an anneal at 1650°C for 2 h.

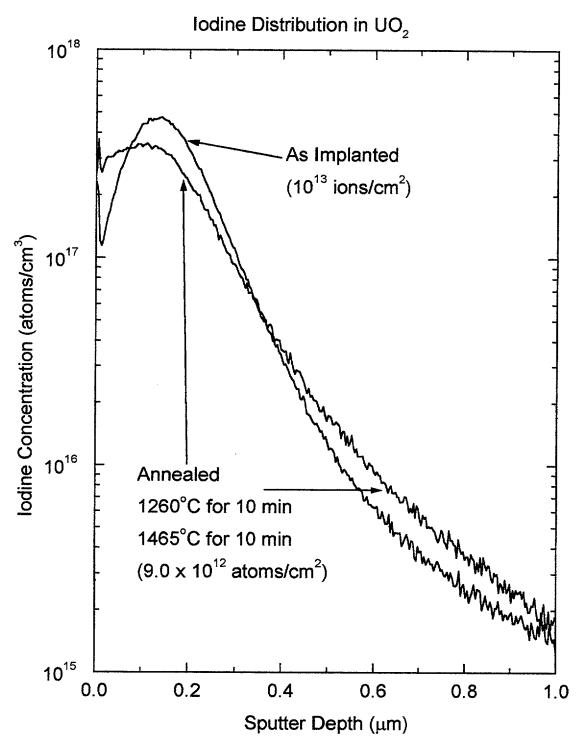


FIGURE 3 SIMS depth profile of a low fluence of iodine implanted at 1 MeV before and after anneals at 1260°C for 10 min, and 1465°C for 10 min.

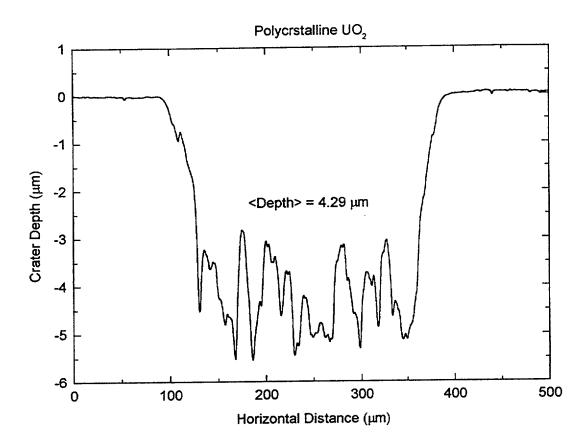


FIGURE 4 Stylus profilometer trace across a representative sputter crater in a sintered UO2 sample implanted with a low fluence of iodine.

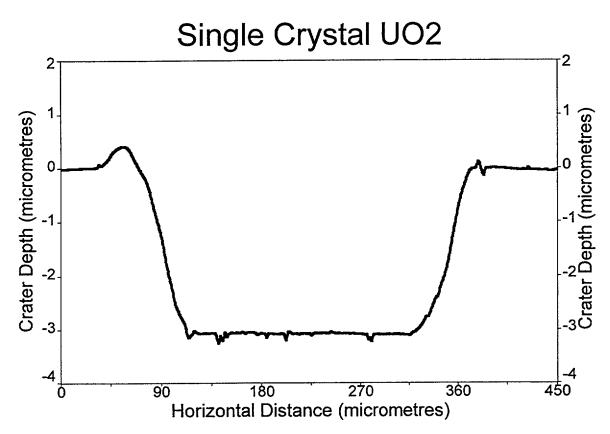


FIGURE 5. Stylus profilometer trace across a representative sputter crater in a single-crystal U sample implanted with a low fluence of iodine.

re-solution because of fission fragments, has largely immobilized the iodine<sup>2</sup> and inhibited diffusion into the bulk.

Figure 3 show the results for a low-fluence implantation at lower temperatures  $-1465^{\circ}$ C. The main observation is that, without fission-fragment effects, the temperature was too low for diffusion into the body of the sample. A second observation is that iodine has migrated towards the sample surface – note the relatively higher concentrations near the surface and the shift of the peak maximum toward the surface. We interpret this as evidence that the damage to the lattice, caused by the ion implantation, enhanced diffusion within the damaged zone. From the sample surface to the peak maximum, the lattice was heavily damaged during the implantation, and diffusion occurred there. On the other side of the peak maximum, however, there was much less damage and little diffusion.

# 2.5. Depth profiling of SIMS craters

As stated above, the craters produced by the SIMS distribution measurements are all depth-profiled to calibrate the sputtering rate for each SIMS run. Profiles of the crater floors from the sintered samples were rough (Fig. 4). Because similar profiles for single crystal samples (Fig. 5) are smooth, and the horizontal scale of roughness is approximately that of the grain size, this roughness was attributed to different sputtering rates for different crystal orientations. For the rough craters, an average depth was used to determine the sputtering rate. Because the SIMS analysis is over a sample area 60  $\mu$ m across, 50 to 100 grains are included in the analysis, and this number provides sufficient averaging to use an average depth in the SIMS calibration.

## 3. SUMMARY

Accelerators and SIMS have been used to develop a new technique for measuring fission-product diffusion rates, both intrinsic and effective (includes holdup at bubbles). Although the methods are outreactor, in-reactor effects of fission fragments are included. Preliminary results show that at 1650°C, iodine at high concentrations is held up at bubbles if re-solution of the bubbles is not included. At low concentrations, fewer bubbles form at 1650°C and diffusion of iodine is seen. At 1465°C, movement of iodine into the bulk  $UO_2$  was not observed in anneals without fission-fragment effects, even at low iodine concentrations, but movement towards the surface through the layer heavily damaged by the implantation was observed. Analysis of samples prepared using a high-energy accelerator to simulate inreactor behaviour that is due to fission fragments will be undertaken in the near future.

The results obtained to date in this study are promising. It appears that a more complete understanding of diffusion rates can be established, and we expect to be able to determine bounding limits on intrinsic and effective diffusion coefficients for specific fission products in  $UO_2$  in the near future.

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 $<sup>^{2}</sup>$ The reduction of iodine peak intensity and the slight asymmetry towards the sample surface is evidence that iodine diffusion through the damaged zone near the surface did occur, possibly during the temperature rise 1650°C before bubble formation was advanced.



## TECHNOLOGIES FOR OBTAINING LARGE GRAIN SINTERED PELLETS

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**Abstract:** A way to increase fuel burn-up is to use a large grains fuel pellets structure. The paper presents a literature review related to the technologies and the methods for large grains sintered pellets manufacturing.

A flowsheet for large grains sintered pellets obtaining by  $Nb_2O_5$  dopant addition in UO<sub>2</sub> sinterable powder, pressing and sintering in H<sub>2</sub> atmosphere is showed.

In the diagrams are presented the dependency of the main sintered pellets characteristics (pore radius distribution, pores volume, density, grains size) as function of the  $Nb_2O_5$  dopants concentration,  $UO_2$  sinterable powder nature and sintering temperature. Other sintered pellets characteristics (electrical conductivity, Seebeck coefficient, high temperature molar heat capacity and thermomechanical properties) are commented.

The beneficial effects resulted from theoretical and practical projects are presented.

# 1. Introduction

The actual tendency all over the world is to manufacture fuel bundles capable to resist high burn-up. The factors affecting the burn-up increase are:

- the pellet-cladding mechanical interaction (PCMI);
- the oxidation and hydriding of the Zircaloy-4 sheath;
- the increase of internal pressure;
- stress corrosion cracking;
- Zircaloy-4 irradiation growth;
- fuel swelling.

To diminish the internal pressure of the Zircaloy-4 cladding, large grain UO<sub>2</sub> pellets can be used, by reducing the fission gas release [1], [2].

One of the ways to increase pellets grain size without increasing sintering temperature and time is the addition of small quantities (< 1% wt M/U) of sintering additives (aliovalent metal or rare earth oxide).

By the addition of certain dopants in the UO<sub>2</sub> powder (TiO<sub>2</sub>, Nb<sub>2</sub>O<sub>5</sub>, Cr<sub>2</sub>O<sub>3</sub>, CaO,V<sub>2</sub>O<sub>5</sub>) the grain size, porosity and the mean free diffusion path are increased, whereas the grain boundary area is reduced [3] - [7].

# 2. Technologies

For the production of niobia doped UO<sub>2</sub> fuel the "direct pelletizing process" which has been developed in relation with the AUC powder technology can be applied without any change beside the admixture of niobia to the UO<sub>2</sub> powder.

In a master mix UO<sub>2</sub> and Nb<sub>2</sub>O<sub>5</sub> powders are added and homogenized. The blended powder is directly pressed without the addition of a lubricant. The green pellets are sintered in the sintering furnace. Under the same sintering conditions, the density of the pellets can be adjusted by U<sub>3</sub>O<sub>8</sub> addition, UO<sub>2</sub> - Nb<sub>2</sub>O<sub>5</sub> pellets with densities between 9.9 - 10.75 g/cm<sup>3</sup> and grain size between 2 - 50  $\mu$ m being obtained [8].

Other methods to obtain uranium dioxide pellets with large grain sizes:

- heating sintered pellet of uranium dioxide at temperatures higher than 1700°C in a hydrogen stream containing silicate vapours, resulting from the aluminum silicate decomposition. The grain size values can be increased to more than 50µm [9].
- production of sintered uranium dioxide pellets by the addition of sintering agent (10 - 55wt% MgO and 90 - 45wt%SiO<sub>2</sub>) or precursor thereof in the composition ranging from 0.1 - 0.8wt% of a sinterable mixture. The resulting mixture is turned into a compact. The sintering process is performed at a temperature where the sintering agent forms a liquid phase to produce a

sintered product. The precursor is thermally decomposed below the sintering temperature [10].

- obtaining of sintered UO<sub>2</sub> nuclear fuel pellets, with the average grain size ranging from about 30µm to about 80µm by the addition of magnesium aluminosilicate in uranium dioxide powder. The pores volume is ranging between 2 and 10% [11].
- addition of Cr<sub>2</sub>O<sub>3</sub> or Al<sub>2</sub>O<sub>3</sub> and of small quantities of SiO<sub>2</sub> into the uranium dioxide powder [12].

At the Institute for Nuclear Research (ICN) - Pitesti a technology for obtaining large grains size UO<sub>2</sub> pellets using dopants [13] was developed. The manufacturing flowsheet is presented in Figure 1.

The  $UO_2$  non-free flowing powder, manufactured by ADU route, was mixed with  $Nb_2O_5$  in a Y - con master mix. The blended powder was pre-pressed and granulated using a 0.5 mm sieve. The resulted granules were mixed with Zn stearate as lubricant. The green pellets were manufactured by bilateral pressing. The compacts were directly sintered (4 hours at 1700°C) in a standard continuos sintering furnace with a dewaxing step at 900°C.

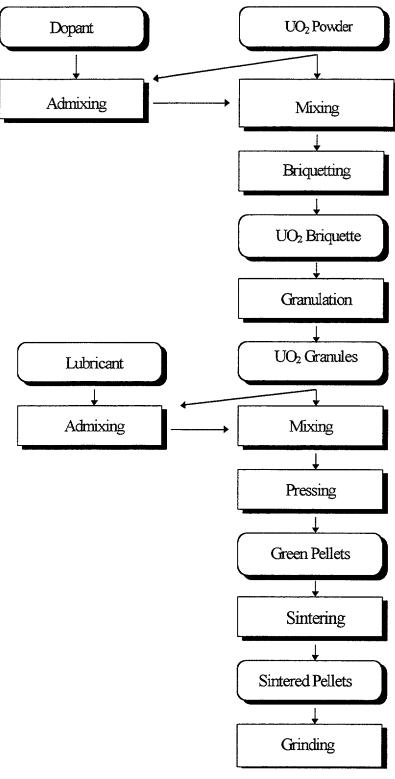
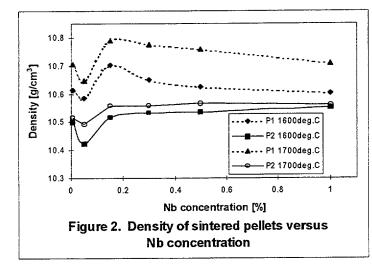


Figure 1. Large grain size sintered pellets manufacturing flowsheet

# 3. Properties of Nb<sub>2</sub>O<sub>5</sub>-doped UO<sub>2</sub> pellets

The physical, thermal, electrical and mechanical properties of  $Nb_2O_5$  doped UO<sub>2</sub> pellets are affected, as compared to the undoped ones.



The variation in densitiy of  $Nb_2O_5$  - doped  $UO_2$  pellets as a function of Nb content is presented in Figure 2 [13]. A very small concentration of  $Nb_2O_5$  ( 0-0.1%Nb/U) leads to a minimum value of the  $UO_2$ pellet density. The density of

 $UO_2$  pellets increases with the increase of the Nb content for low concentrations (0.1 ÷0.2% Nb/U). At concentrations higher than 0.2%Nb/U, the density values are different, depending on the manufacturing routes: the increase of the Nb concentration slowly diminishes the density (P<sub>1</sub> -ADU route) or remains constant (P<sub>2</sub> - IDR route).

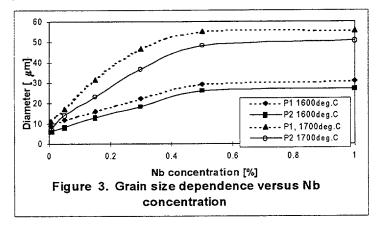
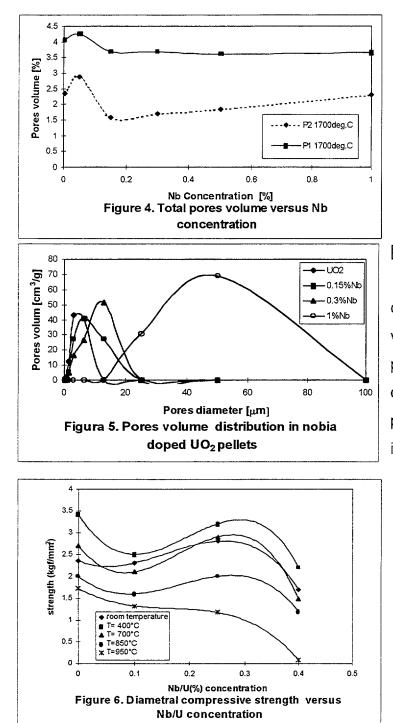


Figure 3 displays the grain size dependence of the UO<sub>2</sub> pellets the Nb on content [13]. The dopant addition determines а significant grain growth. The average grain size, versus concentration, shows an

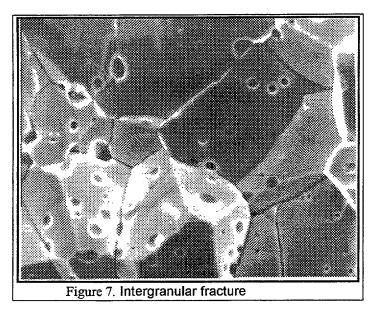
increase up to a maximum value (0.5%) followed by a constant value.



The addition of dopant brings changes into the porosity of sintered pellets. The total pores volume evolution versus Nb concentration in Nb<sub>2</sub>O<sub>5</sub> doped UO<sub>2</sub> sintered pellets is presented in Figure 4 [13].

At low Nb concentrations the pore volume is distributed in small lf pores. the Nb concentration increases, the pores volume is distributed in large pores, that prevail, while small pores volume is negligible (Figure 5) [13]. Nb addition affects the UO<sub>2</sub> pellets mechanical properties [13], [14]. The effect of Nb dopant is to strengthen the UO<sub>2</sub> material (if the Nb concentration increases), up to 850°C. At higher temperatures the

influence of the addition is diminished and disappears completely at 950°C, Figure 6.



The transgranular fracture observed at low temperature ( $\leq$ 400°C) points out the intense cohesion between the grains. If the temperature increases the intergranular fracture is dominant and only this type of rupture takes place at high temperature (  $\geq$  850°C), Figure 7.

The electrical conductivity and Seebeck coefficient of Nb<sub>2</sub>O<sub>5</sub> - doped UO<sub>2</sub> have been measured and compared with undoped UO<sub>2</sub>. Niobium acts as a donor impurity in UO<sub>2</sub>, but is not completely ionized and does not lead to n-type extrinsic behavior. On the contrary, intrinsic conductivity is observed and this is attributed to Nb compensating for acceptor levels due to slight excess oxygen nearly stoichiometric UO<sub>2</sub>. The value of the band gap in UO<sub>2</sub> is measured to be 2.14eV in undoped material and 2.26 in doped UO<sub>2</sub> [15].

High temperature molar heat capacity of  $Nb_2O_5$  - doped  $UO_2$  are practically the same as that of  $UO_2$ , [16].

# 4. Beneficial effects

Using ELES\_2.1, [17], a new Romanian version of the Canadian ELESIM.MOD9 code, the behavior of a CANDU 43 fuel bundle type was estimated. The evaluation has been performed on external fuel elements for a typical overpower envelope power history.

The design nominal manufacturing characteristic values are used as input data, except for the U-235 content (0.9 %wt). At the same time, two kinds of grain size (10  $\mu$ m and 40  $\mu$ m) were used for comparison.

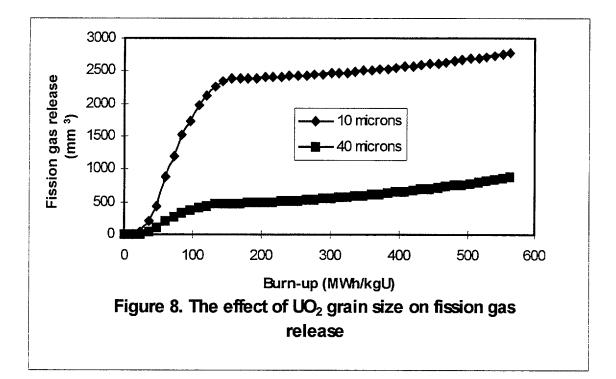


Figure 8 presents the evolution of fission gas release versus burn-up; it is observed that fission gas release is strongly affected by UO<sub>2</sub> grain size for a burn-up over 150 MWh/kgU. Fission gas release volume is smaller for large grain size.

A program covering the manufacturing, irradiation and post-irradiation examination (PIE) of niobia doped fuel at two dopant levels (0.25% and 0.4% by weight of Nb/U) was performed [18].

The fuel was manufactured by BNFL using binderless IDR/CONPOR route. The as-manufactured grain sizes were:

- 6-8 μm undoped pellets;
- 36 μm 0.25% Nb doped pellets;
- 45 μm 0.4% Nb doped pellets.

The fuel was irradiated in the commercial Dodewaard BWR (Netherlands) at various mean burn-ups:

- 25-26 MWd/kgU (4 cycles) fuel rods with 0.25% Nb doped pellets;
- 36-39 MWd/kgU (6 cycles) fuel rods with 0.4% Nb doped pellets.

The post-irradiation examination was carried out by using poolside facilities at the Dodewaard reactor and the hot cells at Winfrith, Berkeley and Harwel. The fission gas release was determined by puncturing and krypton counting. The fuel rod puncturing showed the fission gas release for the niobia doped rods in all cases to be lower than their sibling undoped rods. The difference was a factor of 1.4 to 2.1 for the fuel rods with 0.25% Nb doped pellets and 1.1 to 1.2 for the fuel rods with 0.4% Nb doped pellets.

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# CARA, NEW CONCEPT OF ADVANCED FUEL ELEMENT FOR HWR

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#### Abstract

All Argentinean NPPs (2 in operation, 1 under construction), use heavy water as coolant and moderator. With very different reactor concepts (pressure Vessel and CANDU type designs), the fuel elements are completely different in its concepts too. Argentina produces both types of fuel elements at a manufacturing fuel element company, called CONUAR. The very different fuel element's designs produce a very complex economical behavior in this company, due to the low production scale. The competitiveness of the Argentinean electric system (Argentina has a market driven electric system) put another push towards to increase the economical competitiveness of the nuclear fuel cycle. At present, Argentina has a very active Slightly Enriched Uranium (SEU) Program for the pressure vessel HWR type, but without strong changes in the fuel concept itself. Then, the Atomic Energy Commission in Argentina (CNEA) has developed a new concept of fuel element, named CARA, trying to achieve very ambitious goals, and substantially improved the competitiveness of the nuclear option. The ambitious targets for CARA fuel element are compatibility (a single fuel element for all Argentinean's HWR) using a single diameter fuel rod, improve the security margins, increase the burnup and do not exceed the CANDU fabrication costs. In this paper, the CARA concept will be presented, in order to explained how to achieve all together these goals. The design attracted the interest of the nuclear power operator utility (NASA), and the fuel manufacturing company (CONUAR). Then a new project is right now under planning with the cooperation of three parts (CNEA - NASA - CONUAR) in order to complete the whole development program in the shortest time, finishing in the commercial production of CARA fuel bundle. At the end of the this paper, future CARA development program will be described.

#### 1. INTRODUCTION

In Argentina there are two operating NPPs, Atucha and Embalse in operation. Both used pressurized heavy water as coolant and are fueled with natural uranium, but they have very different engineering solutions for the primary system. Embalse is a standard CANDU 6 reactor (horizontal pressure tube, see figure 1), and Atucha I have vertical fuel channels inside a pressure vessel reactor (Siemens design, see figure 2). Then, nuclear fuel elements are strongly different (see figures 3 and 4). Embalse uses a national developed CANDU 37 rods fuel element, and Atucha uses a long bundle similar to PWR rod type, both supplied by a private owned fuel manufacturing company named CONUAR. That situation of diversified production leads to several complications from the point of view of the production at commercial level, specially when the competitiveness is a central task in electricity generation costs.

Present CANDU fuel is a very active design, with a new bundle generation (CANFLEX) following the present LWR trends, enabling to reach higher burnup with smaller rod diameter and, consequently lower central temperature, linear and surface heat generation rate. Taking account that the argentine electric system dispatch for fuel marginal cost, is reasonable to think about a new generation of advanced nuclear fuels. These fuels must lead, in a first approximation, the most ambitious points mentioned above but with an additional requirement: one fuel element for both types of NPPs and to achieve the smallest costs at small scale commercial production.

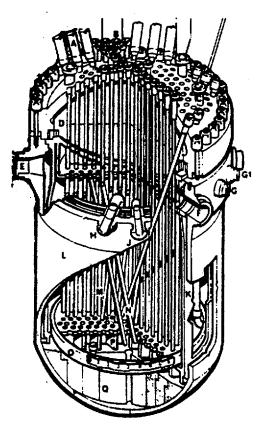


FIG. 1: Atucha I pressure vessel type reactor.

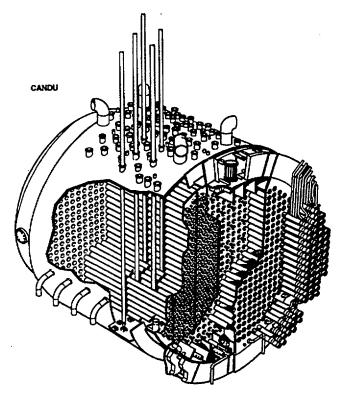


FIG. 2: CANDU reactor.

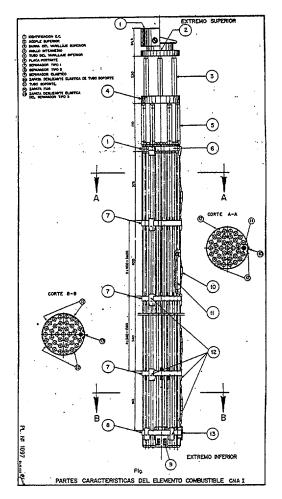


FIG. 3: Atucha I fuel element.

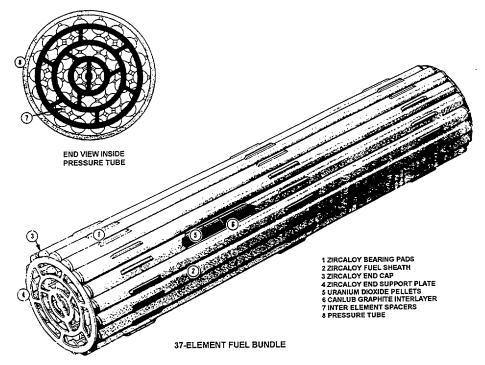


FIG. 4: CANDU fuel element.

#### 2. INITIAL POINT OF VIEW

Instead of small changes in fuel element improvements, we analyze the feasibility study of a completely new fuel element, for both types of NPPs, name CARA (Combustible Avanzado para Reactores Argentinos). This fuel element was set up with the following objectives:

- 1 Use the same fuel for both NPPs.
- 2 Increase the heat transfer area.
- 3 Use a single type of fuel rod diameter.
- 4 Decrease the fuel center temperature.
- 5 Decrease the relation of masses between Zry and Uranium.
- 6 Keep the Uranium contents of the more dense fuel.
- 7 Do not change the hydraulic pressure drop per channel of each NPP.
- 8 High burnup using SEU (Slightly Enriched Uranium)
- 9 Do not exceed the fabrication cost of the CANDU fuel.

Due to the CANFLEX experience, which have some of the same objectives, but with two different rod diameters, CARA fuel must explore new choices. If the number of fuel rods is increased, keeping constant the core pressure drop, the distributed friction will be necessary increased due to the smaller hydraulic diameter. These facts necessary leads to CANFLEX type design, loosing a single rod diameter (condition number 3), or use one type of rod diameter loosing uranium mass (condition number 6). Other option is to change the concentrate pressure drop in order to balance the higher distributed friction with the same uranium mass.

## 3. DEFINITION OF THE NUMBER AND LENGTH OF THE FUEL RODS

Analyzing the pressure drop of an Embalse fuel channel [2], and subtracting the distributed pressure drop [3, 15], we could find the concentrated pressure drop of one CANDU bundle (end plates and spacers). In reference [4] several changes in the CANDU fuel are studied, and using these data, pressure drop of the end plates and the spacers in present CANDU 37 rods bundle could be calculated and checked with our own data set.

As a considerable pressure drop is concentrated in the end plates, if we consider a bundle with twice of length of the usual CANDU fuel (without compatibily problems with CANDU 6 refueling machine), we could decrease de hydraulic diameter, increasing the number of rods.

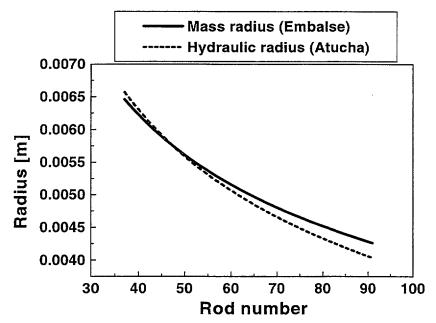


FIG. 5: Curve with the "mass" and "hydraulic" radius of CANDU and Atucha I

To check this approach, two type of curves were developed for a given fuel channel, in which the radius was changed in order to keep constant the uranium mass (mass constant curve) or hydraulic pressure drop ( $\Delta p$  constant curve) for different number of rods. Clearly both curves monotonally decrease for higher rods diameters, and if standard CANDU 6 bundle is taken as a basis, both curves have the same radius at 37 rods number. At higher rods number, if the  $\Delta p$  is keep constant uranium mass must decrease (mass constant curve above the hydraulic pressure drop curve).

But if a twice length bundle is used, both curves cross at 66 rods number. The  $\Delta p$  gain for the end plates number reduction, gives a  $\Delta p$  credit for the distributed friction together with a new concept for spacer function with only 33 % pressure drop reduction. As CARA fuel element must be compatible with two types of fuel channels, the most restrictive curve must be used for mass and  $\Delta p$  curves. Atucha has the most restrictive  $\Delta p$  requirements, and Embalse has the most restrictive mass requirements. Using both curves, with 1 meter long bundle, approximately 50 rods could be used for both NPPs, as is shown in figure 5.

#### 4. GEOMETRY OF THE BUNDLE

For the study of different bundles geometries, symbolic algebra languages enable very simple and fast evaluation; including rod ring rotation and different central rods number. Using this type of approach, 4 final geometries were studied, with 48 to 52 fuel rods. Finally a 52 bundle geometry was chosen, due to good simetry and compactness of the array. This geometry, shown in figure 6, have 4, 10, 16 a 22 rods per ring. The reduction of number or plugs and end plates gives a uranium credit that could be used to increase the bundle uranium mass.

The corresponding pellet diameter is very similar to present smallest CANFLEX radius, but with slightly greater clad thickness and present gap clearance of CANDU 37 rods bundle.

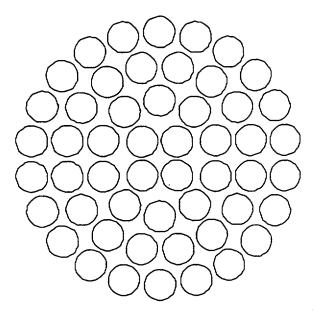


FIG. 6: Geometry of the CARA bundle with 52 fuel rods

## 5. NEUTRONIC CALCULATIONS

Using WIMS D/4 [8] as a neutronic cell code, the burnup could be estimated using the cell reactivity evolution, together with the power peaking factor [7], if the proper power densities, dimensions and geometrical buckling are used. To estimate these results, the burnup could be calculated as the burnup who equalized the mean reactivity of core of an average cell to the required excess reactivity for operation.

TABLE 1. WIMS RESULTS, NEUTRONIC DIFFERENCES BETWEEN THE CARA FUEL
ELEMENT FOR CANDU (CNE) AND ATUCHA I (CNA I).

Characteristic	CANDU 37	Atucha I	CARA (in the CNE)	CARA (in the CNA I)
Natural Uranium				
Burnup [MWd/THU]	7500	6100	7529	6368
Peak Factor [a.U.]	1.1261	1.0936	1.1359	1.07791
Uranium .9%				
Burnup [MWd/THU]	14537	13466	14576	14524
Peak Factor [a.U.]	-	_	1.1484	1.0838

The BOL excess reactivity, power peaking factor and burnup level could be seen in table 1, for natural uranium or SEU rods, for both types of Argentinean NPPs.

Using the power evolution, burnup level and peaking factor calculated with WIMS, together with all the geometric and compositions, a complete thermomechanical behavior could be calculated for the most restrictive CARA rods.

## 6. THERMOMECHANICAL BEHAVIOR OF FUEL ROD

The power history for a CANDU fuel used for calculation is included in the Figure 7. The power history sketched reach high power (and then high temperature) and it includes two shutdowns with a step-by-step increase in the power level after the second shutdown. This hypothetical, but realistic, power history was defined with demand conditions of irradiation for a real fuel element and for the BACO code simulation [9 and 10]. Starting with that power history we extrapolate the respective history for the equivalent CARA fuel conditions in a CANDU reactor correcting by the neutronic cell calculation model. The extrapolation is based on the burnup extension and the adaptation of linear power levels of the CARA fuel. The extension in burnup is 15000 MWd/tonUO<sub>2</sub> and the linear power is reduced up to a 72 % of the original value.

The Figure 8 represents the BACO code output for the temperature center of the  $UO_2$  pellet for a CANDU fuel rod and for the equivalent CARA fuel using its associated power history (Figure 7). CARA fuel allows a decrement of temperature of 500°C at the maximum power level. The Figure 9 represents the local power history of the seventh axial segment of a 5 meter long Atucha I fuel element (from the top of fuel and taking account ten axial segments). The seventh segment is the axial section most demanded during irradiation, it includes a maximum power level of 547 W/cm. The CARA fuel extrapolation corresponds with the forth module of a CARA assembly in Atucha I (the forth CARA module is equivalent with the seventh Atucha segment). The burnup at end of life is 14750 MWd/tonUO<sub>2</sub> and the power level is reduced a 73.4 % of the original Atucha fuel value.

The maximum temperature for the Atucha fuel at the pellet center is 1850°C during the maximum power level (see Figure 10). The calculated temperature for the equivalent CARA module is 1340°C, that is a decrease of 510°C.

The BACO code calculations show: temperature decreasing, smallest fission gas release, no restructuring and no central hole, lowest thermal expansion, and finally a best tolerance of the CARA's dimensional parameters. This allows to best manufacturing tolerance with an improvement in the dishing and shoulder of the pellet, and small plenum.

BACO code validity is sustained with the participation in the IAEA's Co-Ordinated Research Project on Fuel Modelling at Extended Burnup - CRP FUMEX [12, 13 y 14], the Atucha and CANDU experience, irradiation provided by international literature and own experimental irradiation.

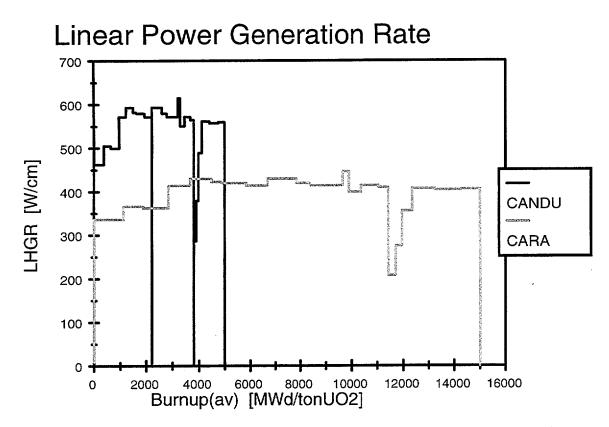


FIG. 7: Averaged power history for a CANDU fuel rod and the equivalent history for the CARA fuel in CANDU NPP.

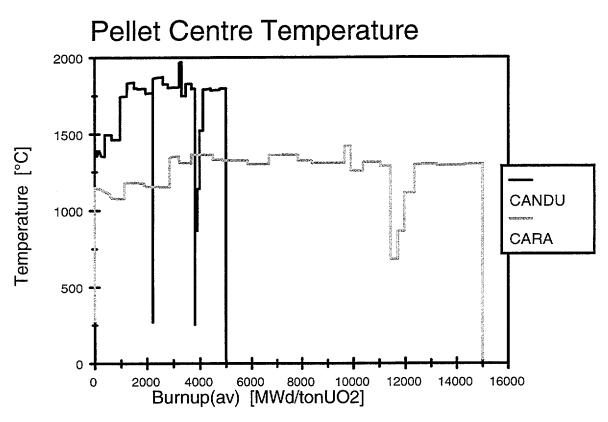


FIG. 8: Averaged temperature at the pellet center of a CANDU fuel rod and the temperature for the equivalent CARA fuel.

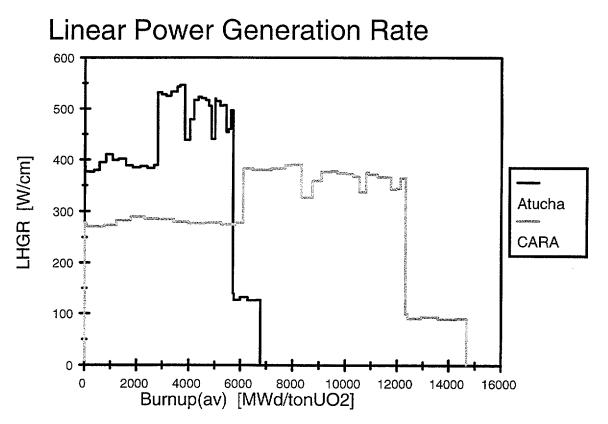


FIG. 9: Local power history for the seventh segment of a fuel rod of the Atucha I NPP and the equivalent history for the CARA fuel in Atucha I.

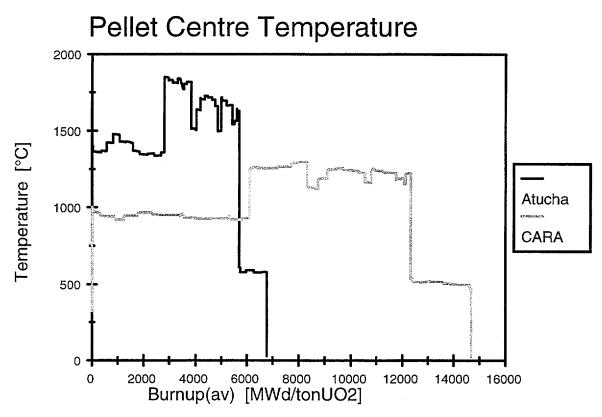


FIG. 10: Local temperature in the seventh segment of a fuel rod for Atucha I and the temperature for an equivalent module of the CARA fuel.

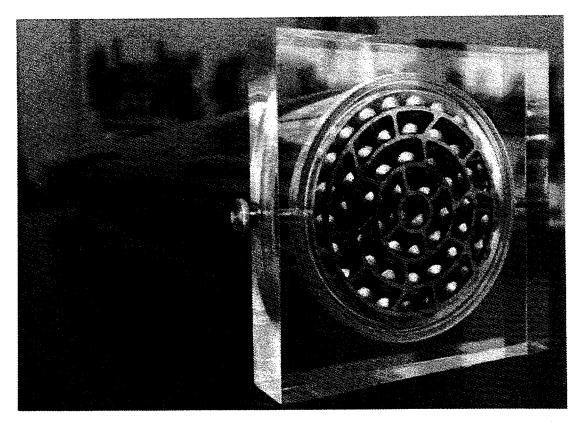


FIG. 11: Picture of the CARA fuel element.

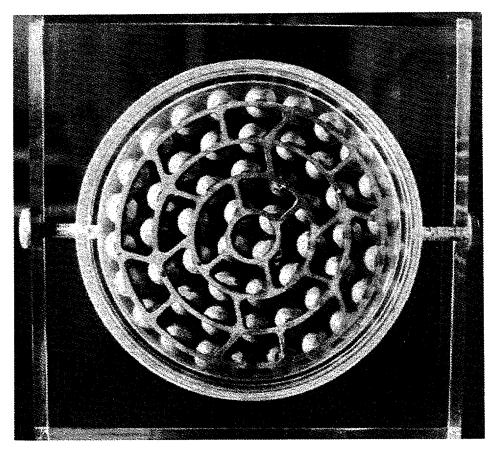


FIG. 12: Picture of the CARA fuel element.

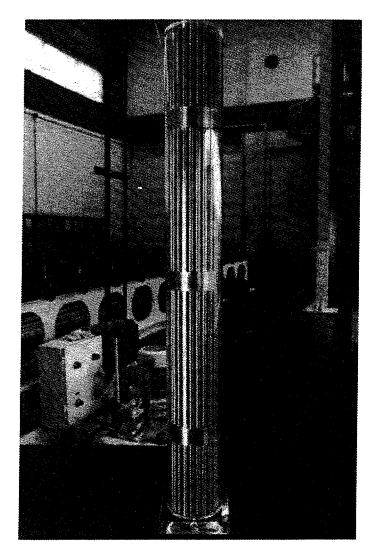


FIG. 13: Picture of the CARA fuel element.

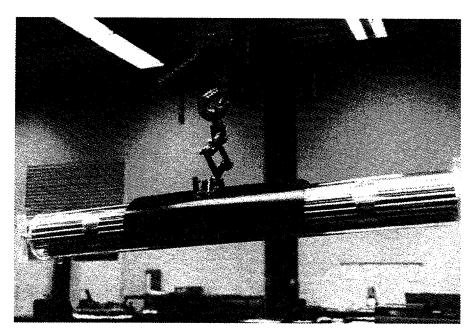


FIG. 14: Picture of the CARA fuel element.

#### 7. CARA DEVELOPMENT PROJECT

The CARA design, attracted the interest of the nuclear power operator utility in Argentina (NASA), and the fuel element manufacturing company (CONUAR). Then a new project is right now under planning with the cooperation of three partners (CNEA - NASA - CONUAR) in order complete the whole development program in the shortest time, finishing in the commercial production of CARA fuel bundle for both type of reactors.

The strong economics advantages of the new fuel, together with the excellent experience for the close to commercial SEU Atucha program, put strong incentives for the fastest fuel development up to commercial level.

Present CARA project, including an Atucha and Embalse program irradiation and post irradiation analysis, looks for an ambitious tasks of 4 years of time span.

At present, the design and independent verification analysis have included mechanical, neutronic, hydraulic and termohydraulic computer code calculations (including sub channel models), using tools already validated in the SEU project.

For Atucha channel assembly, for types of Atucha bundles options are available, using all the options five CARA bundles compatibles with CANDU 6, with minor changes in the end plates for both reactors. These changes in end plates do not affect the standardization costs reduction because the end plate welding is the latest step in bundle fabrication.

One demonstration bundle have been build by CONUAR, that could be seen in figures 11, 12, 13 and 14, and too much activities are now under way in CARA development. All the development centers of CNEA are now involved in CARA project, together with CONUAR. The official presentation of the project have been done in CNEA, and technical meetings with NASA and the National Nuclear Regulatory Authority have been done, with excellent results and prospect for the CARA project

#### 8. CONCLUSIONS

The feasibility study of an advanced SEU fuel element, compatible with CANDU 6 and Atucha type reactor have been successfully done, using a single rod diameter, as an essential task for economic production in Argentina. Then a new project is very advanced with the strongest commitment of the utility and the fuel element fabrication company. The condition of the present project is to developed the CARA fuel element at the shortest time, finishing with the commercial production of CARA bundles.

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## DEVELOPMENT OF ROMANIAN SEU-43 FUEL BUNDLE FOR CANDU TYPE REACTORS

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**Abstract** - SEU-43 fuel bundle is a CANDU type fuel consisting of two element sizes, to reduce element ratings, while maintaining the same bundle power, and an uranium content very close to the uranium content of a standard 37-element bundle. In order to reduce the detrimental effects of the life limiting factors at extended burnup a set of solution have been adopted for fuel element design. As a part of the design verification program, experimental bundles have been fabricated and utilized in typical out of reactor tests conducted at the laboratories of INR, Pitesti. These tests simulated current CANDU-6 reactor normal operating conditions of flow, temperature and pressure. The results are in accordance with the specified acceptance criteria.

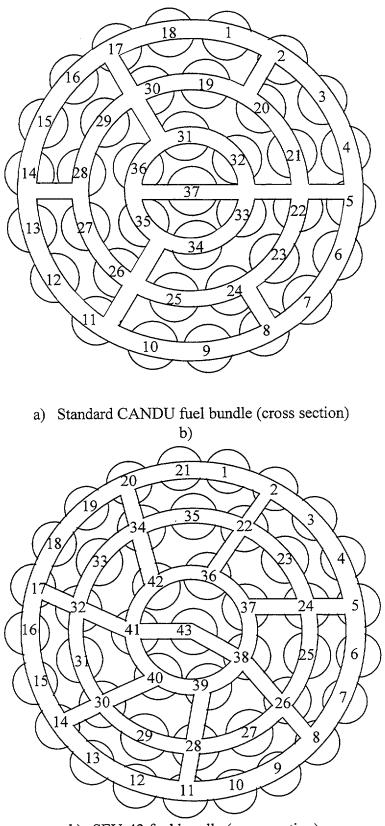
## 1. INTRODUCTION

The CANDU type reactor has the ability to accommodate a wide variety of fuel types. Slightly enriched uranium (SEU) fuel cycle can reduce total fuel cycle cost by 25 to 50% relative to natural uranium fuel cycle, depending on the costs of the uranium and enrichment[Boczar et al, 1987]. SEU fuel cycle offers several benefits which can be obtained by using existing technology. In order to meet extended burnup with SEU fuel in CANDU type reactors, one of the possibilities is a more subdivided bundle design, thus improving fuel general behaviour and its ability to withstand increased operating margins.

INR Pitesti has started in 1990 a general research programme aiming to develop a new fuel bundle for extended burnup operation. It was adopted a step by step strategy, in which every new step is based on the complete utilization of the results obtained in the precedent steps [Horhoianu et al, 1991; Horhoianu, 1992]. The actual version of the design is the result of a long process of analyses and improvements, in which successive preliminary design versions have been evaluated. The most relevant calculations performed on this fuel element design version are presented. Also, the stages of an experimental program aiming to verify the operating performance are briefly described.

## 2. SEU-43 BUNDLE DESIGN

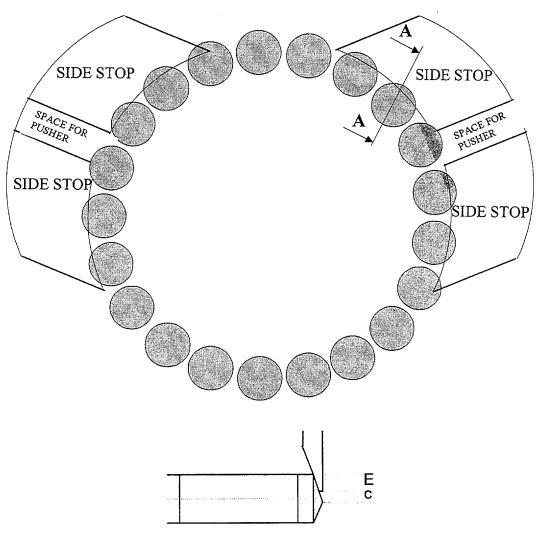
The major feature of SEU-43 bundle is an increase in the number of fuel elements from 37 in the standard CANDU-6 bundle to 43 elements. The SEU-43 bundle consist of 2 fuel element sizes: the 11.78 mm diameter elements in the outer ring, and the 12.40 mm diameter elements in the intermediate, inner and centre rings (see Figure 1). The small-diameter elements in the outer ring allow the peak element ratings in the bundle to be reduced by 16% in comparison to the standard 37-element bundle [Laslau and Serghiuta, 1991]. The larger-diameter elements in the inner rings of the bundle compensate for the fuel volume lost due to the smaller-diameter outer ring elements. To maintain compatibility of the new bundle with the existing CANDU-6 reactor systems, the basic overall dimensions of SEU-43 fuel bundle were designed to the same as those of the 37-element bundle. The small-diameter



b) SEU-43 fuel bundle (cross section)

Figure 1. Comparison of standard and SEU-43 fuel bundle design

elements of the outer ring results in a slightly eccentricity of the welded point fuel element end cap/end plate compared with the standard 37-element bundle. This makes the SEU-43 fuel bundle fully compatible with the side stop/separator assembly of the CANDU-6 fuelling machine. In this conditions an important parameter is the clearance between the bundle's endplate and sidestops, to make sure that there are adequate clearance for normal handling. At the same time, the "engagement" of the sidestops with the endcaps of the fuel elements has to be adequate to prevent damage of the fuel itself. These features are shown in Figure 2. The detailed design features of the bundle have continued to evolve as a result of ongoing design analysis and thermalhydraulics testing. The main specification of the SEU-43 fuel bundle are included in Table 1 and Table 2. Test programs are underway to demonstrate all hydraulic characteristics of the bundle and the irradiation behaviour of the fuel elements.



- Clearance "c" between side stop and endplate of bundle Figure 2. Interaction between SEU-43 and CANDU-6 fuelling machine

Item	Specification	
Structural material	Zircaloy-4	
Type of assembly	Welded bundles of 43 elements in circular	
	array with brazed appendages	
Length of bundle	495.30 mm	
Diameter of bundle	102.29 mm	
Weight (nominal)	23.40 Kg	
End plate thickness (min.)	1.58 mm	
End plate diameter	90.80 mm	

Table 1. Main characteristics of SEU-43 fuel bundle

The main characteristics of the SEU-43 fuel elements, resulted at the end of the design optimisation process are included in Table 2.

Item		Main specification	
		Inner ring fuel	Outer ring fuel
		element	element
Fuel rod	Total length, mm	492.65	492.65
	Active length, mm	477.30	476.85
	Axial gap, mm	3.00	3.50
	Diametral gap, mm	0.09	0.09
	Filling gas	80% He	80% He
Pellet	Diameter, mm	11.95	10.93
	Height, mm	12.57	11.95
	Material	UO <sub>2</sub>	UO <sub>2</sub>
	Enrichement U <sup>235</sup>	(1.2 % wt U235)	(1.2 % wt U235)
	Density, %TD	96.70	96.70
	Grain Size, mm	35.00	35.00
Cladding	Outer diameter, mm	12.40	11.78
	Thickness, mm	0.38	0.38
	Material	Zy-4	Zy-4

Table 2. Main characteristics of SEU-43 fuel element

# 3. FUEL ELEMENT DESIGN OPTIMISATION

The activity of defining feature of the SEU-43 fuel bundle and of the fuel element design has started with the review of the intended design objectives including the specific extended burnup design objectives. In order to reduce the detrimental effects of the life limiting factors at extended burnup a set of solution have been adopted for SEU-43 fuel element design (see Table 3) [Horhoianu et al, 1991; Moscalu and Horhoianu, 1991; Horhoianu and Moscalu, 1990].

Table 3. Design solutions for extended burnup fuel element

Design solution	Specific extended burnup design objectives
-Decreasing of sheath diameter	Decreasing of fuel element linear power and average fuel temperature
-Increasing of initial pellet grain size	Ensuring that fission gas release is within acceptable limits
-Increasing of pellet dish volume -Increasing of pellet land width -Increasing of pellet chamfer	Minimization of the local strain in the sheath ridges and reduction of SCC failure susceptibility in power ramps
-Increasing of axial gap -Increasing of graphite layer thickness	Accommodation of axial fuel stack expansion Reduction of SCC failure susceptibility

# 4. SIGNIFICANT ASPECTS REGARDING SEU-43 FUEL ELEMENT DESIGN EVALUATION

## 4.1 Design evaluation using a deterministic approach

The process of defining the new fuel element design have started with the identification of the extended burnup limitations of the actual fuel element design. The possible design solutions have also been reviewed taking into account the proposed design objectives [Horhoianu et al, 1991; Horhoianu, 1992]. The first step was the review of the design criteria and limits. As long as new extended burnup dependent phenomena have not been identified, the actual design limits for the CANDU type fuel can be maintained. Regarding the fuel element internal pressure, the actual limit (coolant outer pressure) which seems to be very restrictive with respect to extended burnup operation can not be modified or replaced due to specific thin collapsible sheath. So, an important design effort was directed for maintaining the internal pressure below the design limit [Horhoianu and Moscalu, June 1990].

After the initial stages of the design process in which were established the new bundle operational conditions (maximum power, average burnup, typical envelope power histories and coolant parameters), these conditions have been prepared as input data for the design codes. For assessing the influence of the selected design solutions on the fuel element performance, we have primarily used a conservative approach coupled with a comparison with the performance of the standard design in similar conditions. In the cases where the results of the conservative calculations exceeded the design limit, we have used the probabilistic alternative [Horhoianu and Moscalu, 1990]. We have performed detailed analyses for each fuel element performance parameter. The most significant results have been selected and are presented on [Horhoianu, 1992; Horhoianu and Moscalu, June 1990]. The value obtained for the highest fuel central temperature is below the design limit (melting temperature) and also is almost 200 K lower than the central temperature calculated in a standard fuel element in similar bundle power conditions. The cladding end of life circumferential deformation at mid-pellet position calculated in a conservative case (power envelope and the most unfavorable combination of input parameters) is only 0.43%, value which is placed below the design limit and is also in the range of experimental results obtained with standard CANDU type fuel elements.

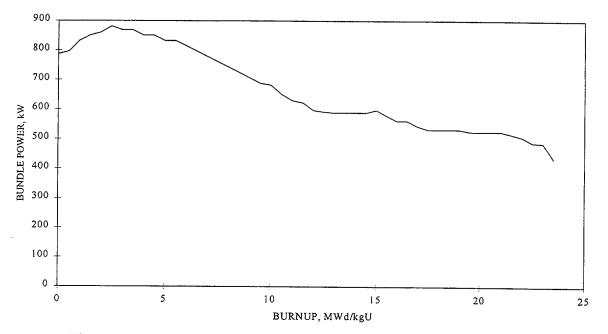


Figure 3. Utilized bundle power envelope - enriched CANDU type fuel

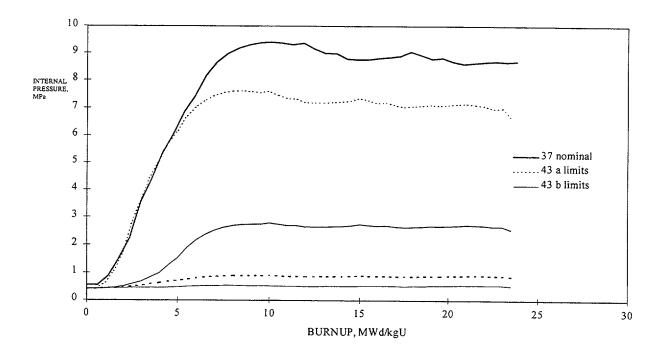


Figure 4. Calculated uncertainty bands of internal pressure for two fuel element designs

Figure 4 includes a series of successive results obtained in evaluating the evolution of outer fuel element internal pressure for three different situations. The curve denoted "37 nominal" represents the evolution of calculated internal pressure in the outer element of a standard geometry CANDU 37 bundle containing enriched  $UO_2$ . It can be noticed that the internal pressure for this nominal case is permanently very close to the coolant pressure. The "worst case" conservative calculation produced significantly higher values and was not figured. This curve was included as a reference for the next results. In the same figure are presented the calculated uncertainty band limits for the outer elements of the new 43 elements bundle design (noted 43a).

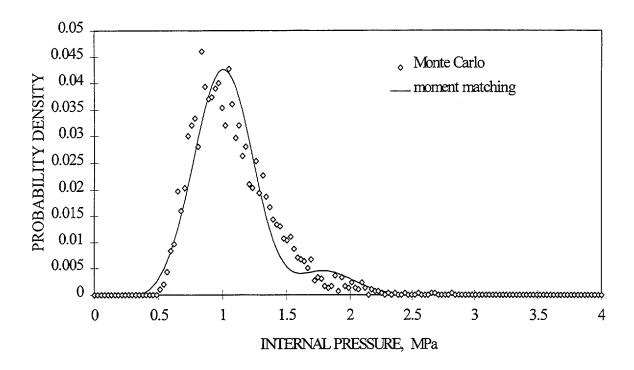
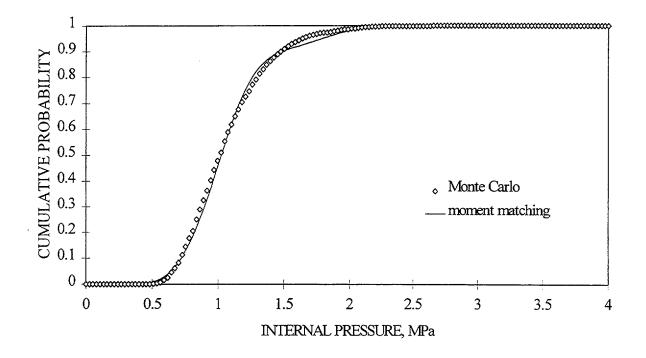


Figure 5. Calculated probability density function and cumulative probability for EOL internal pressure - 43b fuel element design



The local mechanical behaviour of the fuel cladding in typical power ramps, have been analyzed with the ROFEM code in various situations [Horhoianu and Moscalu, June 1990]. Parallel calculations have been performed on the standard fuel in similar bundle power conditions. For example, in the highest anticipated power ramp for the SEU refuelling scheme, the calculated values of the circumferential deformation at the pellet interface position for the SEU-43 element represents only a half of the deformation calculated for the standard design. This behaviour is a direct consequence of the modified pellet geometry.

### 4.2 Application of the probabilistic approach

Burnup extension is a process in continuing progress. Parallel investigations and experimental programs have accompanied this process and knowledge has increased about potential performance affecting phenomena associated with increased burnup. Fuel design analyses using current methodology often only allow limited burnup increases. Therefore effort has to be spent in order to find solutions that permit further increase of burnup. Besides fuel bundle and fuel element design improvements, the removal of excess conservatism from design criteria and from design calculations can contribute to this goal [Eberle et al, 1994]. The main means for eliminating excess conservatism from design calculations at extended burnup are: refining the models, improving the data base and utilisation of probabilistic design methods.

An example of the probabilistic methodology application to the evaluation of a particular CANDU type fuel design is presented in the following. An essential prerequisite for such an analysis is a best-estimate fuel element behavior modelling code that must incorporate an update of knowledge about potential performance limiting phenomena at extended burnup. The above mentioned version of ROFEM code includes such developments (e.g. burnup dependent degradation of  $UO_2$  thermal conductivity [Moscalu, 1995], burnup dependence of radial heat generation rate) and has demonstrated good prediction capabilities in FUMEX blind comparison exercise [Horhoianu et al, 1995]. The probabilistic methodology has been applied for evaluating the effect of variation of fuel element design data within their tolerances on calculated fuel performance parameters. The outer elements linear power histories utilised in this set of calculations were deduced from the envelope bundle power history presented in figure 3, which was calculated from core management simulations involving a 2-bundles shift refuelling scheme.

A version of this design, containing UO<sub>2</sub> pellets with a different grain size range ( $25-45 \mu m$ ) was investigated using the same approach. The calculated uncertainty band limits for this version are also figured (43b), showing a significant reduced range for internal pressure values. Other important additional results have been selected and included in figures 5. It shows the probability density function of EOL internal pressure for the 43b design, calculated using the two available methods on response surface equations. The second graph in figure 5 includes cumulative probability calculated for the same parameter, function that can be used directly for evaluating the probability of exceeding a given value (threshold). In figure 6 is presented the contribution to the variance of calculated EOL internal pressure (43b case) from 5 input parameters that have been selected in a RSM preliminary sensitivity analysis.

A version of this design, containing  $UO_2$  pellets with a different grain size range (25-45 µm) was investigated using the same approach. The calculated uncertainty band limits for this version are also figured (43b), showing a significant reduced range for internal pressure values. Other important additional results have been selected and included in figures 5. It shows the probability density function of EOL internal pressure for the 43b design, calculated using the two available methods on response surface equations. It also includes cumulative probability calculated for the same parameter, function that can be used directly for evaluating the probability of exceeding a given value (threshold). In figure 6 is presented the contribution to the variance of calculated EOL internal pressure (43b case) from 5 input parameters that have been selected in a RSM preliminary sensitivity analysis.

Another important advantage of this approach is the possibility of correctly establishing the "worst case" combination of the input variables for a given case and finding the associated probability of occurrence. This is the practical way of quantifying the conservatism of design calculations and also the basis for designs comparison.

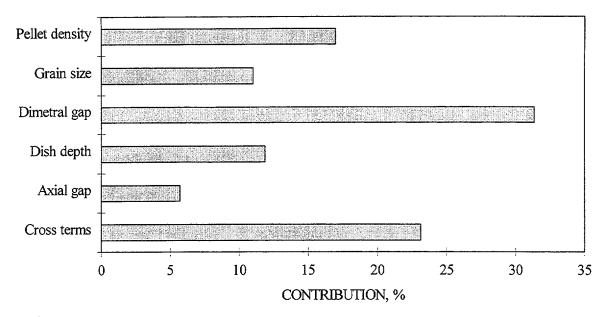


Figure 6. Contribution to EOL internal pressure variance - 43b fuel element design

### 5. FUEL-BUNDLE OUT OF PILE TESTING

A series of pressure drop tests have been performed to verify that the SEU-43 bundle meets the acceptance criteria specified for the pressure drop test and to provide relevant test data to evaluate the new fuel design [Doca and Doca, 1997]. The fuel-string pressure drop test was a light-water test. The pressure measurements were obtained for both SEU-43 bundles and reference 37-element bundles to compare the test results under the same test environment. The conditions for the pressure drop test were flow rates of 10 to 30 kg/s at a temperature of 100 to 289 °C, and fuel channel inlet pressure of 5 to 10.4 MPa. The test results for the SEU-43 and reference 37-element bundle strings are compared in Figure 7.

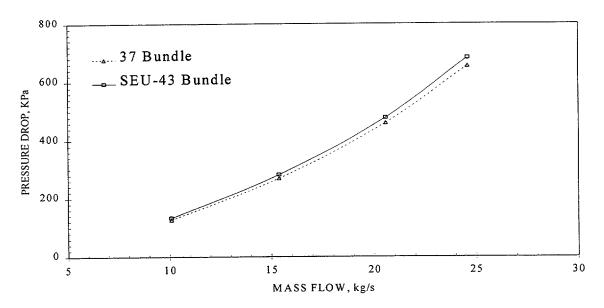


Figure 7. Comparison of 37 bundle and SEU-43 bundle pressure drops test water temperature  $200\ ^{\rm o}{\rm C}$ 

From a hydraulic point of view, the major difference between the 2 bundle designs is a change in the flow area and wetted perimeter. Statistical analysis of all experimental data suggests that the pressure drop for SEU-43 bundle (fully aligned) is about 3.8% higher than that for the 37-element bundles. A pressure drop test using Freon as a modelling fluid rather than light-water can offer the possibility to get more precise measurements of bundle junction pressure drop at Reynolds number closer to actual NPP reactor conditions.

Two flow-based strength tests are required for all bundle designs for CANDU reactors: a strength test to show that the bundle can support the hydraulic drag of a fuel string of bundles on the fuelling-machine sidestops without bundle damage, and a single sidestop test to demonstrate that the hydraulic force of a full channel of bundles can be carried by one sidestop. Both of these tests have been completed at INR. The results indicate that the bundle can withstand without problems the hydraulic forces. A refuelling impact test, simulating impact of a new fuel bundle coming to the rest against the stationary bundles in the channel, is also required. Also flow excitation test is required in order to investigate the vibration excitation characteristics of the small-diameter outer elements in the SEU-43 bundle. It can be anticipated that the small-diameter SEU-43 element had only a slightly lower natural frequency, and a slightly higher vibration amplitude, than a corresponding element of a 37-

Special tests are planned with both the SEU-43 bundle and the standard 37-element bundle geometry to study how the bundles interact with the sidestops under axial loads up to and beyond normal design load. To make these measurements and refine the design of the bundle a special "sidestop fixture" design and fabrication is planned. In this device a bundle could be positioned against sidestops. The final "acceptance" test for fuelling-machine compatibility involve the use of an actual CANDU-6 fuelling machine.

The final test required is a flow endurance test, to demonstrate acceptable vibration and fretting behaviour of the SEU-43 bundles in a fuel channel operating at a specified set of flow conditions.

### 6. IN REACTOR EXPERIMENTS

In order to demonstrate the performance of the SEU-43 fuel element design and to prove the adequacy of the manufacturing teghnologies, experimental fuel elements have been introduced in the irradiation devices of TRIGA, INR Pitesti, research reactor. One of the current objective of these fuel behaviour studies are to investigate and to reliably predict the performance during power cycling operation conditions.

The power cycling experiment has been performed in a special designed irradiation device, capsule C9 [Horhoianu et al, 1996]. The specified variation of fuel element linear power was obtained by mechanical movement of the device into the TRIGA reactor core. During the power cycling test the experimental fuel element has successfully experienced up to 367 power cycles, mostly between 50% and 100% of the specified linear power, pointing out the role of graphite coating in preventing SCC defects. PIE results indicate a maximum cladding strain at ridge of 0.7% in the region with the highest linear power. There are also indications of strong axial interaction between pellet column and end cap.

### 7. CONCLUSIONS

A development programme aiming to introduce extended burnup SEU fuel in our CANDU-6 type reactors has been started at INR Pitesti and the results from its early phases are already available. The following conclusions are obtained:

(i) The major feature of SEU-43 bundle is an increase in the number of fuel elements from 37 in the standard bundle to 43 elements. The SEU-43 bundle consist of two fuel element sizes.

(ii) A new fuel element design, which is included in the SEU-43 fuel bundle, has been developed using a set of selected design solutions able to reduce the failure susceptibility at extended burnup.

(iii) A calculation assessment on the final SEU-43 element version has been performed, pointing out the good performance at extended burnups in comparison with the standard design.

(iv) The probabilistic approach is extremely useful in the process of SEU-43 fuel design development and comparison. It can give a quantitative measure for the design differences and a measure for the importance of the tolerances specified for the fuel. This approach has also the capability of characterizing the degree of conservatism within fuel design analysis by making statements about the probability of occurrence of extreme values.

(v) As a part of the development programme, experimental SEU-43 bundles have been fabricated and utilized in typical out of reactor tests (pressure drop and axial compression). The results of the tests are in accordance with the specified acceptance criteria.

(vi) During the power cycling test the experimental fuel element has successfully experienced up to 367 power cycles.

(vii) A refuelling impact test and a flow endurance test are planned in the future. Also a fuelling-machine compatibility test will involve the use of an special device.

(viii) Fuel performance models and codes will be updated to incorporate the new experimental results to ensure continuing predictive capability for further developments.

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### DEMONSTRATION IRRADIATION OF CANFLEX IN PT. LEPREAU

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### Abstract

The demonstration irradiation of CANFLEX in the Point Lepreau Generating Station (PLGS) in New Brunswick, Canada, will mark a major milestone towards delivering this new fuel to CANDU utilities. One high-powered and one instrumented fuel channel are being fuelled with CANFLEX bundles to establish irradiation experience in a power reactor. As CANFLEX is discharged into the reactor bays, it will be examined by fuel experts from PLGS and AECL. Several irradiated CANFLEX bundles will be shipped to Chalk River for extensive post-irradiation examination. CANFLEX is the latest fuel carrier in the evolution of CANDU fuel. Its design has been driven to provide higher dryout powers and lower peak element ratings, while being fully compatible with existing CANDU stations and addressing the development requirements for future advanced CANDU stations. The design has been tuned through analysis and testing at AECL and KAERI. The final CANFLEX design has undergone extensive analysis, performance testing and critical industry review. Safety performance has been analyzed and documented in a licensing submission to the Canadian regulator, the Atomic Energy Control Board, for approval to proceed with the demonstration irradiation. Because CANFLEX is fully compatible with existing plants, CANDU 6 stations can simply substitute CANFLEX-NU for 37-element fuel and achieve improved reactor operating and safety margins, and higher critical channel powers. For CANDU designers, CANFLEX provides the opportunity to benefit from the use of slightly enriched uranium (SEU) or recycled uranium (RU) from reprocessed spent PWR fuel. Enrichment can be used in one of several ways: to increase the power from a given reactor core size through flattening the radial channel power profile; to increase the fuel burnup and reduce the quantity of spent fuel; to improve fuel cycle economics, both front- and back-end; and, in general, to provide greater flexibility in reactor design.

### **1. INTRODUCTION**

CANDU® fuel has evolved from seven element fuel bundles in the NPD reactor, through nineteen element in the Douglas Point reactor, twenty-eight element in the Pickering Nuclear Generating Station, to the current thirty-seven element in CANDU 6 and Bruce and Darlington plants. The 43-element CANFLEX® bundle is a logical extension in this evolution. Atomic Energy of Canada Limited (AECL) has been developing CANFLEX since 1986 [1]. In 1990, the Korea Atomic Energy Research Institute (KAERI) and AECL completed a joint study on the potential use of CANFLEX in CANDU reactors to provide greater fuel performance and greater fuelling flexibility. Since 1991, AECL and KAERI have pursued a collaborative program to develop, verify and prove the CANFLEX design. New Brunswick Power at the Point Lepreau Generating Station (PLGS) will irradiate 24 CANFLEX fuel bundles over a 2-year period starting in 1998, as a final verification of CANFLEX design in preparation for full-core conversion. This paper describes the CANFLEX program that has driven the design from concept to proven fuel.

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### 2. CANFLEX FEATURES

The principal features of CANFLEX are enhanced thermalhydraulic performance and more balanced radial power distribution, providing CANDU plant operators with greater operating flexibility through improved operating margins [2,3,4]. Critical heat flux (CHF) enhancement appendages on the CANFLEX bundle enable a higher bundle power before CHF occurs, leading to a net gain in the critical channel power of 6% to 8% over the existing 37-element fuel. The maximum linear element rating in a CANFLEX bundle is 20% lower than that of the conventional bundle, reducing the consequences of most design-basis accidents. The lower element rating is achieved by adding extra elements and using larger diameter element in the 2 centre rings and smaller diameter ones in the outer 2 rings [5].

CANFLEX has been designed to have hydraulic and neutronic characteristics that are similar to those of the existing fuel. This feature provides the operators with the ability to introduce CANFLEX bundles during normal on-power refuelling. No hardware changes are required to switch to CANFLEX fuel because CANFLEX is fully compatible with existing fuel handling equipment. Fuel channels containing both CANFLEX and 37-element fuel, in any combination that can occur with normal fuelling, have improved or unchanged operating margins. Transition to CANFLEX fuel can be gradual with no waste of existing fuel.

In addition to providing greater operating margins, the CANFLEX bundle facilitates the use of slightly enriched uranium (SEU) and recycled uranium (RU) [6,7]. CANFLEX-RU offers lower fuelling costs and provides a means to raise reactor power within a fixed core size. The use of RU from LWR reactors promises to be more economical than SEU or NU. The use and economics of RU are being assessed in a collaborative program with British Nuclear Fuels Plc (BNFL).

### 3. CANFLEX VERIFICATION PROGRAM

The CANFLEX bundle has undergone an extensive verification program [8]. The verification program has been conducted under the strategy laid out in the Design Verification Plan (DVP). The verification work consisted of analysis and testing drawing on the capabilities of AECL's facilities in Canada and KAERI's facilities in Korea.

The DVP identifies the performance requirements, specifies the test or analysis required to verify that the requirement is met, and identifies responsibility and procedures. All testing and analysis were performed under the quality standard CAN/CSA-N286.2 [9] or equivalent. The DVP called for preparation of a Test Specification, Test Procedure, Acceptance Criteria and identified the required documentation. The following sections provide a short summary of the main verification activities.

### 3.1 CHF- dryout power testing

The linear dryout power (LDP) is often used to compare the relative difference in dryout power characteristics between fuel bundles of different designs. The ratio of LDP accounts for the differences in heated surface area, flow area and flux distributions, and permits a direct comparison of dryout powers for a given flow rate. In equation form, the ratio is expressed as

$$\left(\frac{LDP_{43}}{LDP_{37}}\right) = \left(\frac{P_{k_{43}}}{P_{k_{37}}}\right) \left(\frac{CHF_{43}}{CHF_{37}}\right)$$

where

 $LDP_{43} = LDP$  for a CANFLEX bundle (kW·m<sup>-1</sup>),  $LDP_{37} = LDP$  for a 37-element bundle (kW·m<sup>-1</sup>),  $CHF_{43} = CHF$  in a CANFLEX bundle (kW·m<sup>-2</sup>),  $CHF_{37} = CHF$  in a 37-element bundle (kW·m<sup>-2</sup>),

 $P_{h_{37}}$  = heated perimeter of a 37-element bundle (m), and

 $P_{h_{43}}$  = heated perimeter of a CANFLEX bundle (m).

CHF experiments were performed by Dimmick et al. [10] in Freon-134a in the MR-3 facility at the Chalk River Laboratories (CRL), on both the 37-element and the CANFLEX simulated fuel strings. The tests were performed on 2 new full-scale simulated fuel strings, constructed according to a strict Q/A protocol, and instrumented with moveable dryout thermocouples in every rod. CHF measurements were obtained with the bundle string located in both an uncrept channel and a channel having a uniform diametral creep of 3.1%. The results are summarized in Figure 1. The average

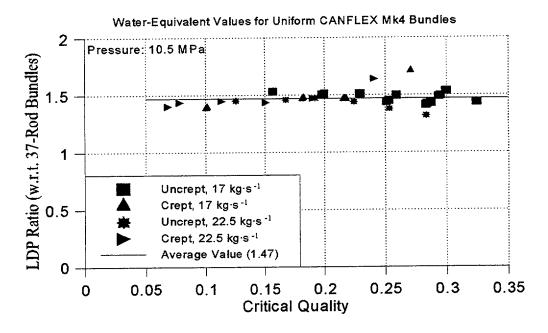


Figure 1: Linear Dryout-Power Enhancement Ratio for the CANFLEX Fuel Bundle With Respect To the 37-Element Bundle

increase in LDP (for a given critical quality) was about 45% for both the uncrept flow channel and the 3.1% crept flow channel tested. Based on this result from the Freon testing, the CANFLEX bundle is estimated to enhance critical channel power by 6% to 10% for a reference channel in a typical CANDU 6.

To license a CANDU 6 reactor with a full core of CANFLEX at a higher critical channel power, water CHF data are required. AECL and KAERI have initiated a contract with the Stern Laboratories in Canada to generate the licensing data. These data will determine the additional operating margin available with CANFLEX.

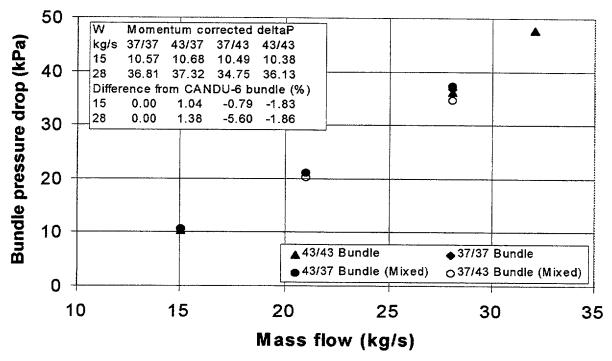


Figure 2: Pressure Drop for 37-, 43-, 37/43- and 43/37-Element Bundles.

### 3.2 Pressure drop testing

The pressure drop characteristics of the CANFLEX bundle were determined in both freon tests and hot and cold water tests. KAERI tested a full string of CANFLEX bundles and 37-element bundles in their hot test loop at normal reactor operating conditions [11]. AECL studied the axial pressure profiles for CANFLEX bundles in the freon MR-3 facility. Mixed junction of 37/43 bundles and the effect of bundle rotation were studied.

Hameed [10] compared the results of the CANFLEX bundles with the corresponding results of the 37-element bundles. The measured bundle pressure drops are shown in Figure 2. The results indicate that, for the same flow conditions, the CANFLEX bundle will reduce the adiabatic pressure drop by about 2% for both the uncrept and the crept channels.

### 3.3 Out-reactor flow testing

Over the last several years AECL and KAERI have subjected the CANFLEX fuel bundle to a set of out-reactor flow tests to simulate reactor conditions and verify that the design is compatible with existing reactor hardware [11]. In addition to the heat transfer and pressure drop tests the following mechanical flow tests have been successfully completed:

• Strength Test: Strength tests showed that the fuel can withstand the hydraulic loads imposed during refuelling. Post-test bundle geometry measurements showed no significant distortion.

• Impact Test: Impact tests showed that CANFLEX bundle can withstand bundle impact during refuelling.

• Cross Flow: Cross flow tests demonstrated that, during refuelling, when the bundle is in the cross flow region, the bundle withstands the flow-induced vibration for a minimum of 4 h.

• Fuelling Machine Compatibility: Fuelling machine compatibility showed that the bundle is dimensionally compatible with the fuel handling system [12].

• Flow Endurance: Flow endurance test demonstrated that the CANFLEX bundle maintains structural integrity during operation; fretting wear on the bearing pads, inter-element spacers and pressure tube remain within design limits over the 3000-h test time.

## Table 1: In-Reactor Irradiation Requirements and Results

Requirements Imposed by	Attribute to be Demonstrated	Irradiation Status
Primary Coolant System	<ul> <li>elements contain fission products;</li> <li>withstand coolant pressure</li> </ul>	AJK successfully irradiated in NRU at ~70 kW/m to 372 MW•h/kg U. Performance acceptable.
Fuel Channel	<ul> <li>bundle compatibility;</li> <li>accommodate</li> <li>expansion, PT sag and creep</li> </ul>	All CANFLEX bundles removed from PT in NRU without problems. Profilometry acceptable
Fuelling System/Physics	Change in power with refuelling	Bundle AJM subjected to power increase in NRU. Performance acceptable.
Fuel Management	<ul> <li>withstand continuous high power,</li> <li>-power changes by ripples,</li> </ul>	AJK - 372 MWh/kgU, ~70 kW/m AJM -450 MWh/kgU, ~65 kW/m AJN - 468 MWh/kgU, ~69 kW/m Performance Acceptable Bundle power envelopes bound
	reactivity shims and refuelling sequences	peaks Performance Acceptable
	- end flux peaking	PIE showed AJK withstood end flux peaking. Performance Acceptable.

### 3.4 In-reactor testing

CANFLEX bundles, AJK, AJM and AJN, were irradiated in the U-1 and U-2 loops in the NRU research reactor to demonstrate performance under expected in-reactor conditions. Typical power changes during refuelling and peak bundle powers during operation were calculated to establish the irradiation conditions for the NRU tests; actual peak powers experienced were over 25% greater than in a CANDU 6. Once the bundles were removed, detailed post-irradiation examination (PIE) was performed. The irradiation requirements and corresponding results are provided in Table 1. All design requirements were met.

### 3.5 Reactor physics testing and analysis

CANFLEX natural-uranium bundles were inserted into the ZED-2 facility at CRL to measure the fine-structure reaction rates, to validate the reactor physics lattice code WIMS-AECL [13]. The data showed excellent agreement with code predictions. Reactor operation over 600 full-power days was simulated to determine peak bundle powers, power changes during refuelling, burnups, and residence times. Various fuel schemes were studied. Fuel performance requirements were established for NRU irradiation tests. The analysis showed that the CANFLEX bundle meets or exceeds all power requirements.

### 3.6 Structural analysis

The CANFLEX design was analyzed for sheath strains, fission-gas pressure, end-plate loading, thermal behavior, mechanical fretting, element bow, end-flux peaking, and a range of other mechanical characteristics. Acceptance criteria established from years of operating experience with 37-element fuel and previous 37-element testing were met by the CANFLEX design.

### 3.7 Formal design review

The final stage in the design verification was a formal review of the design by a Design Review Panel. The Design Review Panel was chaired by AECL's Chief Engineer and consisted of experts from the various disciplines relevant to fuel design and selected from the CANDU utilities and fuel fabricators as well as internal AECL experts not directly involved in the CANFLEX program. The results of the testing and analysis program were evaluated against the design requirements set out in the Design Verification Plan. All CANFLEX performance data were summarized in a Fuel Design Manual. The Panel reviewed the CANFLEX information, raised questions and concerns, which were formally addressed by the Design Team and recorded in a tabular form. Based on the disposition of these issues, the Chief Engineer certified the CANFLEX design for the Point Lepreau Demonstration Irradiation.

### 4. CANFLEX DEMONSTRATION IRRADIATION

The final step in the verification of the CANFLEX bundle before full-core implementation is a demonstration irradiation in a power reactor. CANFLEX has met or exceeded all design requirements in out-reactor tests and irradiation requirements from the NRU research reactor testing. All analyses have shown compliance with the design requirements. Throughout this design verification process, New Brunswick Power has monitored progress, reviewed results and recommended further work. A comprehensive economic assessment was jointly prepared by AECL and the Canadian CANDU 6 utilities, which showed that the additional operating margins available with CANFLEX provided a cost-effective method for offsetting reduced margins resulting from reactor aging. The PLGS has decided to implement a limited 24 bundle demonstration irradiation (DI) as a critical step towards a decision to implement full-core CANFLEX fuelling. This section describes the further reviews conducted by PLGS to secure approval to proceed with the DI.

### 4.1 Demonstration irradiation strategy and plan

Twenty-six CANFLEX bundles were fabricated by Zircatec Precision Industries, Canada, to the Quality Assurance levels normally applied to 37-element fuel supplied to PLGS. Twenty-four of these bundles will be fuelled into 2 channels at PLGS. All configurations of CANFLEX bundles mixed with 37-element bundle in a single channel during transition and full-core refuelling will be tested. The following objectives will be addressed:

- 1. Some fuel should be exposed to as high a power as possible within the allowable operating envelope.
- 2. Some fuel should be exposed to as wide a power variation as possible within the allowable operating envelope.
- 3. At least one channel should have normal dwell with a full CANFLEX fuel string.
- 4. Some fuel should experience normal fuelling-induced power ramps.
- 5. At least one selected channel shall be in the flow-assist-ram-extension region and one in the flow-assist-fuelling region.
- 6. Some fuel should experience the highest burnup that is possible within the allowable operating envelope.
- 7. Some fuel should experience the longest time in reactor that is possible within the allowable operating envelope.
- 8. Some fuel should be in an instrumented channel.
- 9. Some fuel should be exposed to the largest possible amount of acoustic excitation.

Two sets of suitable channels have been identified to meet these objectives. As one channel from each set becomes available for regular refuelling, the CANFLEX bundles will be inserted. The fuelling plan for each channel is as indicated in Figures 3 and 4.

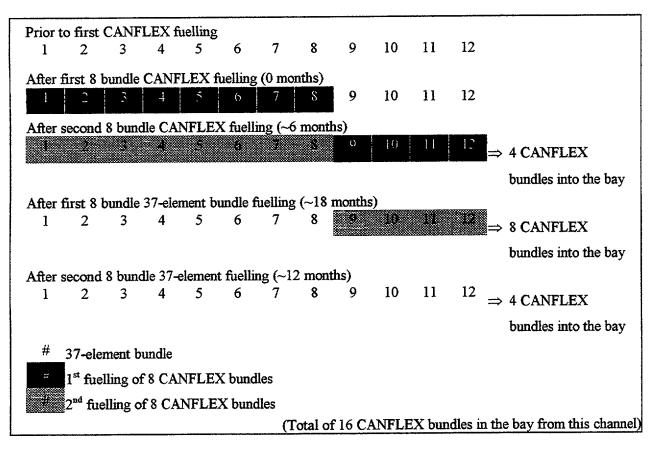


Figure 3: Projected Fuelling History for High-Power Channel

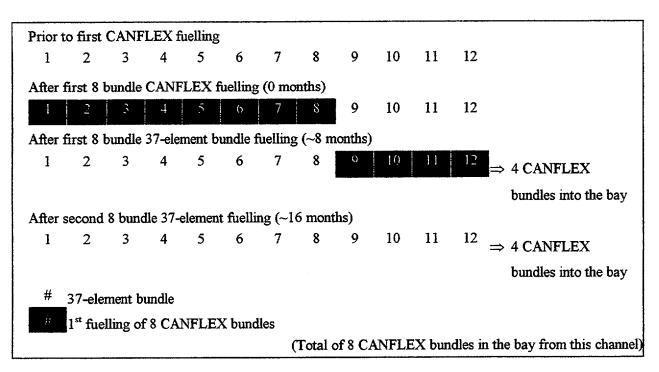


Figure 4: Projected Fuelling History for Low-Power Channel

As CANFLEX bundles are discharged and transported to the bays, they will be visually examined. Plans call for 3 bundles to be shipped to CRL for PIE as follows:

- A bundle from those first discharged from the high-power channel
- A bundle from the high-power channel that has received higher burnup, and
- A bundle from the lower powered channel that has seen longer residence times.

These bundles will be examined in the hot cells at CRL. The PIE will consist of

- Visual examination, bundle profilometry;
- Disassembly and element profilometry;
- Gamma scanning;
- Fission-gas and void volume measurements;
- End-plate weld and button weld strength tests;
- Metallography and ceramography;
- Chemical burnup analysis (high-performance liquid chromatography);
- Alpha, beta and gamma autoradiography; and
- Hydrogen analysis of sheath, button and end plate;

The DI will be fully documented, including PIE reports. Successful demonstration of the new CANFLEX design is critical in making a decision to implement full-core CANFLEX fuelling.

### 4.2 Safety analysis

To secure approval for the DI, PLGS and AECL have analyzed all design-basis accidents to determine CANFLEX performance. The principal design difference between the 37-element and the CANFLEX bundles are:

- 1. higher dryout power providing greater operating margin;
- 2. lower maximum linear element ratings leading to lower centreline temperatures and lower fission-gas release;
- 3. smaller diameter outer elements, which run at lower temperature but could be less rigid;
- 4. higher void reactivity because of the 5% increase in coolant cross-sectional area and greater bundle subdivision, which could lead to higher power increases because of void formation offset by lower initial powers; and
- 5. smaller Zircaloy mass but higher surface area, which could affect hydrogen production.

The safety studies have shown that CANFLEX will maintain acceptable safety margins for all postulated accidents. The results of these studies were summarized in an Information Report submitted to the Atomic Energy Control Board (AECB) requesting approval to proceed with the DI. This Information Report addressed all of the accidents considered in the PLGS Safety Report either through explicit assessment or through qualitative consideration of key design differences, and the other explicit assessments. The results of the explicitly considered design-basis accidents studied, are summarized in the following:

• Large-Break Loss-Of-Coolant Accident (LOCA): Analysis of large-break LOCAs with emergency core-cooling (ECC) available showed that the overpower transient of the CANFLEX bundle is similar to that of the 37-element bundle; fuel temperature will be lower; integrated energy deposited in the fuel is lower; probability of sheath failure is lower because of lower fission-gas release; pressure-tube-to-calandria-tube contact is reduced because of the lower element temperature; axial expansion is less; hydrogen production is moderately higher because of the higher surface area; and, bundle velocity is lower when subjected to reverse flow. In summary, CANFLEX will maintain or improve the safety margins.

• Large-Break LOCA/Loss-Of-Emergency Core-Cooling (LOECC): Analysis of an LOECC showed increased assurance of fuel channel integrity because of reduced subcooling

requirements, slightly reduced fission-product release, and slightly increased hydrogen generation because of the slightly higher Zircaloy surface-to-volume ratio and the higher power per unit mass of CANFLEX. Safety margins are maintained.

• Small-Break LOCA: Analysis of a 2.5% inlet header break showed that the peak fuel sheath temperature is 412°C, moderately higher than the 37-element bundle, but well below the 800°C acceptance criterion. The fuel channel temperature is 388°C, lower than 37-element and well below the limit of 600°C. Thus CANFLEX will maintain safety margins.

• Pressure Tube Rupture: Assessment of the effect of pressure tube rupture coupled with calandria tube rupture on fuel behavior showed significantly lower levels of fission-product release because of the lower free-fission-product inventory resulting from the lower peak linear power rating, pointing to improved safety performance of CANFLEX.

• Channel Flow Blockage: This analysis shows that a channel of CANFLEX bundles fails slightly later than does a 37-element bundle because of the lower outer-element powers. The amount of molten material is marginally higher (5%) because of the higher powers in the inner rings. The fission-product release is essentially the same since both the free and inter-granular inventories are assumed to be released in this accident scenario. Safety margins are maintained.

• Feeder Breaks: For the stagnation break, the timing of the fuel channel failure is moderately delayed, the volume of molten metal is marginally higher and the fission-product release is lower. For off-stagnation breaks, the fuel element temperatures are lower, fuel sheath failure occurs later, and the number of inner element failures is higher, and the fission-product releases are lower. The safety margins for this safety analysis are not eroded by CANFLEX.

• Loss of Forced Cooling: The time to reach fuel centreline melting is the same for the 2 bundle designs in power/flow mismatch situations, such as loss of Class IV electrical power or heat transport pump seizure. Safety margins with this event are maintained with CANFLEX.

• Loss-of-Regulation (LOR)- Loss-of-Reactivity Control: For power/flow mismatch situations such as slow LOR and fast LOR over a range of bundle burnups, the time to fuel centreline melting was shown to be either the same or slightly longer for the CANFLEX bundle. Again CANFLEX is shown to maintain safety margins.

• LOR- Loss of Pressure and Inventory Control: The time to fuel centreline melting for CANFLEX bundles was found to be very close to that of the 37-element design, again maintaining safety margins.

• Fuelling Machine Events: The explicit analysis covered events when the fuelling machine was off the reactor. The CANFLEX bundles in these events were shown to reach peak fuel sheath temperatures very similar to those of the 37-element fuel, again maintaining the safety margins.

### 4.3 Operational review

The PLGS conducted a detailed design review in part through AECL's formal design review but augmented by an internal review. The internal review paid particular attention to the reactor operation issues. Operating tools such as the reactor physics codes were modified to accommodate the CANFLEX bundles. Bundle power and channel powers were recalculated. Operational contingencies were developed to recommend to station staff appropriate responses to various postulated events. A plan was prepared to record pertinent data and information generated throughout the DI. Finally, issues, such an in-bay storage, emplacement of bundles in shipping flasks for shipment to CRL for PIE, were studied and where required, special procedures were drafted. When PLGS station management were convinced that all operational effects were understood and addressed, the request for AECB approval of Demonstration Irradiation was submitted.

### 4.4 Information report requesting AECB approval

The final approval to proceed with the DI must come from the AECB. PLGS prepared a comprehensive document referred to as an information report, which summarized the results of the AECL-KAERI development program, the design review, safety review and operational review. This document was submitted to the AECB on 1998 March 4. The AECB review is in progress with a target date for approval in June when Pt. Lepreau completes a maintenance shutdown.

CANFLEX has been under development for over 10 years. CANFLEX is a prime example of the results that can be achieved through collaborative ventures between Canada and Korea. CANFLEX has been verified through extensive testing by AECL and KAERI. The last crucial phase is the demonstration irradiation of 24 bundles in PLGS. Once successfully demonstrated, the CANFLEX fuel bundle design can be used in CANDU 6 reactors to provide greater operating flexibility, restore operating margins eroded by reactor aging and open opportunities to reduce fuelling costs and uprate fuelling using recycled or slightly enriched uranium. CANFLEX will have gone from a concept to a commercially available product.

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### NUCLEAR FUEL OPTIMIZATION AND PROBLEM OF INCREASING BURNUP AND COST EFFICIENCY OF LIGHT WATER REACTORS IN RUSSIA

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#### Abstract

Brief analysis is given of the results of WWER-1000 and WWER-440 fuel assembly (FA) and fuel rod operation to the average burn-up of 44 and 34 MW·day/kg U, respectively. The reliable operation of the fuel is noted which made it possible to outline the paths of further improvements in their fuel cycles. Based on a series of improvements reactors are being switched on a 4-year cycle; a 5-year cycle is contemplated for the WWER-440.

The fuel cycle of the boiling RBMK reactors is subjected to essential alterations. Specifically, the use of erbium oxide as an integral burnable absorber allows a higher enrichment of uranium and burn-up. To extend the burn-up a new zirconium alloy E-635 is assumed to be used as fuel rod claddings and other FA components; the alloy has higher corrosion and irradiation resistance.

### INTRODUCTION

The current technical level of the PWR and BWR fuel production and the perfected system of the reactor management have allowed to provide the successful operation of PWR NPP to the average burn-up of  $\sim$ 45 MW·day/kg U in all countries of developed nuclear power.

The differences in the reliability and FA serviceability as well as in the number of leaky fuel rods are rather small and the attention is usually not focused on this issue. More urgent and common is the tendency to the cost-efficiency of nuclear power as well as the reliability and safety of NPP. For the optimised fuel cycles of WWER NPP now in operation and new generation reactors that are under development a substantial increase of the burn-up to reach 60 MW day/kgU is contemplated also via the use of MOX fuel. This extended burn-up makes some problems more acute, the ones that at the current burn-up were more or less noticeable. Specifically, they comprise corrosion resistance and mechanical properties of fuel rod claddings; dimensional stability of FA; pellet cladding interaction (PCI), higher FGR etc.

Problems are also encountered that are primarily inherent in improving the safety as well as the cost-efficiency of boiling type reactors RBMK (BWR).

The nuclear power economics is much dependent on the degree to which the nuclear fuel cycle is optimized. In Russia from the time of the nuclear power evolution the closed nuclear fuel cycle concept was adopted. To implement the concept all the needed infrastructure, the spent nuclear fuel reprocessing plant included, was set up; this made it possible to close the fuel cycle in terms of uranium 15 years ago. This in its turn substantially reduced the demand for natural uranium.

### 1. BRIEF ANALYSIS OF WWER OPERATION

To-day 20 units of WWER-1000 and 27 units of WWER-440 are in operation at NPP in Russia and some other countries. The implemented complex of investigations aimed at improving and stabilising the quality of all fuel rod components as well as the optimised operating conditions of the reactors made it feasible to switch the WWER-1000 to a four-year fuel cycle. The mean fuel burn-up in FA discharged in 1994-1995 after operation for four cycles is some 44 MW·day/kg U. It is important to note that the number of leakers does not increase. The more so, as the experience is gained their numbers are decreasing and in 1994-1996 they were  $(1.2 \div 2.2) \times 10^{-5}$ . The fuel rod failures are for the most part of a "gas leakage" nature.

The results of the operation are adequately stable essentially for all 27 units of WWER-440. The average burn-up is 34.1 MW·day/kg U; while the maximum burn-up of 49.3 MW·day/kg U was reached by 4.4% enriched fuel of the four year cycle. The loss of tightness by fuels per cycle is on the average  $0.6 \times 10^{-5}$ ; during 1991-1995 no leakers were detected in many units.

Post-irradiation examinations of FA and fuel rods irradiated to various burn-up allow one to assess the reliability and capability of the fuel rods and FA and to reveal those parameters or factors that limit a further extension of the fuel burn-up.

First of all, the reliable operation of Zr-1%Nb alloy claddings is to be noted in the ammoniaboron-potassium water chemistry. The outer surface of the claddings is coated with a dark uniform oxide film 3-8  $\mu$ m thick; nodular corrosion is not available. At the inner cladding surface the oxide film thickness is variable and can reach 17  $\mu$ m. The claddings show a small amount of zirconium hydrides while the hydrogen content is within  $(3 \div 6) \times 10^{-3}$ % (mass). Thus, judging those factors the Zr-1%Nb alloy claddings have not yet exhausted their potential at the above burn-up.

The situation is somewhat different in relation to the mechanical properties of the claddings. These properties are little dependent on the fuel burn-up up to 43-45 MW·day/kg U. The claddings retain their strength and adequate margin of ductility. During operation the outer diameter of the cladding becomes smaller because of the difference between the pressures effected by the coolant and the gas inside a fuel rod. However, with the burn-up increased to more than 45-47 MW·day/kg U the cladding diameter is increased (fig.1).

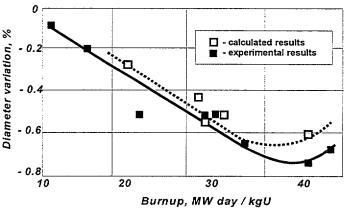


FIG.1. Fuel element diameter variation vs burn-up

This is a consequence of a gradually growing release of FGR with the fuel burn-up (fig.2) and a more intensive PCI.

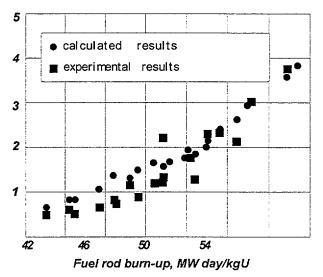


FIG.2. Fission gas release from WWER-440 fuel at externed burnup

The mean fuel rod elongation resulting from the irradiation induced growth at up to 50 MW·day/kg U burn-up obeys the relation  $\Delta L=0.01\times B$ , where  $\Delta L$  is a mean fuel rod elongation (%), B is burn-up (MW·day/kg U). The irradiation growth of fuel rods drastically increases with the burn-up (fig.3).

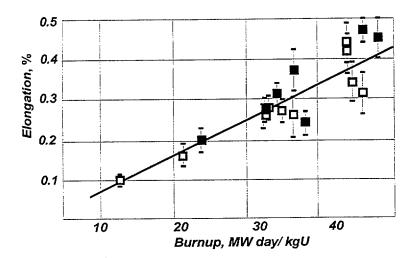


FIG.3. Fuel element dimensional element stability

The Zr-1%Nb alloy claddings have an adequately high crack resistance. Fig. 4 illustrates the crack velocity as a function of the stress intensity factor for claddings of recrystallized Zr-1%Nb alloy and annealed Zry-4, namely, for the conditions in which those alloys are used as fuel rod claddings in commercial reactors. The comparison between the crack resistances of those two alloys in an iodine medium shows that the threshold stress intensity factor of Zr-1%Nb alloy is noticeably higher than that of Zry-4.

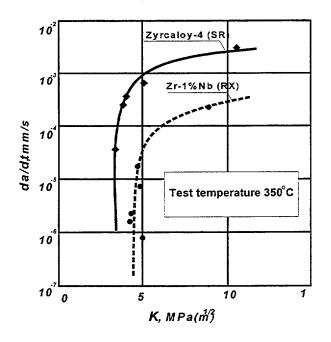


FIG.4.Crack propagation rate versus stress intensity factor for Zr-1%Nb cladding (●) and Zircaloy-4 (◆)

# 2. PERFECTION OF WWER FUEL CYCLE AND WAYS OF BURN-UP EXTENSION

Next to the nuclear power safety the second important criterion is its cost-efficiency. It depends upon many factors. The majority of them directly related to the reactor core can be combined under the general concept, namely, perfection of the reactor fuel cycle. It is clear that each reactor is characterized by the specific features of its fuel cycle. Currently, under improvements are fuel cycles of WWER-1000 and WWER-440.

The main efforts to optimize the WWER-1000 fuel cycle comprise the following.

- Integral fuel Gd<sub>2</sub>O<sub>3</sub> burnable absorber (IFBA) is in the process of introduction. For the purpose a special production was established where fuel assemblies were fabricated in adequately high quantities and loaded into reactors.
- Steel spacer grids and guide thimbles are being replaced by Zr ones. The commercial production of those components has been mastered and currently they are in-pile tested.
- Based on the above measures the reactors are being switched to the four-year cycle.
- The work is under way to increase the fuel content per a fuel rod via the pellet hole reduction and optimisation of the dimensions and tolerances for the diameters of cladding tubes and fuel pellets.
- The low leakage loading pattern with a longer time between reloads is being optimised.

The following is contemplated for the WWER-440 reactors:

- Conversion of all reactors to a four and then five-year cycle.
- Profiled enrichment of fuel across FA.

The reactors were converted to FAs having Zr spacer grids to be operated in the four-year fuel cycle.

In connection with a substantial reduction of steel in the reactor core the issue of a lower Hf content in Zr became urgent. Therefore, beginning from 1993 zirconium is commercially produced having <100 ppm Hf in place of the 500 ppm Hf content that was tolerable previously.

The implementation of the above measures will result in 12-19% reduction of the specific consumption of fuel per unit of electricity generated and a higher cost-efficiency of NPP. The activities to introduce those improvements will be basically completed by 2000. As it has already been mentioned some of the improvements have been implemented.

Besides, the WWER-440 reactors having Zr spacer grids have been switched to the four-year fuel cycle. Under consideration is the feasible conversion of those reactors to the five-year cycle.

A large scope of work is under way to optimize the WWER-1000 FA. To further improve the reactor cost-efficiency and safety an alternative FA design is under developments. This FA has no steel available within the disposition of uranium oxide. The fuel assembly has a support structure fabricated from Zr-alloys including the central channel of the higher strength multicomponent E-635 alloy. This will prevent the FA distortion in a more reliable way: the fuel pellet is subject to some changes, namely, the central hole is reduced from 2.4 mm to 1.4 mm.

The physical calculations indicate that 4.4% enriched fuel integrated to the Gd burnable absorber makes the four-fold reload realizable at the discharged fuel assembly averaged burn-up of 50 MW-day/kg U and the maximum burn-up of  $\sim$ 55 MW-day/kg U.

The E-635 alloy having higher corrosion and irradiation resistances is assumed to be used as fuel rod claddings and spacer grids in reactors of the 60 MW day/kg U burn-up and the 5 year cycle.

# 3. RBMK FUEL CYCLE OPTIMIZATION AND HIGHER FUEL BURN-UP

Every improvement of the RBMK fuel and the core as a whole is assessed in terms of increasing its reliability and safety. These parameters are substantially dependent on the steam void coefficient of reactivity. In the reactors in operation the steam void coefficient of reactivity was lowered via extra absorbers introduced into the core. However, this resulted in a substantial decrease of fuel burn-up and an increase of the FA channel power. However, the calculations demonstrate that this problem may by resolved in a more efficient way through integrating  $Er_2O_3$ , a burnable absorber, to fuel [2]. First, this will eliminate the extra absorber introduction. Second, absorbers available in fresh FAs reduce their power and significantly decrease a local power flash-up when FAs are loaded into a reactor. All this taken together simplifies the reload procedure and control of the power distribution within the core. A lower non-uniformity of the power density will allow a higher enrichment, thus, increasing the fuel burn-up. The production of the uranium-erbium fuel for RBMK-1000 and RBMK-1500 has been mastered. Presently have been fabricated and currently under test are 200 assemblies with this fuel at the Leningrad NPP and 150 assemblies at the Ignalina NPP. All RBMK reactors are assumed to be converted to the uranium-erbium fuel. The erbium content of the fuel is 0.4-0.5% mass. The fuel enrichment was increased from 2.4% to 2.6% for RBMK-1000 and from 2.0% to 2.4% for RBMK-1500. The introduction of erbium has no effect on FGR [3].

One of the distinctive features of the RBMK fuel performance is that at a substantially lower fuel burn-up compared to that of WWER, the cycle time is adequately long (1100-1200 eff. days). In this case the fuel rods operate under the conditions without control for radiolytic gases at the oxygen content of the water up to 20 µg/dm<sup>3</sup>. However, the limitation of the used Zr-1%Nb alloy is a drastic degradation of its corrosion resistance at the oxygen content of the water above 10 µg/dm<sup>3</sup>. This results in nodular corrosion and an aggravation of general corrosion on the waterside, particularly, under spacer grids. This in its turn leads to claddings being superheated, local hydrogen uptake etc. The section of fuel rod cladding under a spacer grid may be decreased by 40%. Those limitations may be obviated via the application of the well-studied Zr-1%Nb-1.3%Sn-0.35%Fe alloy as a cladding material. This alloy is much superior to the Zr-1%Nb one not only in irradiation but also in corrosion resistance and which is very important it is not prone to nodular corrosion. This is particularly attractive for the claddings of boiling reactor fuel rods, operating without control for radiolytic gases. Fuel rods clad in this allow were representatively tested at the Leningrad NPP under the standard operating conditions. The post-irradiation examinations have shown that the fuel rod claddings are coated with a lustrous uniform dark oxide film. No traces of nodular corrosion are available. The local corrosion of the fuel rod claddings under the spacer grids is some 3.5 times lower than that of Zr-1%Nb alloy.

Russia has adopted the concept of the closed nuclear fuel cycle. To implement this concept among other things a plant of the reprocessing capacity 400 t spent fuel/per year was set up [4].

This made it feasible to close the fuel cycle for U. Of the variety of the fuel rods used in the nuclear power of Russia only the reprocessing of RBMK spent fuel rods is not contemplated. They are shipped to be long-term stored. Their low residual content of U-235 and the low content of fissile species in the generated plutonium make their reprocessing cost inefficient. Meanwhile, the RBMK reactors are actively involved in the closed nuclear fuel cycle since their fuel is for the most part fabricated from recovered uranium after adjustments for the U-235 content are made. The use of the recovered uranium has not essentially affected the fuel rod production process or the reactor performance. The recovered uranium is also assumed to be used in the WWER reactors. This significantly reduces the demand for natural uranium.

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# SEU/RU/THORIA FUELS

(Session 3)

Chairperson

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### RECYCLED URANIUM — AN ADVANCED FUEL FOR CANDU REACTORS



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### Abstract

The use of recycled uranium (RU) fuel offers significant benefits to CANDU® reactor operators particularly if used in conjunction with advanced fuel bundle designs that have enhanced performance characteristics. Furthermore, these benefits can be realised using existing fuel production technologies and practices and with almost negligible change to fuel receipt and handling procedures at the reactor. The paper will demonstrate that the supply of RU as a ceramic-grade UO<sub>2</sub> powder will increasingly become available as a secure option to virgin natural uranium and slightly enriched uranium(SEU). In the context of RU use in Canadian CANDU reactors, existing national and international transport regulations and arrangements adequately allow all material movements between the reprocessor, RU powder supplier, Canadian CANDU fuel manufacturer and Canadian CANDU reactor operator. Studies have been undertaken of the impact on personnel dose during fuel manufacturing operations from the increased specific activity of the RU compared to natural uranium. These studies have shown that this impact can be readily minimised without significant cost penalty to the acceptable levels recognised in modern standards for fuel manufacturing operations. The successful and extensive use of RU, arising from spent Magnox fuel, in British Energy's Advanced Gas-Cooled reactors is cited as relevant practical commercial scale experience. The CANFLEX® fuel bundle design has been developed by AECL (Canada) and KAERI (Korea) to facilitate the achievement of higher burn-ups and greater fuel performance margins necessary if the full economic potential of advanced CANDU fuel cycles are to be achieved. The manufacture of a CANFLEX fuel bundle containing RU pellets derived from irradiated PWR fuel reprocessed in the THORP plant of BNFL is described. This provided a very practical verification of dose modelling calculations and also demonstrated that the increase of external activity is unlikely to require any change to current fuel manufacturing or fuel receipt and handling procedures at the reactor. A programme of work being carried out co-operatively by ZPI, BNFL and AECL is described which is aimed at the early introduction and commercialisation in Canada of CANDU fuel containing RU. This programme builds upon the successful technical collaboration undertaken by AECL and KAERI on CANFLEX fuel bundle development with the later support of BNFL for RU.

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### 1. INTRODUCTION

Previous IAEA fuel cycle documents and various conference papers [1,2] have described in detail the technical, economic and environmental benefits achievable from utilising RU as a fuel for CANDU reactors. In summary these are:

- 1. Significantly improved fuel cycle economics;
- 2. Better utilisation of natural resources, through large reduction in fuel requirements and recycling existing uranium stocks;
- 3. Maximising the energy derived from uranium by recycling the RU in CANDU, thus assisting national and global efforts to meet Kyoto targets for the reduction in greenhouse gases; 50% more energy can be derived by recycling the RU in CANDU rather than re-enriching it and recycling in a PWR.

AECL, KAERI and BNFL have a high level of confidence that the technical viability and licensing of CANFLEX/RU in the CANDU reactor system can be achieved.

This paper aims to demonstrate the steps now being taken collaboratively by ZPI, BNFL and AECL to ensure that these benefits are available as quickly as possible to Canadian CANDU reactor operators.

### 2. AVAILABILITY OF RU

The total amount of RU produced from reprocessing operations in Europe and Japan is expected to be around 25000 tes by the year 2000 with additional quantities also having arisen from reprocessing operation in the former Soviet Union. In the UK alone, the average annual output from BNFL's THORP plant over its first 10 years of operation is expected to be 700 tes U derived from reprocessing oxide fuel from Advanced Gas-Cooled Reactors (AGRs) and LWRs. Each 700 tes is sufficient to supply a typical CANDU reactor for 10 years operation. The product will be in the form of slightly enriched UO<sub>3</sub> powder with the majority of the material at an enrichment level up to  $1\%^{235}$ U. Reuse in AGRs or LWRs will require reenrichment. For RU use in a CANDU reactor, however, re-enrichment is not required. In this respect, advantage is being taken of the high neutron economy inherent in the CANDU reactor design which allows use of fuel with a relatively low fissile content and enables burnups to very low fissile content levels to be achieved.

Although much of the RU to arise from reprocessing will be owned by utilities that will recycle the material in their reactors, there is no doubt that sufficient RU will be available post 2000 to fuel any realistic Canadian CANDU programme.

### 3. GENERAL PROCESS DESCRIPTION

The overall activities that need to be considered for the supply and use of Recycled Uranium in CANDU reactors are shown in Figure 1.

### 4. OCCUPATIONAL HEALTH ASPECTS OF RU

The radiological inventory of RU is dependent upon the fuel history prior to reprocessing, the ageing stages and also the various decontamination factors (DF's) of the processes involved. The first step in evaluating the dose implications of all fuel manufacturing and handling operations involving RU is to determine reference radiological source terms that bound material that will eventually be processed. In considering the overall production of fuel via  $UF_6$  and the

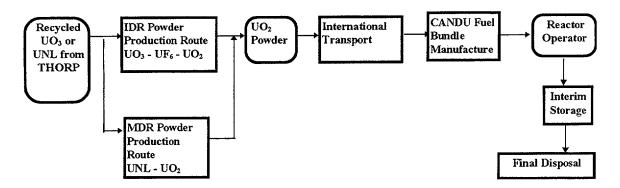


FIG 1. Process Schematic for the Use of RU in CANDU

IDR conversion route, radionuclide inventories for the RU at the various stages of  $UO_3$  conversion,  $UO_2$  powder production and fuel bundle manufacture have been compiled and compared for both natural uranium and for a reference RU material. This reference material has been used within BNFL and ZPI to perform preliminary health physics assessments of fuel manufacturing operations. Again, this reference RU represents a bounding, worst case for radiological assessment.

Using the above data and the International Commission on Radiological Protection (ICRP) material classification that applies to  $UO_2$ , a weighting can be applied to provide an Annual Limit on Intake (ALI) comparison of internal doses, arising from the intake of RU, with natural uranium that can be used to derive criteria to achieve acceptable internal operator dose.

Calculation of the external beta, gamma and neutron dose rates have also been made and compared with dose rates from natural uranium. Doses arising from fission products have been shown to be below the level of detection. The dominant beta source is  $^{234}$ Pa(m), which is present in all types of UO<sub>2</sub> since it is a product of the  $^{238}$ U decay chain and is therefore seen at a similar level. The gamma dose rates are driven by  $^{234}$ Pa(m) and also by  $^{208}$ Tl which is a product of the  $^{232}$ U decay chain and is only present in aged, recycled uranium. The neutron source strength has been shown to be low in all cases and below 1µSv/h contact.

For RU, the major contribution to external, whole body dose will be due to gamma emission and this will be slightly higher than the natural value for the first few tens of days of ageing but will continue to increase linearly for several years. An increase in gamma activity of a factor of around 2 over natural is expected at about 6 months ageing for the bounding, reference material, taking into account the additional contribution by bremsstrahlung radiation which is known to be present and generated by beta radiation absorption in the powder.

The manufacture of the RU CANFLEX bundle displayed at Toronto, referred to earlier, provided an opportunity to perform actual reading dose measurements at various stages throughout the manufacturing process. These show that although both the theoretical and actual reading values are higher than natural uranium, as expected, the increase was only modest. These results, albeit for a small trial quantity of a relatively low burn up material, and calculated data for higher burnup PWR and AGR derived RU compared with natural uranium as shown in Figure 2. Based on these data, preliminary assessments of ZPI's Port Hope facility indicate that satisfactory control of gamma dose can be achieved with the introduction of RU to a fuel manufacturing plant already using natural uranium. Careful planning of fuel manufacture and refuelling operations should ensure that appropriate health physics standards are also achieved at the station.

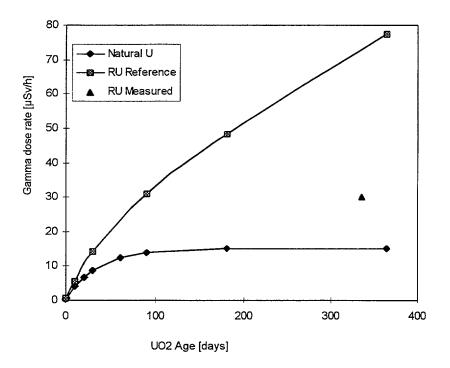


FIG 2. Drum Surface Gamma Dose Rates for RU Powder

BNFL already has extensive commercial scale experience in manufacturing fuel from recycled spent fuel. Some 15000 tes of spent Magnox fuel has been converted to  $UF_{6}$ , re-enriched and used in the manufacture of AGR fuel in the UK. The burn-up levels and consequently <sup>232</sup>U content of the RU were lower than those now arising from LWRs or AGRs but the experience is significant in that no special modification to existing equipment was required, manufacturing procedure changes were minimal and the fuel presented no operational problems to the reactor operators.

It should also be recognised that because the CANDU route does not require re-enrichment of the  $^{235}$ U there is no consequential re-enrichment of  $^{236}$ U and  $^{232}$ U. Thus the dose level associated with the handling of RU for CANDU is greatly reduced compared with the routes that require re-enrichment.

### 5. PROCESSING OF RU TO UO<sub>2</sub> POWDER

For BNFL to supply RU into the Canadian CANDU market, it will be necessary to supply  $UO_2$  powder of a specific enrichment value to a CANDU fuel manufacturer and thus conversion to a ceramic-grade  $UO_2$  powder is required. Furthermore, the enrichment value chosen must take account of the neutron absorbing property of the <sup>236</sup>U content and can be considered an equivalent <sup>235</sup>U enrichment [3].

There are a number of uranium processing options available that would be capable of producing a quality powder product. Following a review of the possible options, BNFL has concluded that the short term business requirements can most cost effectively be supplied from facilities provided to support recycle of RU in LWRs. UO<sub>3</sub> from THORP will be transported to BNFL's Springfields site, converted to UF<sub>6</sub> in the new Line 3 Hex facility and then converted to UO<sub>2</sub> powder using the Integrated Dry Route (IDR) process in the Oxide Fuel Complex.

This route offers the additional advantage in that there is effective removal of  $^{232}$ U decay daughter products which has the potential of reducing operator dose during downstream operations including fuel manufacturing and reactor fuelling operations. This is discussed in more detail in subsequent sections of this paper when the occupational health physics aspects of the use of RU in a CANDU fuel manufacturing plant are examined.

The IDR product is a long-established product and has successfully been used for fuel pellet manufacture for many different types of reactors [4]. In the CANDU context, pellets made from IDR  $UO_2$  will be capable of meeting all quality requirements and will accommodate any future CANDU fuel specifications that may be introduced as fuel burnup increases are sought.

An alternative process, the Modified Direct Route (MDR), has been proven by BNFL at pilot plant scale. This process is based upon kiln-denitration of a uranyl nitrate-ammonium nitrate double salt, uses uranyl nitrate as a feed material, not UO<sub>3</sub>, and does not involve any decay daughter removal steps. In this respect it will be important to more closely couple the THORP purification process to the conversion and ultimately the fuel manufacturing operations to minimise operator dose. In the longer term this process may provide additional economic benefits for CANDU over the existing IDR route. This MDR process has been successfully used to produce THORP-derived RU pellets for a CANFLEX design fuel bundle that was recently displayed at the AECL Sheridan Park Engineering Laboratories during the 5th International CANDU Fuel Conference held in Toronto during September 1997.

### 6. TRANSPORT OF RU

The issues and proposed arrangements for the transport of all necessary material movements involving the reprocessor (BNFL), RU powder supplier (BNFL), Canadian CANDU fuel manufacturer (ZPI) and Canadian reactor operators have been examined jointly by BNFL and ZPI in relation to UK and Canadian national regulations and international regulations as set out in IAEA Safety Standard Document - Regulations for the Safe Transport of Radioactive Material, 1996 Edition No ST-1.

It has been demonstrated that there are no difficulties involved in the transport of RU as either non-fissile material (<1.0%<sup>235</sup>U) or fissile material ( $\ge1.0\%$ <sup>235</sup>U) as defined in the above document.

In the former case, analysis of typical THORP product and processing operations has demonstrated a 205 litre IP1- class drum could be used as the primary transport container within an outer IP2 ISO type container.

For fissile material, BNFL would use its type 3516 (B) container which is currently undergoing testing and is expected to be licensed in 1999.

### 7. FUEL MANUFACTURE

Nuclear fuel has been manufactured in Canada in significant quantities since the late 60's using natural enrichment; enriched fuel has also been manufactured for research reactors in various quantities during this same period. More recently, three styles of CANDU fuel have been routinely produced in high volumes, viz: 28 and two 37 element variants. These fuels are produced in a flow-through continuous mode. This type of production is carried on using a number of identical parallel production lines, with the capability for quick change-over to a different style. Until now, each fuel style has had a single common sized fuel element, albeit with a number of exterior appendage configurations.

### 7.1 CANFLEX MANUFACTURING ASPECTS

CANFLEX fuel uses two different sizes. These new sizes have been produced in small production sized quantities without difficulty at ZPI's Metal Products tubing facilities and are being used for the 24 bundle in-reactor qualification beginning later this year. These bundles have been manufactured using development lab equipment to gain a better understanding for eventual production quantities.

Production quantities of CANFLEX fuels will present the manufacturer with a number of challenges. The single major change will be to batch process a fuel with 2 different sized fuel elements using a separated production line, while simultaneously producing the traditional 28 and 37 element fuel designs.

Segregation of the pellets will be necessary to avoid a plausible misloading into a 28/37 fuel bundle during parallel production. For this reason a separate production line may be required during various phases of production of CANFLEX fuel. Instead of traditional continuous production, one size of fuel pellet will be produced and stored until the second size pellet has been produced; the ratio of production will be in the order of 35:8. Once the pellets are loaded into fuel sub-assemblies, the separation concerns are reduced. Subsequent assembly operations pose a few additional challenges but are manageable. In terms of throughput, a single CANDU 6 fuelling charge will require only limited utilisation of resources even though the total number of fuel bundles will be increased by the ratio 19.1/18.6 or approximately 3% ( due to the reduction in U weight with the CANFLEX design).

From a commercial point of view, the addition of this fuel design will require investment in plant expansion and new processing equipment. When viewed over a reasonable time period, the incremental manufacturing costs to the utility are not expected to be overly significant.

### 7.2 ENRICHED U MANUFACTURING ASPECTS

The change from CANFLEX-NU to SEU, of enrichments of less than 1 % will not present any special problems other than the obvious health physics issues; as mentioned above, these are not expected to present major changes. From a production stand-point, an enriched fuel bundle will require significantly lower production rates, leaving an even larger under-utilisation of equipment resources.

### 7.3 SPECIFIC RU MANUFACTURING CONSIDERATIONS

Several additional considerations need to be addressed regarding the production of RU fuel.

Although physical transportation is not seen as an impediment, nuclear insurance costs for Canadian territorial waters and inland transportation are being assessed; these are not expected to be significant.

The growth of radiological daughter products is a health physics aspect that must be monitored by the manufacturer and utility. For this reason, both the manufacturer and utility will have to increase the existing controls for RU inventory over those presently in use. More significantly, the usual small quantities of contaminated waste generated by normal equipment use/maintenance must also be addressed. Like many countries, Canada's existing waste disposal sites are at a premium and therefore another location or means of disposal or recovery maybe needed. Segregation of material will be even more important for RU, to avoid any cross contamination. This may entail further physical barriers than those being considered for CANFLEX-NU manufacture. In addition to physical plant, additional equipment will also be required.

As in CANFLEX-NU and SEU fuels, the incremental manufacturing costs of RU are not expected to be great. These costs are presently being derived.

### 8. REACTOR OPERATION AND FUEL PERFORMANCE

CANFLEX is the latest fuel carrier in the evolution of CANDU fuel [5, 6]. Its design has been driven to facilitate higher burnups required for advanced fuels such as recycled uranium, to provide higher dryout powers and lower peak element ratings, while being fully compatible with existing CANDU stations [7]. Because CANFLEX is fully compatible with existing plants, CANDU6 stations can simply substitute CANFLEX-NU for 37-element fuel and achieve improved reactor operating and safety margins, and higher critical channel powers.

The enrichment in RU can be used in several ways: to increase the power from a given reactor core size through flattening the radial channel power profile; to increase the fuel burnup and reduce the quantity of spent fuel; to improve fuel cycle economics, both front- and back-end; and, in general, to provide greater flexibility in reactor design.

A 500 full-power day core-follow simulation using both 2 and 4-bundle refuelling schemes with CANFLEX RU resulted in bundle power and bundle power boost envelops that meet current CANDU fuel performance criteria [ 8 ]. Individual maximum bundle and channel power targets are met. There is significant margin in the fuel element power envelops. Practical refuelling schemes are demonstrated to achieve acceptable fuel performance within currently proven CANDU technology.

### 9. SPENT FUEL STORAGE AND DISPOSAL

Spent RU CANDU fuel will probably require some extended pool storage to allow the decay heat to reach the level of spent natural uranium fuel before being transferred to interim dry storage in existing storage containers. In terms of spent fuel disposal, the OECD/NEA study [9] on the disposal costs for high level waste indicated that despite the larger volumes of spent fuel, disposal costs for CANDU natural uranium fuel are roughly equivalent to the disposal costs of spent PWR fuel. The impact on the repository design of the higher quantity spent natural uranium CANDU fuel would be offset by its lower decay heat load, resulting from the lower burnup. Notwithstanding this conclusion, AECL has recently conducted preliminary cost assessments of the impact of SEU on spent CANDU fuel disposal costs of about 20%, compared to natural uranium fuel. The cost savings depend on the size of the repository, cooling times, and disposal method (in-room emplacement vs. boreholes). Hence, a fairly significant reduction in spent fuel disposal costs are possible with RU in CANDU.

### 10. CONCLUSIONS

ZPI, AECL and BNFL have identified and have been addressing the significant technical, health physics and economic assessments relating to their roles in the introduction of RU in Canadian CANDU reactors.

Overall, the recycle of RU into CANDU has beneficial environmental impacts on both the front-end (the mining of fresh uranium is displaced by uranium recycle), and back-end (significant societal gain is achieved by electricity production through a material that might

otherwise be considered a waste). This is an excellent example of the environmental 3R's (reduce, reuse, recycle) as applied to global nuclear energy use.

A demonstration irradiation of 24 CANFLEX bundles containing natural uranium is planned to start in mid 1998 in a Canadian commercial reactor. The next step is seen as the introduction of an RU trial bundle into a commercial reactor with the aim of ensuring that the Canadian nuclear industry starts to obtain the benefits within the next few years from the use of RU fuel as outlined in the introduction.

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# FUEL MANAGEMENT SIMULATIONS FOR 0.9% SEU IN CANDU 6 REACTORS

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### Abstract

Slightly Enriched Uranium (SEU) of 0.9 weight % <sup>235</sup>U enrichment is a promising fuel cycle option for CANDU<sup>®</sup> reactors. An important component of the investigation of this option is the demonstration of the feasibility of on-line refuelling with this fuel type in reactor physics fuelmanagement simulations. Two fuel-management schemes have been investigated in detail during 500-day core-follow simulations; these were a 2-bundle-shift and a 4-bundle-shift axial refuelling scheme. The 43-element CANFLEX<sup>®</sup> fuel design has been used in these studies because of its improved fuel performance characteristics in this application. The results of the studies are discussed in detail in this paper. The most significant conclusion of this study was that both 2- and 4-bundle-shift refuelling schemes with CANFLEX fuel result in bundle power and bundle power boost envelopes that meet current fuel-performance requirements.

### 1. Introduction

The use of Slightly Enriched Uranium (SEU) with 0.9% <sup>235</sup>U by weight is being studied as an attractive fuelling option for CANDU pressurized heavy-water reactors (PHWR). With 0.9% SEU, average discharge burnups of about 13,800 MW·d/t U can be achieved, improving naturaluranium resource utilization by about 34% relative to that achievable in a natural-uranium fuelled CANDU reactor. As well, utilization of SEU reduces spent-fuel volumes and fuelling costs, or it can be used to increase the power available from a given reactor by flattening the radial power profile, or to achieve a combination of these objectives.

To evaluate the effect of introducing 0.9% SEU into a PHWR of the current CANDU 6 design, reactor simulations are required to determine the operating characteristics and fuel operating envelope. Simulations were performed of 500-full-power-days (FPD) of operation of a CANDU 6 reactor refuelled with SEU CANFLEX fuel, with 2- and 4-bundle-shift bi-directional refuelling schemes.

The 0.9% SEU can come either from a supplier of enriched uranium fuel or from a utility owner of recovered uranium (RU) [1] from the conventional reprocessing of spent LWR fuel. RU has the advantage of potentially lower fuelling costs than does SEU.

Previous analyses [2] indicated that, although either a 37-element or CANFLEX fuel design could be used in this application, CANFLEX fuel provides a greater fuel performance margin because its design results in peak fuel linear-element ratings that are about 20% lower. On that basis, the CANFLEX fuel design was selected as the configuration for these detailed fuelmanagement investigations.

### 2. Method of Analysis

All reactor core calculations were performed using the standard code applied for CANDU reactor core design and analyses, RFSP [3]. All the lattice-cell calculations were performed using the

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WIMS-AECL code [4, 5], with an ENDF/B-V-based nuclear data library. The main calculational sequence applied in the analyses reported in this paper were

- 1. Lattice-cell properties for the fuel type being studied were calculated as a function of irradiation using WIMS-AECL;
- 2. The results of the WIMS-AECL calculations were used to prepare "fuel tables" for the representation of specific fuel types in RFSP calculations;
- 3. Time-average fuel-management calculations were performed with RFSP for a CANDU 6 reactor, for the fuel types and fuel-management options under evaluation;
- 4. Quasi-equilibrium patterned-random fuel-age distributions calculated from the time-average irradiation distributions were used to approximate initial instantaneous core states; and
- 5. Explicit time-dependent refuelling simulations were conducted at 2 to 3 FPD intervals to investigate the operating envelope in 500 FPD of reactor operation.

Two fuel types and refuelling schemes were used in these analyses:

- 1. a 2-bundle-shift CANFLEX RU with an enrichment of 0.96%, and
- 2. a 4-bundle-shift CANFLEX 0.9% SEU

The 2-bundle-shift refuelling study was previously documented in Reference 6. The 0.96% enriched fuel was of a typical RU composition, and the 0.9% fuel is SEU; the compositions of both are presented in Table 1. These two enrichments are both within the range of near-term interest for either RU or SEU, and there is no significance to the choice of different enrichments for the two simulations. Adjuster rods and zone-control devices in the reactor core were represented using properties originally derived for a natural-uranium-fuelled configuration; subsequent analysis indicated that these properties were equivalent to those derived for an SEU-fuelled configuration. The reactivity worth of the adjuster rods used in the reported analyses exceeds the current requirements for xenon override following reactor shutdown.

Refuelling decisions during the 500-FPD core-follow simulations were made using a semiautomated procedure:

- 1. All the 380 fuel channels in the core are ranked for suitability for refuelling, according to the power and burnup information for each channel and its neighbours. Higher power and lower burnups result in lower rankings.
- 2. A number of channels are selected as candidates for refuelling according to their rankings. The number of channels selected is that estimated to be required to maintain core criticality over the next burnup step: about 4 per day in the 2-bundle-shift scheme and 2 per day in the 4-bundle-shift scheme.
- 3. A burnup step is simulated, and the power distribution at the end of the time step is checked against several acceptance criteria:
  - (a) The maximum channel power must be less than a target value. This target value was 7100 kW in the 2-bundle-shift study; this value later revised downward to 6950 kW in the 4-bundle-shift study to reflect current CANDU 6 operating practice [7]. The total core power was maintained at 2061.4 MW (thermal) throughout the simulations.
  - (b) The maximum bundle power must be less than a target value. This target value was 882 kW [7] in both studies.

TABLE 1:	Compositions of	Uranium in	RU	and SEU	Refuelling	Simulations
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Fuel	Nuclide	Weight %
0.96% RU	$^{234}U$	0.016
	$^{235}\mathrm{U}$	0.956
	$^{236}U$	0.275
	$^{238}U$	98.750
0.90% SEU	<sup>235</sup> U	0.90
	$^{238}{ m U}$	99.10

- (c) The average zone controller fill must be in the range of 0.3 to 0.7.
- (d) The individual zone controller fills must be in the range of 0.05 to 0.9.
- (e) Zone powers and tilts should be near their normal operating values.

If these acceptance criteria are met, the simulation proceeds to the next time step in step 1. If the acceptance criteria are not met, one or more channels will be excluded from the candidates, and the process is repeated from step 1 for the current step. The channels to be excluded are determined according to the following criteria:

- (a) If the peak channel power occurred at a channel that had just been refuelled, it is excluded from the list, or
- (b) If the peak channel power occurred at a channel that was not just refuelled, that channel and its eight nearest neighbours are excluded from the list.

In practice, the automated refuelling scheme worked quite well, although some manual intervention was occasionally required.

The reference power distribution in the 4-bundle-shift simulation was radially flattened relative to the 2-bundle-shift configuration (resulting in lower time-average maximum channel powers), in order to accommodate the high-power ripple caused by that refuelling scheme.

	2-Bundle Shift		4-Bundle Shift		
	Time-Average	Core Follow	Time-Average	Core Follow	
Average Discharge	14130	13960	12460	12300	
Burnup (MW·d/t U)					
Maximum Channel	6820	7021*	6249	6825*	
Power (kW)					
Maximum Bundle	771	857	730	840*	
Power					
<b>Refuelling Rate</b>	4.1	3.9	2.3	2.3	
(channels/day)					
<b>Refuelling Rate</b>	8.2	7.8	9.3	9.4	
(bundles/day)					

TABLE 2: Primary Characteristics of 0.9% SEU-Fuelled CANDU Reactors

\*Average of the maximum values

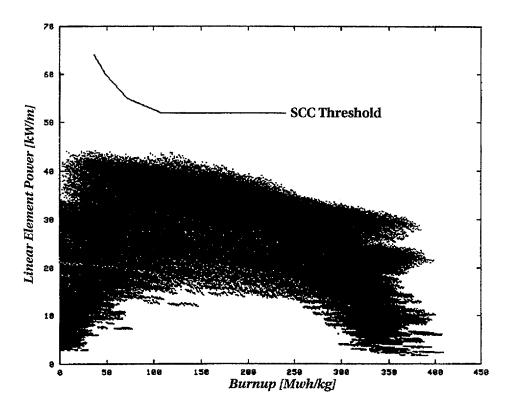


FIG. 1: Fuel-Element Operating Power for 2-Bundle Shift

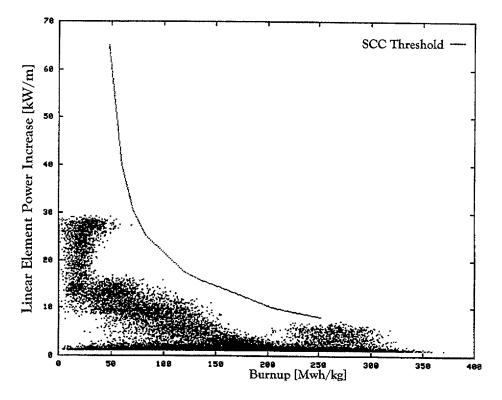


FIG. 2: Fuel-Element Power Boost for 2-Bundle Shift

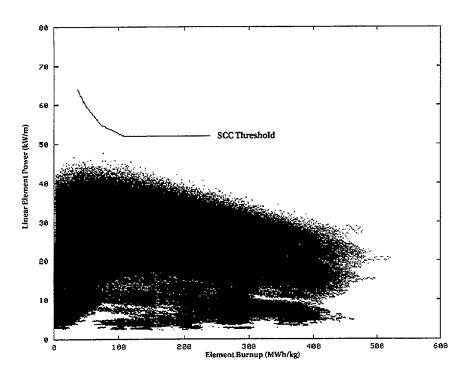


FIG. 3: Fuel-Element Operating Power for 4-Bundle Shift

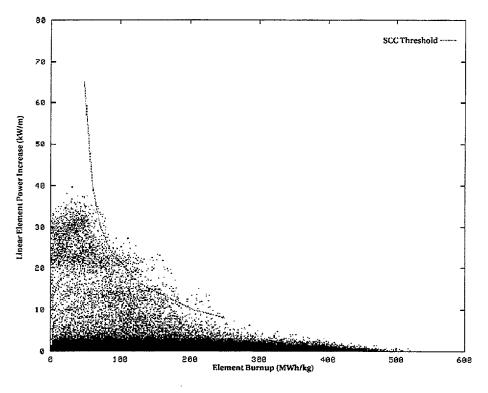


FIG. 4: Fuel-Element Power Boost for 4-Bundle Shift

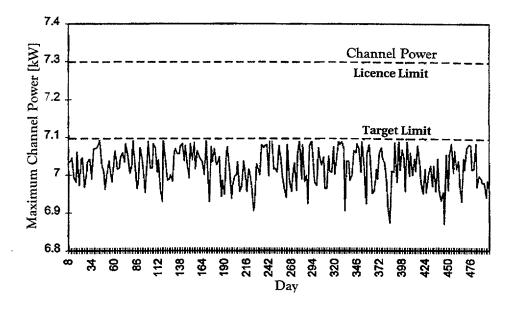


FIG. 5: Maximum Channel Power for 2-Bundle Shift

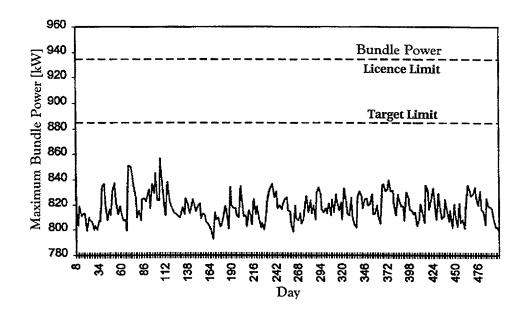


FIG. 6: Maximum Bundle Power for 2-Bundle Shift

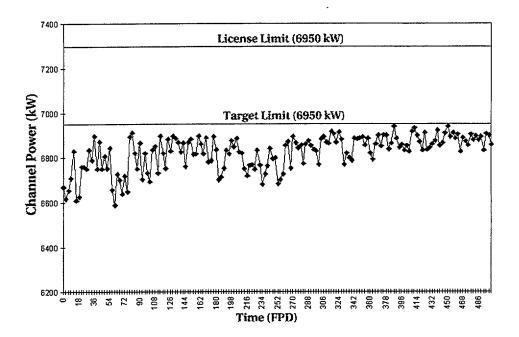


FIG. 7: Maximum Channel Power for 4-Bundle Shift

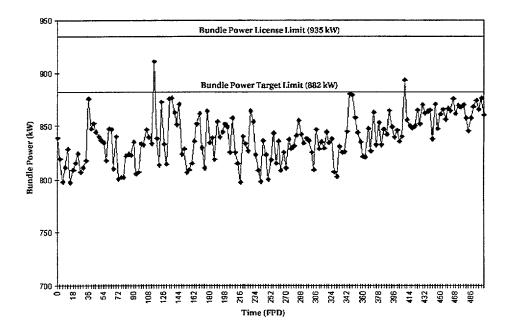


FIG. 8: Maximum Bundle Power for 4-Bundle Shift

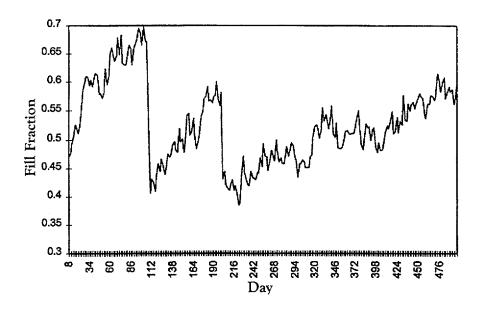


FIG. 9: Average Zone Fills for 2-Bundle Shift

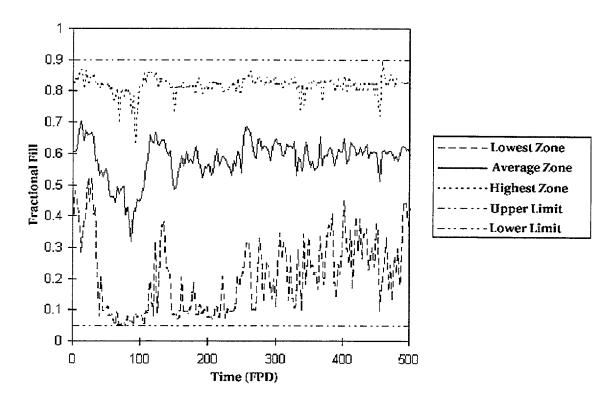


FIG. 10: Zone Fills for 4-Bundle Shift

# 3. Results of Simulations

The primary time-average characteristics of the 0.9% SEU-fuelled CANDU reactor for the two fuelling schemes studied are presented in Table 2. The significant features presented in this table are

- 1. The individual maximum bundle- and channel-power targets were met in both cases.
- 2. The reduction in <sup>235</sup>U enrichment of from 0.96% to 0.90% between the 2- and 4-bundle-shift simulations results in a reduction in average burnup of about 12%.
- 3. As expected, the 4-bundle-shift scheme results in a fuel channel visit frequency half that of the 2-bundle-shift scheme.

Figures 1 and 2 present the fuel-element linear power versus burnup and fuel-element power increase versus burnup in the 2-bundle-shift refuelling scheme, respectively, as well as the curves indicating the limits of acceptable fuel operation. Corresponding information is presented in Figures 3 and 4 for the 4-bundle-shift refuelling scheme. Exceeding both limiting curves at the same time corresponds to a 1% fuel failure threshold, and is generally considered unacceptable when evaluating new CANDU fuelling concepts. The results indicate that there is a significant margin in the fuel element operating power envelope in all cases and thus no fuel failures are anticipated, although the power-boost limit curves are exceeded over some burnup ranges in the 4-bundle-shift case and approach the high-burnup end of the boost threshold curve in the 2-bundle-shift case.

Figures 5 through 8 present the maximum channel and bundle powers in the 2- and 4-bundleshift cases during the 500-FPD simulations. The target analysis limits on maximum channel power were met throughout the simulations in both cases. The channel-power target for the 2bundle-shift simulation had been 7100 kW, but it is expected that the limit of 6950 kW would have been achievable. In the 2-bundle-shift case, the bundle power limit of 882 kW was comfortably met at all times. The 882 kW bundle power limit was exceeded at only 2 points in the 4-bundleshift simulation; these 2 exceptions are not considered significant and would have been easily accommodated with different refuelling choices if the refuelling logic had considered them.

Figures 9 and 10 present the zone controller fills during the 500-FPD simulations. The applicable zone-fill limits were met in all cases.

# 4. Conclusions

Fuel-management calculations conducted for 0.9% SEU-fuel CANDU 6 reactors indicate that either a 2- or 4-bundle-shift refuelling scheme with CANFLEX fuel bundles would meet current CANDU fuel-performance criteria. The use of 0.9% SEU would result in a 90% increase in bundle discharge burnups and an improvement of 34% in natural-uranium resource utilization, relative to current natural-uranium fuel. The results of the analyses reported here indicate that practical refuelling schemes may be used to achieve acceptable fuel performance, within currently proven CANDU technology.

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### NEUTRONICS AND THERMALHYDRAULICS CHARACTERISTICS OF THE CANDU CORE FUELED WITH SLIGHTLY ENRICHED URANIUM 0.9% U<sub>235</sub>

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#### Abstract

The interest concerning the slightly enriched uranium (SEU) fuel cycle is due to the possibility to adapt (to convert) the current reactor design using natural uranium fuel to this cycle. Preliminary evaluations based on discharged fuel burnup estimates versus enrichment and on Canadian experience in fuel irradiation suggest that for a 0.93% U-235 enrichment no design modifications are required, not even for the fuel bundle. The purpose of this paper is to resume the results of the studies carried on in order to clarify this problem. The calculation methodology used in reactor physics and thermal-hydraulics analyses that were performed adapted and developed the AECL suggested methodology. In order to prove the possibility to use the SEU 0.93% without any design modification, all the main elements from the CANDU Reactor Physics Design Manual were studied. Also, some thermal-hydraulics analyses were performed to ensure that the operating and safety parameters were respected. The estimations sustain the assumption that the current reactor and fuel bundle design is compatible to the using of the SEU 0.93% fuel.

#### 1. Calculation methodology

The analysis methodology used in the studies regarding the neutron characterization of the SEU 0.9 % fueled core represents an adaptation and development of similar methods used by AECL for this kind of analyses. It can be considered that the principal aspects characterizing this type of reactor which influence the choice of calculation methods are:

- utilization of heavy water as moderator;
- separation of moderator from coolant;
- horizontal fuel channels and perpendicular reactivity devices;
- continuous refueling.

By the reduced moderator absorption, the PHW lattices are well moderated. This makes possible that the neutron balance be well approximated in the four factors formula, and the neutron flux be well approximated in the Westcott formalism.

Subsequently, the neutron parameters are built most often in the age-diffusion approximation. For natural Uranium, one uses the POWDERPUFS (PPV) computer code (ref. /1/). For detailed and more rigorous information, one resorts to the LATREP code (ref. /2/), which deals with the multigroup flux distribution by collision

probabilities. Though these two codes are the most used programs for the CANDU reactor design analysis, one uses the WIMS code too for the multigroup transport calculation in some situations (ref. /3/).

For higher values of  $U_{235}$  content in the fuel some questions rise for the PPV results just because these results are based on the experimental data obtained for natural Uranium. That is why one uses especially the LATREP and WIMS computer codes for determining the cell parameters in the case of SEU fuel cycle. The flux distribution determination is done by two group, 3D diffusion calculations. The tridimensional model is required by the geometrical configuration with horizontal channels and perpendicular reactivity devices. An important feature is the existence of a neutron properties distribution induced by the presence of a different burnup fuel distribution as a result of a continuous bidirectional refueling. These aspects are taken into account by the way of building the macroscopic constants used in the diffusion calculations, i.e.:

- 'homogeneous calculations', which treat both global and local neutron balance, considering a uniform distributed fuel with averaged properties, up to the discharge burnup;

- 'time average calculations', which corespond to a similar method distinguished by the fact that one considers average compositions that differ along the channel, the time average being done up to irradiations depending on channel power and residence time, in compliance with the refueling scheme;

- 'instantaneous calculations', which treat the problem by appointing each bundle properties calculated at the burnup that results from the history of irradiation.

The 'instantaneous' constants approximation is used especially for the refueling simulation or for studies regarding the refueling strategies.

For advanced fuel cycles, the SERA program was developed, a program similar to FMDP, ref. /4/. The SERA program, ref. /5/, has a modular structure in which the macroscopic constants can be automatically generated with the LATREP code or can be taken from a WIMS built file. It can be used in all the three aproximations mentioned above. The code provides the possibility of building the constants in function of the real irradiation history for each bundle. In the multigroup diffusion approximation the reactivity devices are represented by specific homogenized cross sections, based on proper local calculations.

The PIJXYZ code was used. This integral transport program solves the multigroup equations in the whole supercell volume (ref. /6/). The needed material properties, of the fuel, pressure tube, reactivity device are determined by the WIMS program.

# 2. Cell parameters and reactivity effects

The neutron balance assessment at the cell level enables us to evaluate the reactivity effects produced by the variations of different parameters. These reactivity effects underlie the requirements for the different reactivity devices and, by

comparison with similar natural Uranium effects, one can know if it is necessary to change them.

The reactivity effects for SEU 0.9 % were analyzed by E. Nichita, for thermalhydraulics parameters variations in ref. /7/ and by P. Laslau for the Xenon concentration variation in ref. /8/.

The multiplication factor evolution with burnup for SEU 0.9 % is shown in figure no.1. The comparison with natural Uranium shows that, obviously, it starts from a higher initial k value and the Plutonium peak does not appear. This can be explained by the higher fissile concentration that makes the variation in the multiplication factor due to  $U_{235}$  exhaustion exceed the one due to the Pu build-up.

In figure no. 2 the integrated multiplication factor evolution is shown, a parameter that can be linked with the global balance in the hypothesis that the bundles in the core are characterized by a uniform irradiation distribution between zero and the discharge irradiation. This interpretation permits the discharge burnup assessment, considering a reactivity need for the neutron leak and reactivity devices and incidental absorption compensation. For example, in figure no. 2, one can estimate a discharge burnup of 14983 Mwh/t, resulting from the intersection between the multiplication factor evolution curve and the 47 mk line that corresponds to the assessed reactivity need.

In figure no. 3 the radial power distribution evolution for average bundle is shown; this evolution is the result of superimposing the effects due to changes in fissile isotopes concentrations and in flux microdistribution.

The moderator temperature reactivity effect is shown in figure no. 4, in which the multiplication coefficient variation with the moderator temperature for SEU 0.9 % is presented.

The fuel temperature reactivity effect is shown in figure no. 5, in which the multiplication coefficient variation with the fuel temperature for SEU 0.9 % is presented.

The void reactivity effect is shown in figure no. 6, in which the multiplication coefficient variation with the coolant void for SEU 0.9 % is presented.

The SEU 0.9 % and natural Uranium reactivity effects comparison shows the multiplication factor variations have the same behavior regarding each perturbation parameter and the same evolution trend with irradiation even if, for fuel at equilibrium, there are some small differences.

The reactivity effects due to Xenon have a special importance both in the steady state, by its large reactivity, and while operating, by the important neutron balance variations, that are induced at passing from one state to another and by the associated transients.

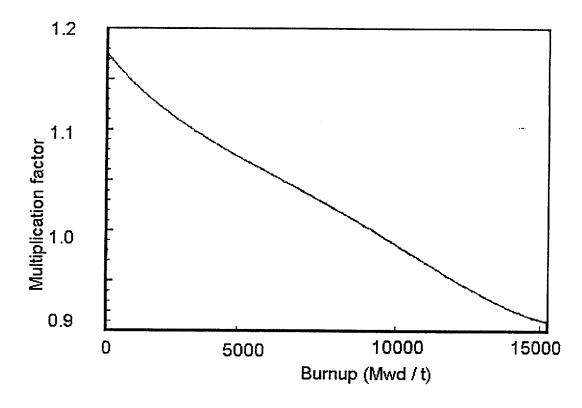


Figure 1. Multiplication factor versus burnup

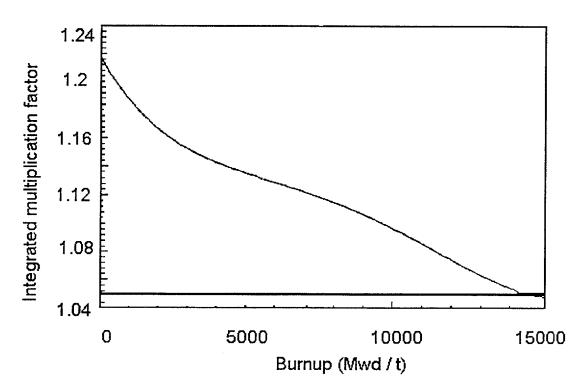


Figure 2. Integrated multiplication factor versus burnup

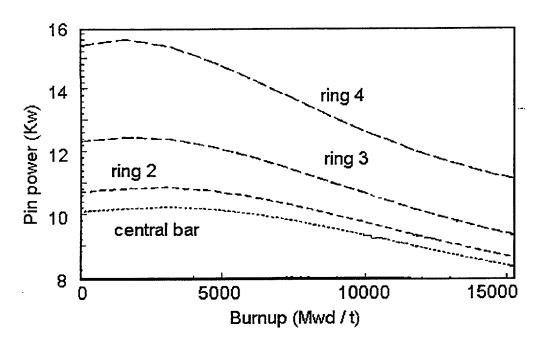


Figure 3. Pin power versus burnup

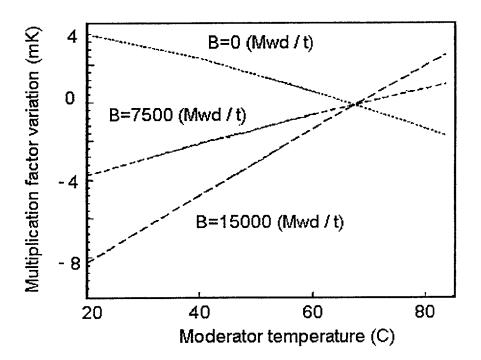


Figure 4. Multiplication factor variation versus moderator temperature

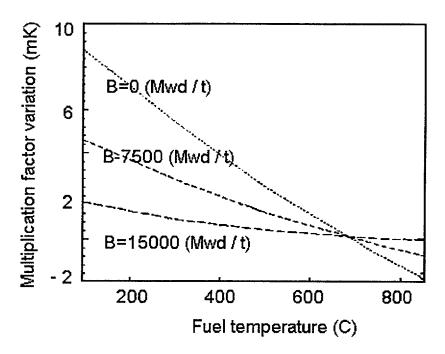


Figure 5. Multiplication factor variation versus fuel temperature

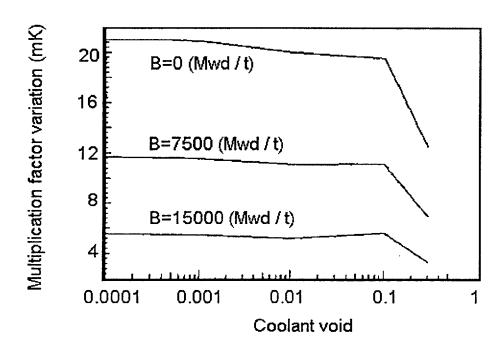


Figure 6. Loss of coolant reactivity effect

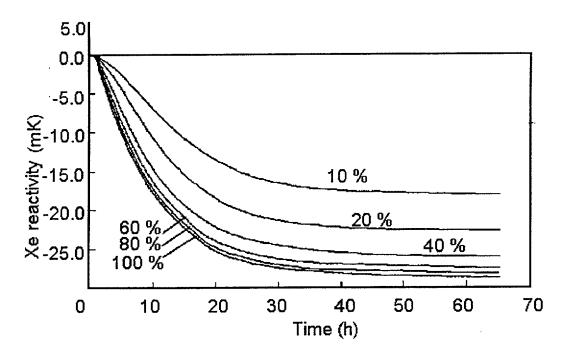


Figure 7. Xe reactivity transients after startup

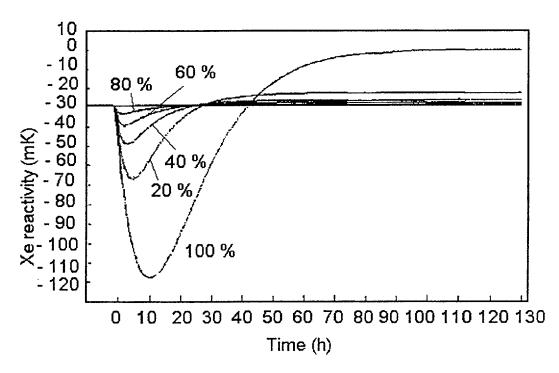


Figure 8. Xe reactivity transients after reduction of power from 100 %

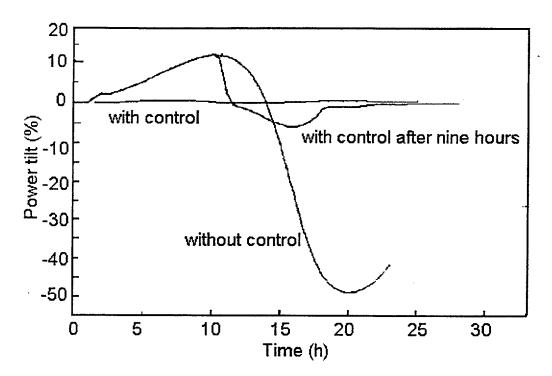


Figure 9. First azimuthal mode evolution

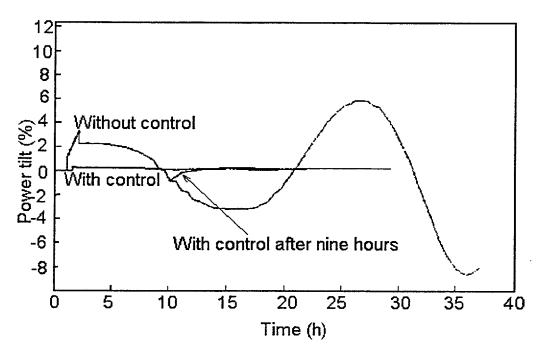


Figure 10. First axial mode evolution

The cumulated yield is over 7 %, for SEU fuel, from which over 10 % is prompt component. By production and loss of Xenon, by absorption and desintegration, a equilibrium state is reached, that, for SEU fuel, coresponds to a reactivity of 28-29 mk at nominal power. This can be identified in figure no. 7, where the Xe transients at startup for different power levels are shown.

In figure no. 8, the Xe reactivity transients resulted by the reduction of power up to different levels are shown, starting from nominal value. These transients are generated by the mismatch between reduction and production, whose decrease is delayed by Iodine desintegration. For that reason, in the beginning, a Xe concentration increase is taking place, and consequently, its negative reactivity increases, followed by a decrease to the equilibrium value given by the power value on each level.

Another important aspect that needs attention is the stability of the core with respect to the local Xe transients. In the PHW reactors, usually very large, with a strong flux flattening and relatively high power densities, Xe density spatial distribution oscillations can be observed. For setting the conditions in which these oscillations could appear and if they would naturally diminish or increase, special analyses are required to link the different Xe distribution spatial forms and the transient mode corresponding to them.

In this case, one was able to avoid the mentioned analyses, having a set of conclusions of such analyses for the natural Uranium core. In fact, one analyzed the transients of the first two modes, in the instability order, identified for natural Uranium. This procedure is justified by the fact that the Xe effects look alike for the two fuel types.

The results obtained for SEU fuel are gathered in figures 9 and 10, where the relative deformations of the power distribution characterizing these two modes, respectively left-right and front-back, are shown. As a general observation, one can state that the SEU fuel core is more unstable at the Xe oscilations, especially for the axial modes due to the additional axial flattening.

# 3. Equilibrium power distribution and refueling strategies

For SEU fueled core, due to the sharper channel neutron properties variations, the time average approximation calculations become the base of simulation and not only an improvement of the results obtained by homogeneous calculations. Because the average time calculations imply handling scheme elements, the equilibrium power distribution and refueling strategy are settled at the same time. The SEU utilization implies new refueling schemes, especially due to the sharper decrease with burnup and to larger power deformations after refueling.

There are some options which were identified and presented in scientific literature, for example ref. /9/ and /10/.

For choosing a convenient scheme, a large option range was analyzed. These analyses were conducted on a 7x7 channels lattice, watching the residence time and

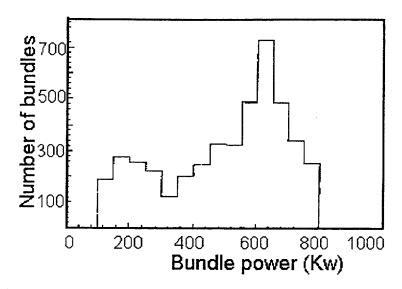


Figure 11. Number of bundles for different power intervals

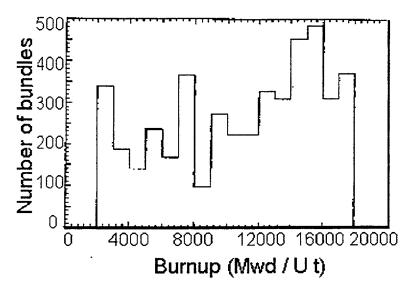


Figure 12. Number of bundles for different burnup intervals

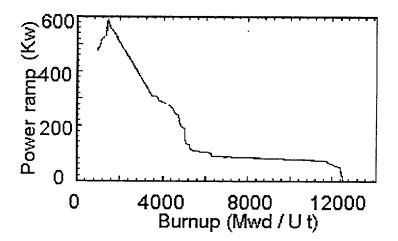


Figure 13. Power increase envelope during refueling power ramps (time average calculations)

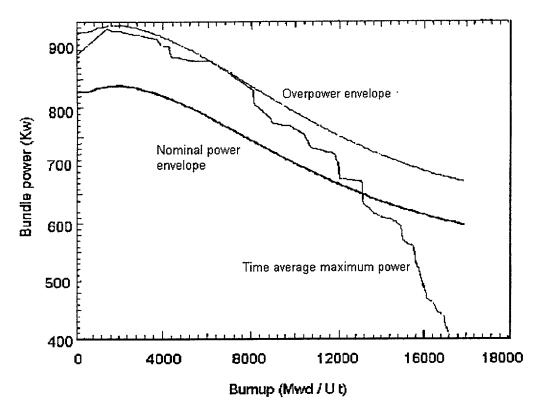


Figure 14. Power envelopes

the requirement conformation for different axial handling schemes. Among these schemes, to be analyzed at the core level, the following axial handling schemes were selected:

- 4 bundles;
- 4+2 bundles;
- 2 bundles.

These were analyzed with regard to a parameter series which define the core configuration, for example:

- burnup zones number;
- burnup zones dimensions;
- axial shift scheme

and the restrictions imposed by the maximum channel and bundle powers.

The tridimensional diffusion calculations were coupled with thermalhydraulic analyses for selecting an adequate configuration set. These have the role of providing critical channel power estimations for the chosen axial power distribution. For that, one can use as a refueling scheme selection criterion a maximum ratio between the critical and nominal channel powers.

The refueling scheme which proves convenient consists in feeding the central zone with 2 bundles and the external zone with 4 bundles. For this scheme, one have 3 burnup concentric zones on which one imposes different discharge burnup degrees.

# TABLE 1. BURNUP ZONES MAP

A	1	2	3	4	5	6	7	8	9 1	10 1	11 1	12 1	13 1	14 1	15	16	17	18	19	20	21	22
В						1	1	1	1	1	1	1	1	1	1	1	1					
ĉ					1	ĩ	1	1	1	3	3	3	3	1	1	1	1	1				
Ď				1	1	ĩ	3	3	3	3	3	3	3	3	3	3	1	1	1			
Ē			1	1	1	3	3	3	3	3	3	3	3	3	3	3	3	1	1	1		
F			1	1	1	3	3	3	3	3	3	3	3	3	3	3	3	1	1	1		
Ĝ		1	1	ī	1	3	3	3	3	3	3	3	3	3	3	3	3	1	1	1	1	
H		î	1	ĩ	3	3	3	3	3	3	3	3	3	3	3	3	3	3	- 1	1	1	
J	1	1	1	3	3	3	3	3	2	2	2	2	2	2	3	3	3	3	3	1	1	1
ĸ	1	1	1	3	3	3	3	3	2	2	2	2	2	2	3	3	3	3	3	1	1	1
Ĺ	1	1	1	3	3	3	3	3	2	2	2	2	2	2	3	3	3	3	3	1	1	1
M	1	ī	1	3	3	3	3	3	2	2	2	2	2	2	3	3	3	3	3	1	1	1
N	ī	1	1	3	3	3	3	3	2	2	2	2	2	2	3	3	3	3	3	1	1	1
0	1	Ī	1	3	3	3	3	3	2	2	2	2	2	2	3	3	3	3	3	1	1	1
P		1	1	1	1	3	3	3	3	3	3	3	3	3	3	3	3	1	1	1	1	
Q		Ī	1	1	1	3	3	3	3	3	3	3	3	3	3	3	3	1	1	1	1	
R			1	1	1	3	3	3	3	3	3	3	3	3	3	3	3	1	1	1		
S			1	1	1	3	3	3	3	3	3	3	3	3	3	3	3	1	1	1		
Ť			_	1	1	1	3	3	3	3	3	3	3	3	3	3	1	1	1			
Ū					1	1	1	1	1	3	3	3	3	1	1	1	1	1				
v						1	1	1	1	1	1	1	1	1	1	1	1					
W									1	1	1	1	1	1								

#### TABLE 2. REFUELING SCHEME

			-		-			<u>^</u>		••						• /		10	10	•		
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22
A									4	-4	4	-4	4	-4								
В						4	-4	4	-4	4	-4	4	-4	4	-4	4	-4					
C					4	-4	4	-4	4	-4	4	-4	4	-4	4	-4	4	-4				
D				4	-4	4	-4	4	-4	4	-4	4	-4	4	-4	4	-4	4	-4			
E			4	-4	4	-4	4	-4	4	-4	4	-4	4	-4	4	-4	4	-4	4	-4		
F			-4	4	-4	2	-2	2	-2	2	-2	2	-2	2	-2	2	-2	4	-4	4	4	
G		-4	4	-4	4	-2	2	-2	2	-2	2	-2	2	-2	2	-2	2	-4	4	-4	4	
н		4	-4	4	-2	2	-2	2	-2	2	-2	2	-2	2	-2	2	-2	2	-4	4	-4	
J	4	-4	4	-2	2	-2	2	-2	2	-2	2	-2	2	-2	2	-2	2	-2	2	-4	4	-4
ĸ	-4	4	-4	2	-2	2	-2	2	-2	2	-2	2	-2	2	-2	2	-2	2	-2	4	-4	4
L	4	-4	4	-2	2	-2	2	-2	2	-2	2	-2	2	-2	2	-2	2	-2	2	-4	4	-4
Μ	-4	4	-4	2	-2	2	-2	2	-2	2	-2	2	-2	2	-2	2	-2	2	-2	4	-4	4
N	4	-4	4	-2	2	-2	2	-2	2	-2	2	-2	2	-2	2	-2	2	-2	2	-4	4	-4
õ	-4	4	-4	2	-2	2	-2	2	-2	2	-2	2	-2	2	-2	2	-2	2	-2	4	-4	4
P		-4	4	-4	2	-2	2	-2	2	-2	2	-2	2	-2	2	-2	2	-2	4	-4	4	
Q		4	-4	4	-4	2	-2	2	-2	2	-2	2	-2	2	-2	2	-2	4	-4	4	-4	
R			4	-4	4	-2	2	-2	2	-2	2	-2	2	-2	2	-2	2	-4	4	-4		
S			-4	4	-4	4	-4	4	-4	4	-4	4	-4	4	-4	4	-4	4	-4	4		
Т				-4	4	-4	4	-4	4	-4	4	-4	4	-4	4	-4	4	-4	4			
U					-4	4	-4	4	-4	4	-4	4	-4	4	-4	4	-4	4				
V						-4	4	-4	4	-4	4	-4	4	-4	4	-4	4					
W									-4	4	-4	4	-4	4								

BURNUP ZONE	1	2	2	3
AXIAL REFUELING SCHEME	2	2	4	4
BURNUP(MWD/T)	15410	16050	16050	13530
AVERAGE BURNUP(MWD/T)		150	50	

#### TABLE 4. CHANEL POWER DISTRIBUTION (KW)

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22
Α													3494									
В														4475								
С					3453	4125	4718	5131	5362	5274	5248	5249	5275	5363	5132	4719	4127	3454				
D				3602	4357	4996	5299	5585	5734	5751	5647	5647	5751	5735	5586	5300	4998	4359	3603			
E			3529	4453	5140	5455	5772	5959	6025	6001	5877	5878	6002	6026	5960	5774	5457	5142	4454	3531		
F			4356	5193	5698	5836	6009	6099	6050	6009	5948	5948	6010	6051	6101	6011	5838	5700	5195	4358		
G		3971	4959	5711	5977	5988	6132	6186	6119	6098	6117	6117	6099	6120	6188	6133	5990	5980	5713	4961	3973	
н		4501	5455	6043	5961	6066	6157	6203	6179	6182	6220	6221	6183	6180	6204	6159	6068	5873	6046	5457	4503	2//0
J	3651	4855	5794	6053	6065	6124	6179	6224	6228	6295	6311	6312	6296	6290	6225	6181	6127	6067	6055	5796	4857	3652
K	3859	5130	6029	6204	6137	6163	6224	6257	6316	6291	6217	6218	6291	6317	6259	6226	6165	6140	6207	6032	5132	3861
L	3991	5273	6164	6328	6306	6310	6293	6291	6330	6282	6176	6177	6283	6332	6293	6296	6313	6308	6331	6167	5276	3993
М	3993	5279	6181	6364	6369	6373	6329	6307	6336	6283	6176	6176	6284	6337	6308	6331	6375	6371	6367	6183	5282	3994
Ν	3861	5141	6067	6296	6342	6364	6317	6292	6326	6291	6216	6216	6292	6327	6294	6319	6367	6345	6299	6069	2143	3802
0	3644	4854	5815	6127	6254	5310	6254	6243	6286	6289	6306	6306	6290	6288	6245	6256	6312	6257	6129	5424	4820	3043
Р		4470	5422	6024	5971	6076	6139	6169	6147	6162	6212	6212	6162	6149	6171	6140	6078	5913	6026	5974	4472	
Q		3904	4862	5577	5812	5829	6000	6083	6045	6061	6114	6114	6061	6046	6084	6002	2831	5814	5579	2804	3903	
R																			4962			
S			3365	4193															4195	3300		
Т				3385										5517					3380			
U					3256									5123				3230				
v						2974	3532	4008						4260	4009	3333	2915					
w									3239	3316	3389	5589	3316	3239								

The burnup zone distribution is shown in table no. 1 and the distribution of the regions in which the shift is done with 2, respectively with 4 bundles is shown in the table no. 2. One can see the burnup zones limits do not correspond to those of the different handling zones limits, in the second zone being both 2 bundles fed channels and 4 bundle fed channels. This aspect comes even more clearly from table no. 3, in which the discharge burnup on the 3 regions are shown.

The principal elements that characterize the equilibrium core, considering this refueling scheme, are presented in table no. 4, which contains the channel power distribution, and in figures from 11 to 14, as follows:

- the bundles number distribution versus power intervals (figure no. 11);

- the bundles number distribution versus burnup (figure no. 12);
- power increase envelope during refueling power ramps (figure no. 13);

- power envelope (figure no. 14).

For setting the fuel bundle operating requirements the instantaneous power distribution is essential. To obtain the data required for fuel behavior analysis the core operation and refueling were simulated. These simulations results are reported by I. Patrulescu in ref. /11/. The calculations started from an instantaneous situation generated by the random age method, simulating the refueling on a 200 days interval

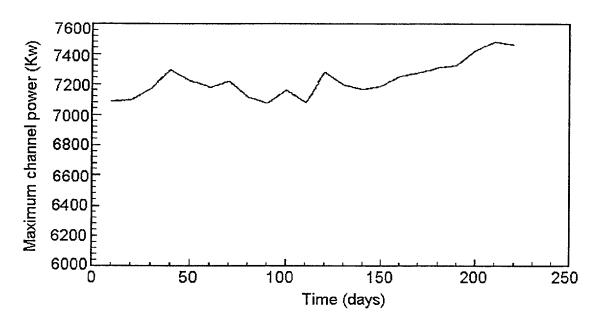


Figure 15. Maximum channel power refueling transient

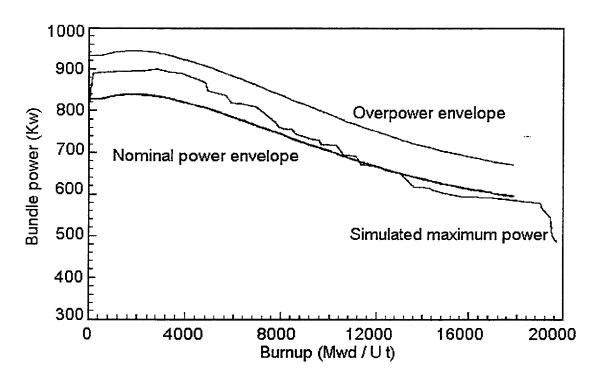


Figure 16. Bundle power envelopes (refueling simulation)

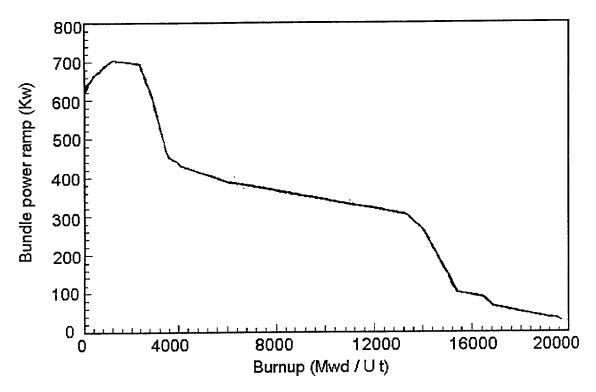


Figure 17. Power increase envelope during refueling power ramps

TABLE 5. DRY OUT CRITICAL CHA	VEL POWER DISTRIBUTION (KW).
-------------------------------	------------------------------

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22
Α									5825	6112	5845	5845	5285									
в						5697	5992	7043	7311	7347	7669	7669	7346	7311	7043	5992	5697					
С					5993	6471	7347	7757	8049	8414	8498	8498	8414	8049	7757	5347	6471	5993				
D				6046	6815	7593	7945	7644	8853	8977	9041	9041	8976	8853	7644	7946	7593	6815	6046			
Ē			6332	6773	7830	8319	8678	8901	9103	9189	9454	9454	9189	9103	8901	8678	8319	7830	6773	6332		
F			6913	7739	8457	8823	9000	9183	9148	9235	9194	9194	9235	9148	9183	9000	8823	8457	7739	6913		
Ġ		6509											9157								6509	
й													9089									
ī	5979												9199									5979
ĸ													9145									
1													9110									
м													8802									
N													9136									
ö	6321												9112									
p	0321												9188									
ò													9286									
Q R		0410											9266								• • • •	
s			5891										9484									
о т			J091										9033							5702		
1				0118									8805						0110			
U					5737													5151				
V						5356	6013	6833					7259		0833	0013	2220					
w									5862	3982	6226	6227	5981	3862								

with a 10 days step. The refueled channel determination was done off-line, following the procedure and the refueling rules for natural Uranium. A 4 % deviation from the adequate value was admitted for the maximum channel power, based on the previous NUCCP thermalhydraulics analyses of relative differences of critical power (dry out). In figures no. 15 to 17 the results of these simulations are given, respectively:

- maximum channel power refueling transient (figure no. 15);
- bundle power envelope (figure no. 16);
- power increase envelope during refueling power ramps (figure no. 17);

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22
A B						(000	7115	77 47	7121	7449 8080		8299	7449									
Ċ					6940					8795							7691	6041				
Ď				6990						9193								** • •	6990			
Ē			7153	7781	8291	8767													7780	7154		
F			7711	8249	8704	8986													-8249	7711		
G		7444	8167	8550	8910	9160	9730	10009	10397	10523	10373	10373	10523	10397	10009	9730	9160	8910	8550	8167	7444	
Н		7888	8467	8931	9086	9341	9884	10263	10475	10422	10474	10474	10422	10475	10263	9884	9341	9086	8931	8466	7888	
J	7211	8155	8686	9054	9380	9615	10305	10702	10562	10596	10518	10518	10596	10562	10702	10305	9615	9380	9054	8686	8154	7212
К	7427	8292			9570																8292	
L		8852			9871																	
M	8273	8407			10045																	7570
N		8348			9815																	
0 0	7359	8177			9482																	7359
P					9188																	
Q R		7417			9151																7417	
S					8702 8133															7717		
т			/010	6996		8186														7018		
Ū.				0770	6843	7646								8703					0990			
v					0045	6709		7647		7995					7647			0645				
Ŵ						0,00	1.400	/04/		7403					1041	1400	0700					

## 4. SEU fuel thermalhydraulics conditions evaluations

These evaluations were conducted only for defining the requirements imposed by the thermalhydraulics conditions and estimating the compliance with these conditions. The primary circuit characteristics are considered unchanged with respect to those for natural Uranium, the principal change reffering to both the radial and especially axial core power distributions. To perform these analyses the NUCCP code was used, a code used by AECL for designing the primary circuit and setting the operation conditions (ref. /12/). In the first phase, the global circuit characterization calculations were done. For this geometry one determines the thermalhydraulic parameters distribution in the entire circuit, especially the pressure drops and flows. The results were very close to those obtained for natural Uranium, the differences being not essential.

In the second phase, analyses were conducted channel by channel, considering the circuit part between the two headers in detail and imposing boundary conditions, respectively maintaining unchanged the pressure drop between the two headers. For each channel, the detailed thermalhydraulic parameters distribution was determined, for example: temperatures, pressure drops, flows, title and critical power (dry out and melting).

In ref. /13/, L. Bratu estimated, by this method, the thermalhydraulic parameters for SEU 0.9 % fueled core. These critical powers distributions are shown in the table no. 5 ( for dry out) and table no. 6 (melting). The importance of these assessments consists in the fact that they permit a global and rapid evaluation for the safety requirements compliance. One can see the critical ratios ( the ratio between the critical and the nominal channel powers) are, generally, higher for SEU than for natural Uranium. This is due to the axial channel distribution asymetry, with the maximum

shifted to to the channel inlet and having lesser values at the channel outlet, where the conditions of dry out are more rapidly created. This constitutes an important advantage, allowing an increased flexibility in reactor operation by larger margins to the safety limits. This advantage was properly taken into account when the simulation of refueling was conducted.

In this direction a preliminary study series was done to analyze to what extent can the existing ROP system for natural Uranium be used for SEU 0.9%. In ref. /14/, O. Nainer determined the critical parameters associated to a limited set of 48 distributions selected and computed by V. Raica in ref. /15/. Although they have an introductory character, these studies allow one to be aware of the possibility of using the detectors system associated ROP for natural Uranium, eventually considering a new set of starting thresholds.

## 5. The reactivity devices performance evaluation

One of the physics analyses objectives for SEU reactors is to demonstrate that the reactivity devices can perform the same functions as for the natural Uranium reactor. This means two main issues:

- setting the requirements, that is indentifying how much these must be modified, in terms of the reactivity need, at the change between natural Uranium and SEU;

- the devices effectiveness modification evaluation at the switch between natural Uranium and SEU.

The requirements modification appraisal was made based on the previous assessments of the reactivity effects.

The modification of the devices effectiveness has two major reasons:

- the devices perturbing effect modification that must be identified at super cell level and comes into the incremental cross sections modification which characterizes the particular device;

- the flux macrodistribution modification at the core level that implies the modification of the weight the respective perturbation has in the neutron balance. This effect is treated at the same time with the core flux and power distribution.

Following, a brief presentation of the principal results on these reactivity devices performances evaluations done by P. Laslau in ref. /16/ is shown.

#### The mechanical control absorbers:

The preliminary results indicate a reduction from 11 mk (for natural Uranium) at 9 mk. For SEU fuel at equilibrium, the positive reactivity that must be covered at reactor shutdown is less than 4 mk, having a temperature reactivity coefficient with 10 % less than that for natural Uranium. The value is greatly overcome by rods effectiveness. In reality, the rods were dimensioned to satisfy this requirement for fresh fuel, situation in which the temperature reactivity effect is more stressed but does not

present interest for these analyses, in which one can assume the equilibrium comes by transition from natural Uranium.

## The liquid zone control system:

The fuel burnup results in a reactivity variation that changes from 0.4 mk/d for natural Uranium to 0.5 mk/d for SEU. The changes in thermalhydraulic condition imply positive or negative reactivities with respect to the changing parameter and they are generally smaller for SEU with about 10 %. The reactivity need for power level modification depends on required speed and dynamic characteristics of the core. One can consider that all these needs will be generally smaller, taking into account that both the lifetime and the delayed neutron fraction have smaller values.

Preliminary evaluations indicated a total zone control system efficiency from 7 mk for natural Uranium to 5.5 mk for SEU. Till now, it was not identified any situation in which the zone control system could not properly react. This result may be explained either by the reduction of reactivity requirements at the switch between natural Uranium and SEU or by the possible overdimensioned system capacity for natural Uranium. A particular aspect which confirms this statement is linked with the control system capacity to suppress Xenon oscillations, that were analyzed in full detail together with the Xenon effects. In figure no. 9, which presents the left-right flux divergences evolution for setting off the Xenon divergent oscillations for the first azimuthal mode, the same deformations evolution in the case of control system activation is shown. One can see the difference is reduced in the magnitude and damps rapidly. This aspect stands out in figure no.11, where the front-back first axial mode induced deformation is shown.

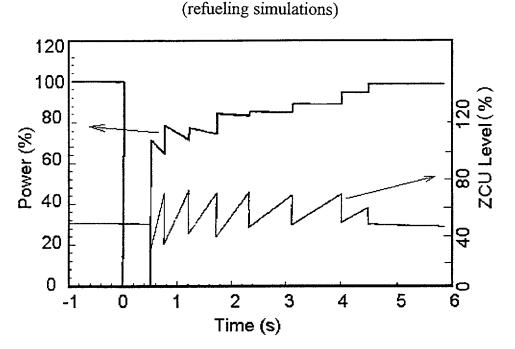


Figure 18. Restart power transient, actual scheme

#### The adjuster rod system

From the point of view of requirements, the value of reactivity margin is reduced both for control and for Xenon. Keeping in mind that the adjuster rods are permanently present in the core, influencing directly the power distribution, the adjuster rod system was treated in more detail, even in these preliminary evaluations. The adjuster rods role was virtually treated in the equilibrium core calculations by determining the power distribution that complies with the design requirements.

P. Laslau analyzed the possibility of restarting after a short shutdown, demonstrating in ref. /17/ that the adjuster rods can be used for this situation in the same manner as for natural Uranium, as one can see in figure 18, in which this dynamic analysis is shown. In ref. /18/ one analyzed the reactivity shim at power reductions. The calculations show that at a power reduction of 50 %, the reactivity due to Xenon reaches after 2.4 hours a maximum of 39.6 mK, compared with 27.8 mK initial value.

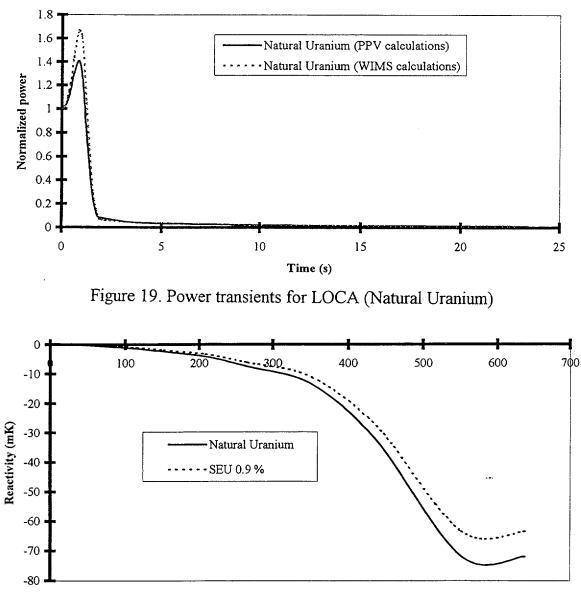
This represents the limit situation that could still be compensated by adjuster rods system, considering their reactivity is 11.8 mK, and by their extraction, the flux deformation decreases the shape factor to 0.356. In this paper, one analyzed the possibility of using the adjuster rods for compensating the negative reactivity induced by fuel burnup, in the situation of loss of refueling, determining a prolongation of operation by 2 days ( at the same time with the power reduction up to 95 %) by the second bank extraction. These results were obtained for the actual scheme of banks, the analyses proving that for SEU fuel there are more effective grouping possibilities, regarding the functioning at refueling capacity loss and with similar restarting performance.

## 6. Shutdown system performances

For natural Uranium, the shutdown system performances are evaluated on the power transient basis, in the LOCA time. For that purpose, for SEU 0.9% one must analyze to what extent this transient evolution is modified by reactivity effect increment due to the void and by the shutdown system effectiveness reduction. An additional problem is associated with the uncertainties related to the prediction of the void effect reactivity for the irradiated fuel case. The differences between WIMS and POWDERPUFS estimations are well known even for natural Uranium.

To illustrate this, in figure no. 19 the power evolution for natural Uranium fed core is shown, computed by WIMS and PPV constants for the same evolution of coolant density (corresponding to a RIH break of 20 %). The evolution is more rapid for the transient computed by WIMS constants, reaching a maximum of 160% of the nominal power, compared with 140 % obtained in the PPV based calculations. This is explained by the more emphasized void effect predicted by WIMS.

For the SEU 0.9 % reactor, one has a higher value of void effect for the fresh zone. For equilibrium fuel yet, one has some small differences between natural Uranium and SEU for void effects. This is the result of the void effect behavior which diminishes with burnup following the Pu gathering. The void effect for SEU, though



Insertion depth (cm)

Figure 20. Reactivity insertion for Natural Uranium compared with SEU 0.9 %

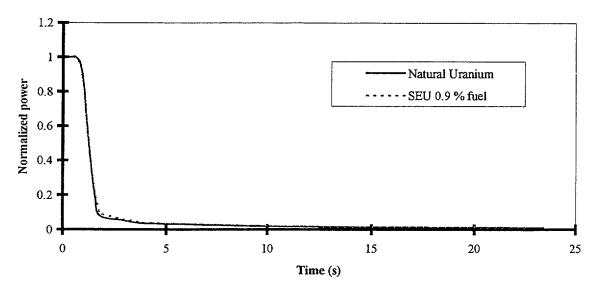


Figure 21. Power transients for reactor shutdown (SDS 1)

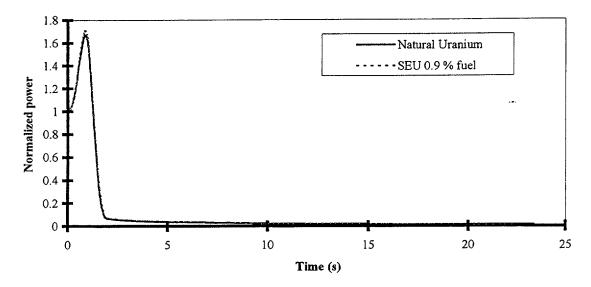


Figure 22. Power transients for LOCA

initially has a higher value, will present a sharper reduction because of the almost double discharge burnup. In this way, for SEU 0.9 % at equilibrium, one will obtain values close to those obtained for natural Uranium, effect predicted both by PPV and WIMS.

For the SDS 1 efficiency modification, compared with natural Uranium, for SEU 0.9 %, though one can see a reactivity insertion curve difference illustrated in figure no. 20, the effect in power evolution is greatly diminished, especially on the first part where usually the critical conditions are reached, as one can see in figure no. 21 where the power transients generated by rods insertion for the two types of fuel are shown.

A comparison of power evolution for the two types of fuel is shown in figure no. 22. This shows an assessment of the shutdown system capacity to limit the power evolution in the LOCA case, more exactly an assessment of the modification of this capacity at the switch between natural Uranium and SEU 0.9 %. To set off the differences, WIMS calculations were done both for natural Uranium and SEU 0.9 %, considering the same density evolution curve. One can remark the very close evolutions, the differences being of a few percent order, containing both the void effect modification and the efficiency reduction at the switch between natural Uranium and SEU fuel.

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## CANFLEX-RU FUEL DEVELOPMENT PROGRAM AS ONE OPTION OF ADVANCED FUEL CYCLES IN KOREA



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#### Abstract

As one of the possible fuel cycles in Korea, RU (Recycled Uranium) fuel offers a very attractive alternative to the use of NU (Natural Uranium) and SEU in the CANDU reactors, because Korea is a unique country having both PWR and CANDU reactors. Korea can therefore exploit the natural synergism between the two reactor types to minimise overall waste production, and maximise energy derived from the fuel, by burning the spent fuel from its PWR reactors in CANDU reactors. Potential benefits can be derived from a number of stages in the fuel cycle: no enrichment required, no enrichment tails, direct conversion to U0<sub>2</sub>, lower sensitivity to <sup>234</sup>U and <sup>236</sup>U absorption in the CANDU reactor, expected lower cost relative to NU and SEU. These benefits all fit well with the PWR-CANDU fuel cycle synergy. RU arising from the reprocessing of European and Japanese oxide spent fuel by 2000 is projected to be approaching 25,000 te. The use of RU fuel in a CANDU-6 reactor should result in no serious radiological difficulties and no requirements for special precautions and should not require any new technologies for the fuel fabrication and handling. A KAERI's feasibility shows that the use of the CANFLEX bundle as the carrier for RU will be compatible with the reactor design, current safety and operational requirements, and there will be no significant fuel performance difference from the CANDU 37-element NU fuel bundle. Compared with the 37-element NU bundle, the RU fuel has significantly improved fuel cycle economics derived from increased burnups, a large reduction in fuel requirements and spent fuel arisings and the potential lower cost for RU material. There is the potential for annual fuel cost savings to be in the range of one-third to two-thirds, with enhanced operating margins using RU in the CANFLEX bundle design. These benefits provide the rationale for justifying R & D effort on the use of RU fuel for advanced fuel cycles in the CANDU reactors of Korea. The RU fuel development is an international collaboration between KAERI, AECL and BNFL. It is expected that the work will be completed before 2005, and there should be no impediment to the use of RU fuel in the CANDU-6 reactors in Korea, if the RU in the world is available and competitive with NU and SEU on price.

#### 1. INTRODUCTION

In Korea, twelve nuclear power plants, (10 PWRs and 2 CANDUs) are currently in operation, and six plants (4 PWRs and 2 CANDUs) are under construction. In 1997, the existing power plants represent about 25 % (10,316 MW) of the domestic installed generating capacity, and produced about 34 % (77,086 GWh) of the gross electrical energy generation. In 2002, a total nuclear power generation capacity of 15,742 MWe will be installed in Korea, where 18 % of the capacity will be contributed by the 4 Wolsong CANDUs. Korea is therefore a unique country in the world having both PWR and CANDU reactors, and can exploit the natural synergism between these two reactor types to minimise overall waste production, and maximise energy derived from the fuel. The synergism can be exploited through several different fuel cycles [1]. In conventional reprocessing, which is currently available from

several sources, uranium and plutonium are separated from the fission products and other actinides in the spent fuel. The plutonium could be recycled as MOX fuel, in either LWR's or in CANDU reactors. If the political and non-proliferation considerations in the Korean peninsula led to the decision to reprocess the Korean spent PWR fuel, then the resultant recovered uranium, which constitutes the vast majority of the spent fuel, and which still contains valuable <sup>235</sup>U (typically about 0.9%), could be recycled as-is in CANDU reactors, without re-enrichment. The fuel burnup in CANDU would be about double that of natural uranium fuel, and about twice the energy would be extracted, compared with re-enrichment and recycling in a PWR. However, the use of RU in Korean CANDU reactors is not dependent on reprocessing Korean spent PWR fuel; RU is a nuclear fuel commodity available from several sources, as is natural uranium, and enriched uranium. Hence, RU as a fuel cycle option in Korea is particularly attractive for use in the CANDU reactors, with the advantage of potentially lower fuelling costs than both NU and SEU.

KAERI (Korea Atomic Energy Research Institute) has a comprehensive product development program on CANFLEX (CANDU Flexible Fuelling) - RU (Recovered Uranium) fuel[2]. This is seen as an economical alternative to natural uranium as a fuel for use in either existing or future CANDU reactors. The aim is to introduce CANFLEX into CANDU reactors in Korea and have a clear vision of how the product will evolve over the next 10 years. The key targets of the program are enhanced safety and economics, the reduction of spent fuel volumes, using the inherent characteristics and advantages of CANDU technology. The specific activities of the program take account of the domestic and international environment concerning non-proliferation in the Peninsula of Korea[2]. These involve showing an overall evaluation and identification of the potential benefits, risks, and costs associated with the use of RU fuel to a CANDU-6 utility by 1999. This will provide a rationale to justify the R & D efforts on it for the advanced fuel cycle of CANDU reactors in Korea. The justification includes security of supply issues for RU and the overall possibility of satisfying the licensing issues in the Korea Safety Review Guideline (KSRG)[3]. These external influences and justifications have been, and will be applied to all fuel and fuel cycle R & D in Korea. The RU fuel R & D program has been enhanced by an international collaboration between KAERI, AECL(Atomic Energy of Canada Limited) and BNFL(British Nuclear Fuels Plc), since the end of 1996. The prime objective of this joint program is the small-scale demonstration irradiation of 20 to 100 bundles in a CANDU power reactor, followed by the post irradiation examination of selected irradiated bundles. This is a necessary prerequisite to a full-scale conversion to RU. The program includes the necessary analysis and out-of-reactor tests.

The intent of this paper is to evaluate the advantages and feasibility of CANFLEX-RU in order to provide a rationale for justifying the R & D efforts on it for an advanced fuel cycle in Korea.

# 2. CANFLEX AS THE REFERENCE CARRIER OF RU IN CANDU

To allow the benefits of RU to be maximised, an appropriate carrier is required. This is achieved through the provision of enhanced operating margins in the CANFLEX bundle design [4]. Since 1991, KAERI and AECL have pursued a collaborative program to develop, verify and prove the design of CANFLEX which is a 43 element CANDU fuel bundle and acts as the reference carrier of the RU fuel. The CANFLEX bundle has the same bundle diameter and length as a CANDU-6 37-element natural uranium (NU) bundle. The principle features of CANFLEX are enhanced thermalhydraulic performance and more balanced radial power distribution, providing CANDU plant operators with greater operating flexibility through improved operating margins. Critical heat flux (CHF) enhancement appendages on the bundle enable a higher bundle before CHF occurs, leading to a net gain in the critical channel power of 6 % to 8 % over the existing 37-element fuel bundle. The maximum linear element rating in a CANFLEX bundle is 20 % lower than that of the conventional bundle,

reducing the consequences of most design-basis accidents. The lower element rating is achieved by adding extra elements and using larger diameter element in the 2 center rings and smaller diameter ones in the outer 2 rings. These features will provide larger operating margins in existing CANDUs, and will allow higher burnups. New Brunswick Power at the Point Lepreau Generating Station in Canada will irradiate 24 CANFLEX-NU fuel bundles over a 2-years period starting in 1998, as a final verification of CANFLEX design in preparation for full-core conversion [5].

## 3. CANDU-6 REACTOR PHYSICS, THERMALHYDRAULICS, SAFETY AND FUEL PERFORMANCE OF CANFLEX-RU

AECL and KAERI have performed reactor physics simulations to evaluate the feasibility of CANFLEX-RU fuel being used in a CANDU-6 reactor by taking the isotopic composition of typical RU UO<sub>2</sub> produced by the MDR route.

RU is one of the products of conventional chemical reprocessing of spent uranium oxide fuel. The extra isotopes in RU have minimal effect on the reactor physics characteristics in CANDU. Spent PWR fuel contains typically 0.4 % <sup>236</sup>U with a range from 0.2 % to 0.7 %, originating from neutron capture in <sup>235</sup>U in the original PWR fuel, that has a strong resonance at 5.5 eV. Because of the softer neutron spectrum in a CANDU reactor, the absorption worth of the <sup>236</sup>U is an order of magnitude lower in a CANDU than in a PWR. Also, the <sup>235</sup>U would be burned down to low levels (i.e. 0.2 to 0.3 %) in a CANDU reactor because of the good neutron economics provided by the heavy water moderator and coolant, compared with PWRs (0.8 % to 1.0 %)[6]. Therefore, the main determinant in CANDU reactor physics with RU is the <sup>235</sup>U level.

In AECL[7], 500 FPD (Full Power Days) core-follow simulations were made for CANFLEX-RU with 0.96 w/o<sup>235</sup>U, using a bi-directional 2-bundle-shift refuelling scheme, where standard computer codes and methods were used for the simulations and analysis. WIMS-AECL[8] with ENDF/B-V nuclear data library was used to construct fuel tables for use with a core code, RFSP[9], which modelled the reactor core. To facilitate the decisions that must be made during refuelling, an automated method was used to do most of the editing and calculations required to perform the steps. The results of the CANFLEX-RU core-follow simulations show that the RU fuel would be a satisfactory fuel in a CANDU-6 equilibrium core : maximum bundle power of 857 kW(c.f. the license limit of 935 kW); maximum chnnel power of 7.021 MW (c.f. the license limit of 7.3 MW); average discharge burnup of 1394 MWd/kgU. An assessment was made of the probability of stress-corrosion cracking (SCC) through power boosting using the results of the AECL refuelling simulation. The CANFLEX-RU elements do not come close to approaching the SCC element-power threshold, and none of the linear-element powers were above 44 kW/m as shown in Fig. 1.

In KAERI, the reactor lattice calculations for various bundle types of CANFLEX-0.9 % RU were preliminarily performed with WIMS-AECL code to investigate the bundle types with respect to Korean safety regulation of power coefficient. The results of the WIMS-AECL reactor lattice calculations show that the power coefficient of the CANFLEX bundle type with RU in all elements is positively increased, compared with that of a 37-element fuel bundle with natural uranium as shown in Table 1. However, the power coefficient of a CANFLEX-RU bundle with, for example, stainless steel in the centre element is negatively increased, compared with that of the 37-element fuel bundle. RFSP time-averaged (reaction rate averaged) calculation results maximum bundle power of 775 kW (c.f. the licence limit of 935 kW), maximum channel power of 6.57 MW (c.f. the licence limit of 7.3 MW) and average discharge bunrup of 13,375 MWd/MTU which is 88 % higher than that of the 37-element fuel bundle with natural uranium. Statistic reactivity worths of adjuster rods, zone controllers and mechanical control absorbers in CANDU-6 equilibrium core were investigated for the CANFLEX-RU bundle with 0.90 w/o <sup>235</sup>U, using with a bi-directional 4-bundle-shift refuelling

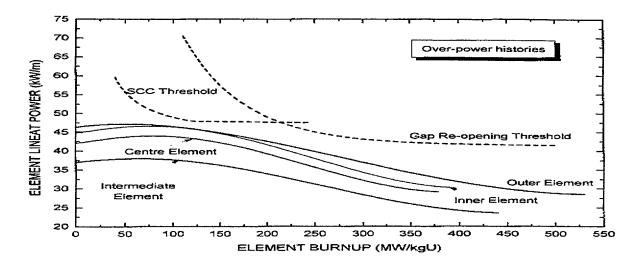


Fig. 1. Defect Analysis of CANFLEX-RU(0.98 % U-235 in total U) Fuel Bundle With Respect To SCC Threshold

Table 1. Characteristics of	WIMS-AECL Time-Averaged Lattice Parameters for
CANDU-6 Existing	Fuel and Possible Bundle Types of CANFLEX-RU

	<u> </u>	Coolant	Fuel	Coolant	Moderator	Average	Power
	MLHR	Void	Temp.	Temp.	Temp.	Discharge	Coeff.
		Reactivity	Coeff.	Coeff.	Coeff.	Burnup	000011.
Fuel Types	(kW/m)	(mk)	(mk/ <sup>o</sup> C)	(mk/ <sup>o</sup> C)	(mk/ <sup>0</sup> C)	(MWd/MtU)	(mk/%FP)
37-elem.(NU37)	58.031	14.49664	-0.00123	0.05302	0.02870	7,087	~0.0
CANFLEX(NU43)	48.879	15.51087	-0.00143	0.05616	0.02987	7,002	-0.001096
CANFLEX(RU43)	49.398	15.91435	-0.00104	0.05919	0.03937	13,040	0.002872
CANFLEX	51.319	15.44110	-0.00114	0.05810	0.03910	12,993	0.001792
(C1+RU42)							
CANFLEX	57.206	12.69371	-0.00146	0.05132	0.04217	12,389	-0.002323
(C8+RU35)							
CANFLEX	50.682	13.39525	-0.00207	0.05188	0.01972	10,474	-0.007426
(ST1+RU42)							
CANFLEX	50.792	13.73889	-0.00190	0.05305	0.02313	10,889	-0.005730
(Fe 1+RU42)							
CANFLEX	51.245	15.32182	-0.00116	0.05764	0.03847	12,820	0.001552
(Al 1+RU42)							
Remarks	NU : Natu	Iral Uranium	RU : Recov	ered Uraniu	n(0.9 %U-235	in total U);	
						d in center ring	
					n center ring,		,
		ber of rods			an oonton mig,		
L		Der of Tous			<del> </del>		

Table 2. Static Reactivity Worth of Reactivity Control System in CANDU-6 Equilibrium Core

Fuel Types	Adjuster Rods	Zone Controllers	Mechanical Control Absorbers
37-elem.	16.6 mk	6.5 mk	-11.3 mk
(NU37)	(> Xe buildup:13 mk)		
CANFLEX	14.7 mk	7.3 mk	-8.5 mk
(RU43)	(> Xe buildup:12 mk)		
Remarks	Compatible with	Compatible with	Required to be investigated
	CANDU-6 design	CANDU-6 design	in more detail

scheme. In CANDU-6, the adjuster rods are provided for xenon override capability needed to restart the reactor after a short shutdown, power manoeuvering during startup or power derating, reactivity shim when fuelling is temporarily interrupted, and shaping the thermal flux distribution in the core for optimum reactor power and fuel burnup. The light water zone control system is designed to perform two main functions: bulk control (control of gross power output) and spatial control (control of flux/power shape). The mechanical control absorbers are to initiate rapid power reduction if required by operations and to override the reactivity increase following a power reduction (due to the negative fuel temperature coefficient). As shown in Table 2, the CANFLEX -0.9 % RU bundles would not required any design change or hardware modification in CANDU-6 reactor, even if the mechanical control absorbers are required in more details of investigation for the static reactivity worth. In addition, criticality of the fresh fuel on the reactor site and criticality and heat removal capability of the irradiated fuel in the storage pool are investigated to be that there are no requirement of any design or hardware modifications of the reactor site. Based on the results of RFSP time-averaged (reaction rate averaged) calculation, the radial and axial heat flux distributions for the CANDU-6 equilibrium core with the CANFLEX-RU fuel led to increase critical channel power by about 4 %, compared with the CANDU-6 equilibrium core with the 37-element natural uranium fuel. A CANDU fuel element performance analysis code, ELESTRES [10], predicted that the internal pressure of the outer CANFLEX-RU elements in normal power operation was below 2.5 MPa, which is lower than that of the outer elements of the 37-element NU fuel bundle by a factor of 2. The maximum fuel stack length of the outer and inner CANFLEX-RU elements increased by 0.46 % through thermal expansion, which is equivalent to a reduction of less than 0.2 mm in the axial gap between the fuel stack and the end cap. A preliminary safety assessment of a CANDU-6 shows that, for all the shorter half-life isotopes, the gap (or "free") inventory with CANFLEX-RU fuel is 5 - 10 times smaller than that for 37-element NU fuel, and the total inventory with RU-fuel is very similar to that for 37-element NU fuel. For the longer half-life isotopes such as <sup>137</sup>Cs, the gap inventory with CANFLEX-RU is very similar to that with 37-element NU fuel, but the total inventory with CANFLEX RU fuel is about 2 times higher than that for 37-element NU fuel, because of the higher burnup. In a preliminary fuel channel analysis for 35 % reactor inlet header (RIH) break in CANDU-6 reactor with the CANFLEX-RU, the maximum fuel centreline and sheath temperatures are resulted to be lower by 338 °C and 122°C, respectively, than those for the existing 37-element natural uranium fuel. The fuel channel integrity shows to be negligibly affected by the axial power distribution shape change of the CANFLEX-RU bundle's channel following the bundle refuelling scheme change.

#### 4. AVAILABILITY AND PROCESSING OF RU

The cumulative quantity of RU projected to arise from the reprocessing of European and Japanese spent fuel by 2000 is approaching 25,000 te [6]. This RU, which is owned by the utilities or reprocessors, is an alternative fuel source to new natural uranium for use in LWR and CANDU reactors. Each country and utility will determine its strategy for RU based upon local factors. Theoretically this 25,000 te would provide sufficient fuel for 500 CANDU-6 reactor years operation, since the initial core load of uranium for a CANDU-6 reactor is 85 t, and annual refuelling requirements for a RU fuel burnup of 13 MWd/kgU are around 50 t/a.

Current reprocessing technology has been optimised to produce an RU product suitable for interim storage pending re-enrichment and recycle into LWR reactors. BNFL uses thermal denitration to convert UNL (Uranyl Nitrate Liquor) to  $UO_3$ . COGEMA uses the ADU route to convert UNL to  $U_3O_8$ . Further processing would be required to convert this to sinterable powder. Several processes exist to convert the RU from its form used in storage, to ceramic grade sinterable powder. For example the  $UO_3$  from BNFL's THORP reprocessing plant could be further processed to UF<sub>6</sub> and the existing IDR (Integrated Dry Route) facilities used to convert the UF6 to ceramic grade  $UO_2$ . Alternatively, BNFL has a prototype facility in operation, which converts the UNL directly to a ceramic grade  $UO_3$  (subsequently to  $UO_2$ ) by

the MDR (Modified Direct Route) process. This route offers significant savings in the longer term if sufficient CANDU RU demand develops.

## 5. FABRICATION AND HANDLING OF RU CANDU FUEL

The isotopic composition and activity of un-enriched recovered U0<sub>2</sub> powders depend inter alia on the reactor type, initial enrichment and discharge burnup of the PWR fuel, the time between spent fuel discharge and reprocessing, the route chosen to convert the UNL to UO<sub>2</sub>, and the delay until fuel fabrication. RU contains typically ~1 ppb <sup>232</sup>U which decays with a half-life of 69.8 years. The daughters in the <sup>232</sup>U decay chain are removed during reprocessing but grow during storage. Conversion processes via UF<sub>6</sub> also remove daughter products. The first daughter in the chain is <sup>228</sup>Th with a half-life of 1.9 years. Since all the other daughters in the chain have much shorter half-lives, including the radiologically important <sup>208</sup>TI and <sup>212</sup>Bi, they are all in secular equilibrium with <sup>228</sup>Th. Therefore, the <sup>228</sup>Th build-up governs the rate of build-up of gamma activity and indicates the gamma activity with time relative to the quasi-equilibrium level attained after about 10 years. RU also contains <sup>234</sup>U that contributes to a higher specific alpha activity compared to NU. However, the level is about the same as in conventional enriched PWR fuel, since the source of the increased <sup>234</sup>U is the initial enrichment of NU. RU also contains trace fission product gamma and beta emitters, and transuranic alpha emitters.

An initial assessment of the health physics aspects of manufacturing and handling RU as a reactor fuel for CANDU was done in the joint program between BNFL, KAERI and AECL, and before that in a joint program between AECL and COGEMA [6]. BNFL has converted reprocessed spent PWR fuel into 200 kg of  $U0_2$ . The characteristics of the recovered  $U0_2$  powder met CANDU specifications, both in terms of chemical impurity contents and physical characteristics. The powder was granulated and pressed into green pellets, which were sintered under the normal conditions for CANDU fuel. The finished pellets met all the physical and chemical specifications for CANDU fuel.

The conversion took place one year after reprocessing. Activity level measurements made on the finished CANFLEX-RU bundle were 1.3 times higher than a natural uranium bundle, when measured at 30 cm distance. This CANFLEX-RU bundle was displayed at AECL's Sheridan Park Engineering Laboratories (SPEL) during the 5th International Conference on CANDU Fuel, 1997 September 21-25, in Toronto, Canada, where delegates were able to see and handle both RU and natural uranium bundles. Consequently, because the total fuel quantity required can be reduced by around 50% using RU, the overall dose uptake to the workforce during the fabrication and handling of RU bundles will be comparable with that presently seen for natural uranium fuel. By reducing the time from reprocessing to conversion, fuel fabrication, and insertion into the reactor, the dose uptake will be reduced even further.

During the sintering, the release of <sup>137</sup>Cs and other volatile fission products from RU was below detectable levels. Also, AECL[6] earlier concluded that no significant fields in a commercial fuel fabrication plant would build up due to release of <sup>137</sup>Cs during sintering, even after decades of production.

#### 7. FUEL CYCLE COSTS FOR RU

Most countries and/or utilities, which adopt a reprocessing strategy, do so for strategic energy self-reliance and/or for waste management reasons. Generally, RU is owned by the utility that contracts for reprocessing of spent uranium oxide fuel. The uranium and plutonium recovered from reprocessing are often held as "low or zero cost" stocks by the utilities. Hence, there is the possibility that RU will be competitively available on the open market.

The potential annual saving to a CANDU utility by the utilisation of RU is significant, but strongly dependent on the price paid for the RU powder and fuel fabrication.

The costs of the front-end of the fuel cycle (excluding back-end storage and disposal costs) in US dollars were assessed for RU in CANDU and re-enriched RU in a PWR by Boczar et al.[7], for a range of RU-cost assumptions. This parametric survey indicated that, with RU at no cost, CANDU fuelling cost with RU is >70% lower than for re-enriched RU in PWR. With RU at no cost, the CANDU fuelling cost is reduced relative to NU fuelling by 45% with NU at 25 \$/kgU, and by 67 % with NU at 80 \$/kgU. With RU at NU cost, the fuelling cost savings in CANDU with RU are 28 % for NU at 25 \$/kgU, and 34% for NU at 80 \$/kgU. With RU at NU cost, the fuelling costs are 10 - 15 % lower than for 1.2 % slightly enriched uranium (SEU), which is the economic optimum SEU enrichment.

KAERI also assessed relative annual savings of CANFLEX-RU to existing 37-element NU fuel bundles in CANDU-6 by assuming that the fabrication cost of the RU fuel bundle is about 16 % higher than that of the 37-element bundle. With recycled  $UO_2$  priced at 25 % of the natural  $UO_2$  price, the annual fuelling costs will represent a 64 % saving relative to that of NU in 37-element bundles. Similarly with recycled  $UO_2$  priced at 124 % of the natural  $UO_2$  cost, the annual fuelling cost of the RU fuel bundles would show a saving of 31 % relative to that of the 37-element bundles. Break-even between RU and NU  $UO_2$  is represented with recycled  $UO_2$  priced at 210 % of the natural  $UO_2$  price.

Ongoing work will reduce the uncertainties in the fuelling costs for RU, namely the cost of ceramic-grade UO<sub>2</sub> powder, and the cost of CANFLEX-RU fuel fabrication. Finally, another AECL paper [11] quantifies significant cost savings in the back-end of the fuel cycle with SEU (or RU).

## 8. CONCLUSIONS

Korea can exploit the natural synergism between the two reactor types of PWR and CANDU reactors to minimise overall waste production, and maximise energy derived from the fuel, by recycling the spent fuel from its PWR reactors in CANDU reactors. As one of the possible fuel cycles, RU fuel is a very attractive alternative to the use of NU and SEU in CANDU reactors, offering among other benefits, the advantage of potentially lower fuelling costs. The RU fuel development program and international collaboration between AECL, KAERI and BNFL is a part of KAERI's comprehensive development program of CANDU advanced fuel and includes a clear vision of how the product will evolve over the next 10 years. The key targets of the program are safety and economic enhancements, and reduction of spent fuel volume, using the inherent characteristics and advantages of CANDU technology.

RU is one of the products from conventional reprocessing of spent uranium oxide fuel and typically will have an overall nominal <sup>235</sup>U content of 0.9 %. The composition of un-enriched RU depends on the reactor type, initial enrichment and discharge burnup of the PWR fuel, the time between spent PWR fuel discharge and reprocessing, the route chosen to convert the UNL to UO2, and the delay until fuel fabrication. It is only slightly more radioactive than NU. The RU can be used directly in CANDU reactors. A number of options exist for the conversion of the UNL to ceramic-grade UO<sub>2</sub>, including direct conversion using MDR and ADU processes, or fluorination to UF6, followed by the IDR route. The RU available to utilities and reprocessors in Europe and Japan by 2000 cumulatively is expected to be approaching 25,000 te. This quantity of RU, if used solely for recycle in CANDU reactors, would provide sufficient fuel for some 500 CANDU-6 reactor-years of operation, since the initial core load of uranium for a CANDU-6 reactor is 85 t, and annual refuelling requirements

for RU with burnup of 13 MWd/kgU are ~ 50 t/a. Security of supply is not an issue, since SEU could be substituted for RU. The suitability of RU as a reactor fuel for CANDU has been shown: CANDU fuel fabricated from RU meets CANDU specifications; RU does not pose serious radiological difficulties, and no special precautions or technologies are required for handing RU, because the dose fields associated with RU are just slightly higher than NU; fuel management is particularly simple.

Taking the CANFLEX 43-element CANDU fuel bundle as the carrier of RU fuel, some preliminary evaluations of CANDU reactor physics, thermalhydraulics, safety and fuel performance of CANFLEX-RU indicated that the fuel would not cause excessive channel or regional overpowers, or significant risk of fuel element failure in spite of its higher burnup and slight enrichment relative to natural uranium. However, future detailed analyses of the RU fuel are required to provide a detailed rationale for the justification of the R & D efforts on it for the advanced fuel cycle of CANDU reactors in Korea. The justification includes the licensing issues in the KSRG.

With RU available free-issue, the annual fuelling costs could be reduced by ~ 30 - 60%, compared to NU fuel. With ceramic RU powder at NU cost, the fuelling costs are 10 - 15% lower than for 1.2% SEU, which is the economic optimum SEU enrichment. These cost savings are strongly dependent on the cost of ceramic grade  $U0_2$ , and the cost of CANFLEX-RU fuel fabrication. Fuel management with RU is considerably simpler than that for 1.2% SEU, and good fuel performance is assured as a result of the lower ratings with CANFLEX. In the current collaborative program between KAERI, AECL and BNFL, RU fuel development and proof testing will be completed by around 2005, and there should be no impediment to the use of RU fuel in the CANDU-6 reactors in Korea, if RU is competitively available.

RU is very attractive option of advanced fuel cycles not only in Korea, but also in World, because the use of RU in nuclear reactors improves uranium utilisation and saves new natural uranium for our second generation.

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# FUEL-MANAGEMENT SIMULATIONS FOR ONCE-THROUGH THORIUM FUEL CYCLE IN CANDU REACTORS

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## Abstract

High neutron economy, on-power refuelling and a simple fuel bundle design result in unsurpassed fuel cycle flexibility for CANDU<sup>®</sup> reactors. These features facilitate the introduction and exploitation of thorium fuel cycles in existing CANDU reactors in an evolutionary fashion. Detailed full-core fuel-management simulations concluded that a once-through thorium fuel cycle can be successfully implemented in an existing CANDU reactor without requiring major modifications.

## **1. Introduction**

The abundance of thorium in the earth's crust is about three times that of uranium. Hence, the thorium fuel cycle ensures a long-term nuclear fuel supply. For countries with abundant thorium reserves, the thorium fuel cycle in CANDU reactors would enhance both the sustainability of nuclear power and the degree of energy independence, using a single reactor type.

There are two major options for implementing the thorium fuel cycle in CANDU reactors. The first is the recycling option, in which the <sup>233</sup>U in the spent thorium fuel is recycled into the fresh fuel. The second is the once-through thorium (OTT) option, where the rationale for the use of thorium does not depend on recycling. Thorium recycling is not a near-term option because it requires extensive research and development efforts. Therefore, recent thorium fuel cycle studies conducted at AECL have focused on the OTT as the nearer-term option.

Two methods can be used to introduce the OTT fuel cycle into existing CANDU reactors. The first is a "mixed-core" approach where a large number of driver channels containing enricheduranium fuel are used to support a relatively small number of channels dedicated to thorium irradiation. Because of the disparity in reactivity and power output between driver channels and thorium channels, very sophisticated fuel-management schemes will be required to shape the channel and bundle power distributions in the mixed core in order to achieve the nominal reactor power output. This approach is theoretically feasible, but its practicality has not been investigated in detail.

An alternative approach is to fuel the whole core with mixed-fuel bundles, which contain both thorium and enriched-uranium fuel elements in the same bundle. This "mixed-fuel bundle" approach is a practical means of utilizing thorium in existing CANDU reactors, while keeping the fuel and the reactor operating within the current safety and operating envelopes established for the natural-uranium fuel cycle.

Detailed fuel-management studies were conducted using the RFSP<sup>1</sup> code for a CANDU 6 reactor using the mixed-fuel bundle approach. The lattice parameters were calculated by WIMS-AECL<sup>2</sup> for CANFLEX<sup>®</sup> mixed-fuel bundles having slightly enriched uranium (SEU) fuel in the outer 35 elements and natural thorium in the central 8 elements.

CANDU<sup>®</sup> is a registered trademark of Atomic Energy of Canada Limited (AECL).

## 2. Fuel-Management Flexibility in CANDU

The on-power refuelling feature of CANDU reactors allows the reactors to operate with the optimal channel and bundle power distributions for a wide variety of fuel cycles. In general, the reactor is divided into different regions for fuel-management purposes. Each region may contain a different type of fuel with different fissile content. Reactor criticality and global power shape can be controlled by fuelling each region at a different rate. Local power peaking can be minimized by judiciously selecting individual channels to be refuelled, and by limiting the number of fresh fuel bundles inserted into a channel during each refuelling operation.

Figure 1 shows the RFSP model of a CANDU 6 reactor divided into three fuelling regions. The inner and the outer regions are further subdivided into smaller zones having slightly different fuelling rates in order to optimize the reactor global power shape. Figure 2 shows the configuration of the fuel elements in a CANFLEX fuel bundle where the natural-thorium fuel in the 8 inner elements is surrounded by slightly enriched uranium (SEU) fuel in the outer 35 elements. There is a high degree of flexibility in the choice of the amount and the nature of the fissile material in the enriched driver fuel elements to achieve specific goals in different fuel cycles. The combination of on-power fuelling with a simple and flexible fuel bundle design offers many options for burning thorium in existing CANDU reactors.

## 3. Options for Burning Thorium in CANDU Reactors

Two options have been developed for burning thorium fuel in an existing CANDU 6 reactor. In Option 1, only one fuel type was used throughout the entire core, and the adjuster rods were removed. The reference fuel design is a CANFLEX fuel bundle with 1.8 wt % slightly enriched UO<sub>2</sub> fuel in the outer 35 elements and natural ThO<sub>2</sub> fuel in the inner 8 elements. The initial fissile content was chosen to maximize the burnup of the thorium fuel elements without exceeding the current limits on maximum channel and bundle power.

Adjuster rods provide flux and power flattening with natural-uranium fuel, but they are not needed for this purpose with enriched fuel. In fact, they would over-flatten and reduce the power in the centre of the channel, resulting in an undesirable asymmetric double hump in the axial flux and power distributions in CANDU reactors using enriched fuel. The flux and power distortions caused by the adjuster rods can be eliminated by using a sophisticated fuelling scheme<sup>3</sup>, or by using a small amount of burnable poison in the fuel bundle.

The second option illustrates the flexibility of existing CANDU reactors to accommodate both thorium fuel and adjuster rods. In Option 2, each of the three regions shown in Figure 1 contains a different type of thorium fuel bundle. The fuel in the 196 outer-region channels is the same as that used in Option 1. The fuel in the 124 inner-region channels is identical to that in the outer-region channels, except that the central  $ThO_2$  element contains 6.0 wt % of gadolinium to shape the axial flux distribution. The gadolinium-doped bundles are used only in the inner-region, which is under the influence of the adjuster rods.

The 60 periphery channels in Option 2 contain thorium bundles designed to achieve burnups of over 50 MW.d/kgHE. These high-burnup thorium bundles use natural  $ThO_2$  in all 43 fuel elements. However, the initial fissile content in the outer 35 elements is increased from 0 wt % to 1.7 wt % using 20 wt % enriched uranium. These high-burnup thorium bundles are located strategically at the edge of the core to utilize a large percentage of the leakage neutrons to produce power. This arrangement significantly increases the amount of thorium fuel in the core and improves the overall fuel efficiency of the thorium-burning reactor.

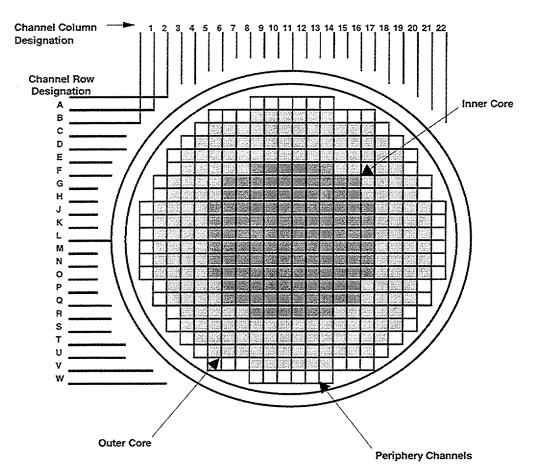


Figure 1: Reactor Core Model of a CANDU 6

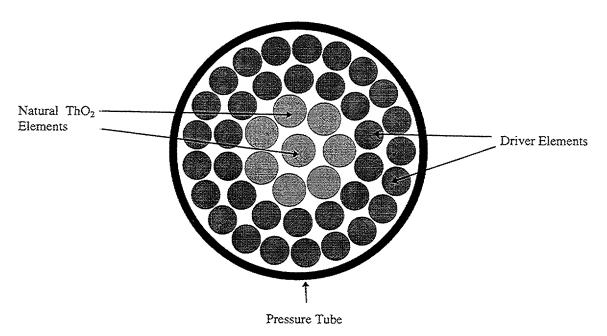


Figure 2: Configuration of Fuel Elements in a CANFLEX Bundle

## 4. Lattice Properties of Mixed-Thorium Fuel

Table 1 gives the initial fuel composition of the three types of thorium fuel bundles used in the current study. The basic physics properties of these fuel lattices, such as the variation of lattice k-infinity, of fissile content and of lattice coolant-void reactivity, are shown in Figures 3, 4 and 5, respectively as functions of bundle-average fuel burnup. Although natural- $UO_2$  and natural- $ThO_2$  fuel bundles are not used in this study, their physics properties are also shown in these figures for comparison purposes.

The initial fissile content of the high-burnup thorium bundles has been carefully chosen so that the depletion rate of the fissile material is almost the same as the conversion rate of the fertile <sup>232</sup>Th into fissile <sup>233</sup>U. Consequently, the reactivity and the fissile content of the high-burnup thorium bundles are almost constant throughout the entire lifetime. These high-burnup thorium bundles can theoretically reside indefinitely in the reactor. The attainable fuel burnup is limited only by the mechanical integrity of the fuel bundle.

The main purpose of the gadolinium is to shape the axial flux distributions so that the resulting bundle flux and power distributions are similar to those in the thorium-burning reactor without adjuster rods. Figure 6 shows that the gadolinium effectively eliminates the bundle power distortion caused by the adjuster rods. As expected, the effect of gadolinium on lattice reactivity is evident only during the initial stage of the fuel lifetime. The fast burnout rate of gadolinium suppresses the reactivity of the fresh bundle without causing significant burnup penalty over the lifetime of the fuel. This effect also reduces the channel and bundle power ripples caused by refuelling. The presence of a neutronic poison, gadolinium, in the central element also reduces coolant-void reactivity<sup>4</sup>. This results in a significant reduction in the core-averaged coolant-void reactivity.

	1.8% SEU-Th No Gd	1.8% SEU-Th 6% Gd (central pin)	1.7% SEU-Th No Gd
	Whole core, Option 1	0% Ou (central phi)	No Ga
	Outer Core, Option 2	Inner Core, Option 2	Periphery Channel Option 2
Ring 1			
<sup>235</sup> U	0	0	0
<sup>238</sup> U	0	0	0
<sup>232</sup> Th	0.510	0.510	0.510
Gd(natural)	0	0.0306	0
Ring 2	•		
<sup>235</sup> U	0	0	0
<sup>238</sup> U	0	0	0
<sup>232</sup> Th	3.590	3.590	3.590
Ring 3			
<sup>235</sup> U	0.102	0.102	0.087
<sup>238</sup> U	5.578	5.578	0.349
<sup>232</sup> Th	0	0	4.700
Ring 4			
<sup>235</sup> U	0.153	0.153	0.131
<sup>238</sup> U	8.367	8.367	0.523
<sup>232</sup> Th	0	0	7.050
Total			
<sup>235</sup> U	0.256	0.256	0.218
$^{238}U$	13.944	13.944	0.872
<sup>232</sup> Th	4.100	4.100	15.850
Gd(natural)	0	0.0306	0

## Table 1: Initial Fuel Composition (kg/bundle) of Mixed SEU-Thorium Bundles

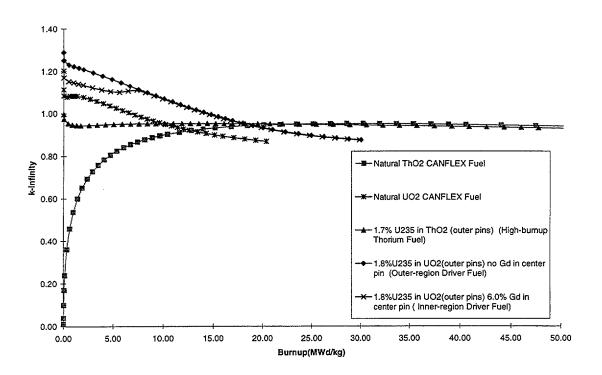


Figure 3: Lattice k-infinity vs. Fuel Burnup

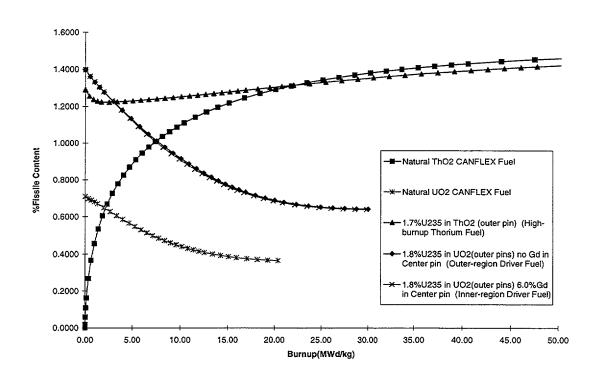


Figure 4 : Fissile Content vs. Fuel Burnup

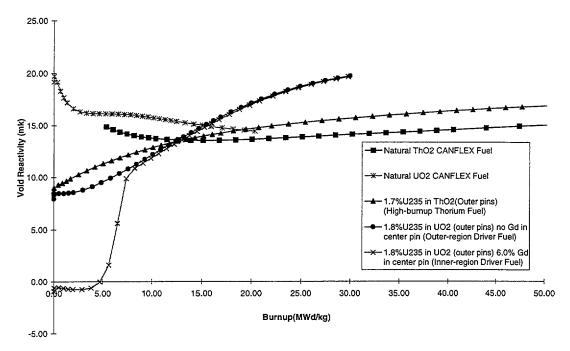


Figure 5: Lattice Void Reactivity vs. Fuel Burnup

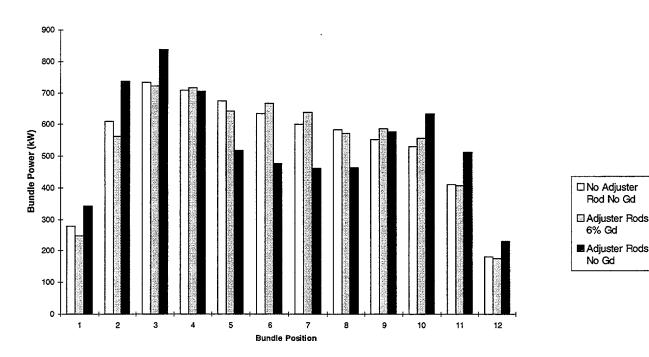


Figure 6: Effect of Adjuster Rods and Gadolinium on Time-Average Bundle Power Distribution

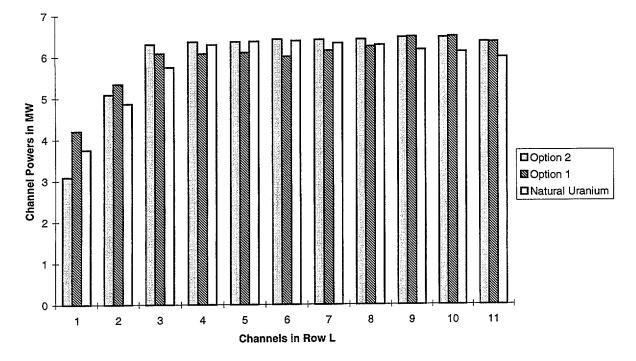


Figure 7: Time-Average Channel Power Distributions

	Option 1 (without Adjuster Rods)	Option 2 (with Adjuster Pode)
ΥΥ		(with Adjuster Rods)
Uranium Utilization	130	138
(MgNU/GW(e).a)	05	24
Percentage of Thorium in Reactor Core (by volume)	25	36
Core-Average Fuel Burnup	22.1	20.3
(MW.d/kgHE)		
Core-Average Thorium Burnup	10.4	9.1
(MW.d/kgHE)		
Fuelling rate	5.5	6.0
(Bundles per Full-Power-Day)		
Fuelling Scheme	2	2
(bundle-shift)		
Maximum Channel Power (kW)	6491	6505
(Time-Average)		
Maximum Bundle Power (kW)	741	739
(Time-Average)		
Maximum Channel Power (kW)	6855	6849
(Instantaneous)		
Maximum Bundle Power (kW)	785	781
(Instantaneous)		
Reactor Leakage (mk)	30.6	25.8
Full Core Coolant-Void Reactivity (mk)	12.6	10.6

Table 2 : Summary of WIMS/RFSP OTT Fuel	I-Management Studies
in a CANDU 6 Reactor	

## 5. Characteristics of Thorium-Burning CANDU Reactors

The RFSP code was used to perform time-average core calculations for Options 1 and 2 using a uniform 2-bundle-shift fuelling scheme. Instantaneous core calculations were conducted using randomly generated age patterns. Figure 7 shows that the channel power distributions for the two thorium-burning reactors are very similar to those of a typical natural-uranium CANDU reactor. Figure 8 shows that the axial bundle power distributions in the thorium-burning reactors are flatter than those in a natural-uranium CANDU and are more skewed towards the coolant-inlet end. This skewed axial power profile should improve the thermalhydraulics performance. Major reactor physics results for both options are summarized in Table 2.

The fuelling rates, maximum channel power and maximum bundle power for both options are well within the limits established for current CANDU reactors using natural-uranium fuel. There is also a significant reduction in the coolant-void reactivity from that of a natural-uranium reactor under comparable conditions. Option 1 gives 21% better uranium utilization than that for a natural-uranium CANDU reactor. About half of the improved fuel efficiency is due to the removal of the adjuster rods. The other half can be attributed to the energy produced in the thorium fuel. Option 2, which uses the existing adjuster rods, gives 14% better uranium utilization than that for a natural-uranium reactor with the additional advantage of a significantly lower coolant-void reactivity.

## 6. Effect of Flux-Dependence on Thorium Physics Calculations

The dependence of the lattice parameters of thorium-bearing fuel on the flux and power history of the fuel was reported in a previous study <sup>5</sup>. This dependence arises because the creation of the fissile isotope <sup>233</sup>U from the fertile isotope <sup>232</sup>Th is flux-dependent. This process is analogous to the production of the fissile isotope <sup>239</sup>Pu from the fertile isotope <sup>238</sup>U in uranium-based fuel cycles. The major difference is that the equilibrium level of <sup>233</sup>U in <sup>232</sup>Th is about 1.5%, and it is sensitive to flux level, whereas the equilibrium level of <sup>239</sup>Pu in <sup>238</sup>U is only about 0.4%, and it is relatively insensitive to the flux level.

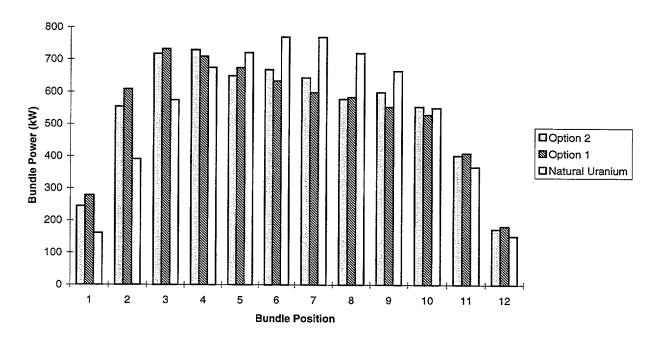


Figure 8: Time-Average Bundle Power Distributions in a High-Power Channel

The lattice parameters for the mixed SEU-Thorium fuels were calculated with WIMS-AECL using a constant cell-average thermal flux of  $2.0 \times 10^{14}$  n/cm<sup>2</sup>.s. Those for the high-burnup thorium fuel were calculated using a constant cell-average thermal flux of  $1.0 \times 10^{14}$  n/cm<sup>2</sup>.s. These flux levels are consistent with the flux levels obtained from RFSP core calculations. The results of the RFSP calculations are not expected to be significantly affected by the flux-dependence of the lattice parameters because the thorium-bearing fuel is placed in very low-importance regions. The high-burnup thorium fuel is located at the outermost channels and the thorium in the mixed SEU-Thorium fuel bundles is limited to the inner two fuel rings, where the thermal neutron flux level is relatively low.

WIMS-AECL calculations were performed to assess the effect of flux-dependence on the lattice k-infinity and fissile content of the mixed SEU-Thorium fuel up to a core-average fuel burnup of 22.1 MW.d/kgHE for Option 1 and 20.3 MW.d/kgHE for Option 2. Two methods for incrementing the fuel burnup were used:

- the fuel burnup was calculated by simulating the shifting of a thorium fuel bundle from the inlet to the outlet using a typical time-average axial flux profile and resident time at each position calculated by RFSP, and
- the fuel burnup was calculated by using a constant cell-average thermal flux level consistent with the time-average axial flux profile.

The results are summarized in Table 3. As expected, there are small differences between these two sets of calculations. However, the variations in the <sup>233</sup>U content are not significantly greater than the variations in the fissile plutonium content. The sum of <sup>233</sup>U and <sup>233</sup>Pa is the important quantity at discharge and is generally quite constant for each case type. For the cases with the axial thermal neutron flux distribution modelled, the flux level drops appreciably for the final irradiation period. During this irradiation, the decay of <sup>233</sup>Pa to <sup>233</sup>U is enhanced and the level of <sup>233</sup>U at discharge is greater than in the constant flux situation. This increases the k-infinity somewhat and also reduces the amounts of the other fissile nuclides that were used up. The maximum discrepancy in the lattice k-infinity is less than 10 mk at discharge. The uncertainty in the mixed SEU-Thorium fuel lattice properties is expected to be small and should not have a significant impact on the results of the reactor core calculations.

	WIMS calculations based on Core- Average Constant Cell Flux	WIMS calculations based on RFSP Time-Average Axial Flux Profile
Option 1		
Core-Average Burnup	22.1	22.1
(MW.d/kgHE)		
WIMS k-infinity	0.898	0.904
<sup>235</sup> U	20.6	20.5
<sup>233</sup> U	48.9	50.7
<sup>233</sup> Pa	4.6	3.1
$^{239}$ Pu + $^{241}$ Pu	46.8	47.1
Option 2		
(outer-region fuel)		
Core-Average Burnup	20.3	20.3
(MW.d/kgHE)		
WIMS k-infinity	0.916	0.922
<sup>235</sup> U	26.6	26.6
<sup>233</sup> U	47.0	48.6
<sup>233</sup> Pa	4.2	2.8
$^{239}$ Pu + $^{241}$ Pu	46.6	46.8

# Table 3 : Effect of Flux Dependency on Lattice Reactivity and Fissile Contents (g/bundle) in Mixed SEU-Thorium Fuel Bundle

The RFSP code was used to conduct time-dependent refuelling simulations in the Option 1 core for a period of 100 full-power-days using lattice parameters that are consistent with the power history of individual fuel bundles. These local lattice parameters are based on WIMS-AECL calculations using a very efficient computational scheme<sup>6</sup>. The maximum channel power and maximum bundle power during this simulation period using history-dependent lattice parameters are  $6.8 \pm 0.1$  MW and  $780 \pm 10$  kW respectively. These results are very similar to those calculated by the RFSP code using traditional non-history-based lattice parameters.

## 7. Conclusions

The current study represents only a first look at practical fuel-management strategies for the OTT fuel cycle. Two options for implementing the OTT fuel cycle in existing CANDU reactors were identified. For both options, the uranium utilization is better than that of the natural-uranium fuel cycle. The reactor and the fuel perform within existing envelopes without requiring major modification to the current reactor design. Coolant-void reactivity is significantly lower than that of a natural-uranium reactor under comparable conditions.

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## A THORIUM BREEDER REACTOR CONCEPT FOR OPTIMAL ENERGY EXTRACTION FROM URANIUM AND THORIUM

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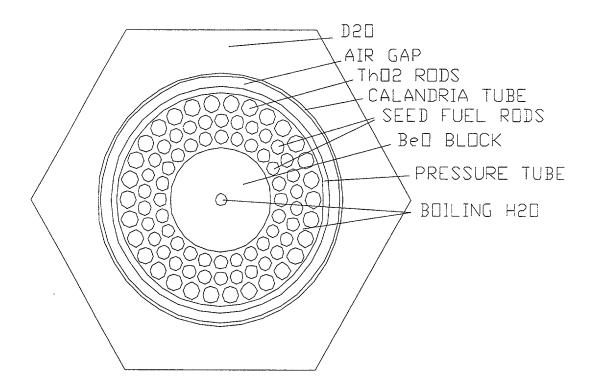
## Abstract

An attractive thorium breeder reactor concept has been evolved from simple physics based guidelines for induction of thorium in a major way in an otherwise enriched uranium reactor [1]. D<sub>2</sub>O moderator helps to maximise reactivity for a given enrichment. A relatively higher flux level compared to LWRs offers the advantage of higher rate of <sup>233</sup>U production in thoria rods. Thus fresh thoria clusters consider no feed enrichment. In an equilibrium core, a full batch of pure thoria clusters are loaded during each fuel cycle. They undergo irradiation for about one year duration. By this time they accumulate nearly 70% of the asymptotic stable concentration of <sup>233</sup>U, if they face a flux level of the order of 10<sup>14</sup> n/cm<sup>2</sup>/sec. In the next fuel cycle, these thoria rods in ring cluster form are juxtaposed with the fresh enriched fuel rods, also in ring cluster form. Such integrated fuel assemblies are then irradiated for four or five fuel cycles, at the end of which U as well as Th rods attain a reasonably high burnup of about 30-32 MWD/kg. The core characteristics are quite attractive. The core excess reactivity remains low due to large thoria inventory which makes the net burnup reactivity load to be below 1%. The core is capable of being operated in an annual batch mode of operation like a LWR. The control requirement during power operation is negligible. Xenon over-ride requirement is low and can be managed by partial withdrawal of a few thoria clusters. Void reactivity is nearly zero or negative by the optimum design of the fuel cluster. Reactivity changes due to temperatures of fuel, coolant and moderator are also small.

## **1. INTRODUCTION**

For generation of fission nuclear power both uranium and thorium have comparable gross energy potential. The present day nuclear reactors use mainly uranium because one needs some external feed enrichment for thorium, while it is intrinsically present in uranium. This paper explores the possibility of avoiding such feed enrichment. It is conjectured that if thoria rods are placed like control clusters in a thermal reactor using enriched fuel, and are allowed to face a fairly large flux of the order of 10<sup>14</sup> n/cm<sup>2</sup>/sec, they would accumulate significant <sup>233</sup>U in one fuel cycle. Subsequently they can be used as fuel rods along with normal fresh enriched fuel rods and irradiated for four or five more fuel cycles. The physics based guidelines to evolve the new reactor concept are enumerated in Ref.1. They can be briefly summarised as follows.

- $D_2O$  moderator is necessary to enhance the reactivity as well as the flux level incident on thoria rods.
- A fairly large sized fuel cluster with lattice pitch less than the optimum ensures that the coolant void coefficient is near zero or negative. The moderation role is shared by inchannel hot coolant (boiling  $H_2O$ ) and the outchannel cooler moderator  $D_2O$ .



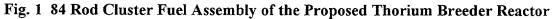


Table-1

**Core and Fuel Design Parameters** 

Reactor Power		1875 Mwt (6	O Mwe)	
	nnes/hr)	27 X 10 <sup>6</sup>	<u></u>	
	(w/cm)	160		
Height of the core	360			
No. of rods in a fuel cluster	84 (54 seed rods + 30 ThO <sub>2</sub> fertile rods)			
No. of rods in pure thoria cluster	· · · · · · · · · · · · · · · · · · ·	30 ThO <sub>2</sub>		
No. of fuel clusters in the core	360			
No. of pure thoria cluster in the core	72 (varied from 72 to 90)			
$Ring \rightarrow$	Inner	Middle	Outer	
No. of rods		24 (seed)	30 (seed)	30 (fertile)
Enrichment		see Table-2	see Table-2	insitu
Pitch circle dia	(cm)	10.4	13.0	15.8
Clad ID/OD	(cm)	1.0/1.14*	1.0/1.14*	1.26/1.40
Clad Material		Zr-Nb(1%)	Zr-Nb(1%)	Zr-Nb(1%)
Assembly Lattice Pitch (hexagonal)	(cm)	30 (32 cm als	o used for 'eue	eut' fuel)
Average Fuel Temperature	600°C			
Average Coolant Temp. (Boiling H <sub>2</sub> O - 101)	286°C			
Central Moderator Block		BeO		
ID/OD	(cm)	1.0/9.0 (inclu	sive of Zr-line	r)
Pressure Tube (PT) Zr-Nb (2.5%) ID/OD	(cm)	17.6/18.70		
Calandria Tube (CT) Zr-2 ID/OD	(cm)	20.40/20.70		
Moderator Material/Temperature		D <sub>2</sub> O - 80°C		
Additional locations for Shutoff Rods		7		
Radial D <sub>2</sub> O reflector thickness	(cm)	60 to 70		
Axial D <sub>2</sub> O reflector thickness	(cm)	60		
Calandria Tank Size		~8 m dia X 4.	8 m height	

For seed type 'tptpt' thinner fuel dia was considered.

Fuel Type	eueut	npnpt	nunut	tptpt	tutut
Seed zone	enriched	$PuO_2$ in nat.	<sup>233</sup> U in	PuO <sub>2</sub> in	<sup>233</sup> U in
	UO <sub>2</sub>	ŪO <sub>2</sub>	nat.UO2	ThO <sub>2</sub>	$ThO_2$
Fuel Pellet Dia (mm)	10	10	10	9	10
Seed content	4.5% <sup>235</sup> U	9% Pu	3.3% (92%	10% Pu	4.9%
in inner ring		(74% fissile)	<sup>233</sup> U)	(74% fissile)	(92% <sup>233</sup> U)
Seed content	3.6% <sup>235</sup> U	5% Pu	2.6% (92%	6% Pu	3.9%
in middle ring		(74% fissile)	<sup>233</sup> U)	(74% fissile)	(92% <sup>233</sup> U)

Table-2Description of Seed Zones

The reactor is akin to that of a SGHWR (Steam Genearting Heavy Water Reactor). Fig.1 gives the schematic diagram of the proposed reactor fuel cluster. Table-1 gives the description of the core and fuel design parameters. The fuel can be briefly described as follows: There are three rings of fuel rods in a fuel cluster. Inner 24 rods and middle 30 rods constitute the seed zone. The outermost ring of 30 thoria rods is the fertile zone. As mentioned earlier, these fertile rods undergo prior irradiation for one fuel cycle duration and then only they are integrated with the fresh seed rods. BeO block with Zr-liner is used as filler material within each fuel cluster. This helps to achieve a low value of power peak within the fuel cluster. Pure thoria clusters contain only 30 ThO<sub>2</sub> rods and hence will need bigger BeO blocks as filler material. It is necessary to minimise the water volume to a bare minimum value in the core as well as in axial reflector zones in order to gain reactivity and also to reduce axial power peak.

In this paper, the potential of the proposed reactor concept for long term fuel cycle strategies involving a variety of seed zones is also briefly explored. Reprocessed  $^{233}$ U or Pu as feed enrichment in either natural UO<sub>2</sub> or ThO<sub>2</sub> are conceived as the seed zones in addition to  $^{235}$ U enriched UO<sub>2</sub> fuel. These 5 types of seed zones are described in Table-2. The outer ring of 30 ThO<sub>2</sub> fuel rods is common to all types and is vital for realisation of a possible thermal breeder.

The lattice calculations were performed with the CLUB module [2] of the PHANTOM code system [3]. 69 group WIMS cross section library was used. The core is assumed to have a hexagonal lattice structure.

For core calculations, a new core followup code TRISUL (Thorium Reactor Investigations with Small Uranium Loading), has been developed. This code is an extension of the TRIHEX-3D code [4] which uses few group diffusion theory and finite difference method with hexagonal or triangular meshes. TRISUL is a coupled neutronics cum thermal hydraulics code. Approximate thermal hydraulics calculations are done with the models which are used for Tarapur BWR square fuel assembly geometry.

## 2. RESULTS AND DISCUSSIONS

Preliminary investigations were done for all the five types of seed zones with TRIHEX-3D code. In order to compare the overall core characteristics, a flat zone-wise burnup model, typical average void value and a mean flux level for thoria clusters were assumed. The last parameter is crucial because the reactivity characteristics of the core are strongly influenced by the flux level prevalent in thoria assemblies and the integrated fluence achievable in their first fuel cycle of irradiation.

The observations from the above coarse studies are : The burnup reactivity swing is small (<1%) in all cases. The reactivity load of 72 pure thoria clusters is found to be ~9% at BOC and about ~5% at EOC for seeds 'eueut', 'nunut' and 'tutut'. In case of seed materials 'npnpt' or 'tptpt' the worth of thoria clusters is lower by 1%. A discharge burnup of 35,000 MWD/T is possibly achievable in all cases.

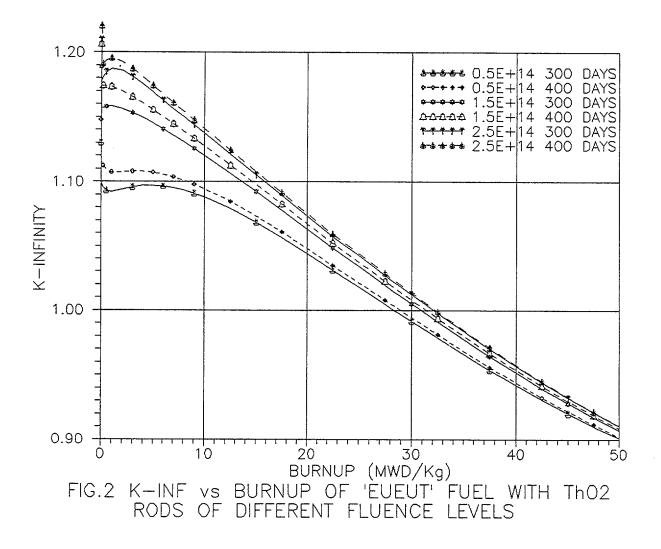


 Table-3
 Results of Core Follow-up Study with TRISUL for Core with 'eueut' Seed

 78
 Fuel Assemblies per Batch - <sup>235</sup>U enrichment - 4.7% and 3.8%

Full Power Days	Core Average Burnup	Core Average Void	K <sub>eff</sub>	Reactivity mk	Peaking factor
	GWD/T	Fraction			
0 (BOC)	10.912	0.285	0.996379	-3.63	2.184
50	11.743	0.285	0.993944	-6.09	2.273
100	12.580	0.281	0.993484	-6.56	2.292
150	13.427	0.276	0.993224	-6.82	2.294
200	14.285	0.279	0.992936	-7.11	2.252
250	15.153	0.282	0.992282	-7.78	2.193
300 (EOC)	16.031	0.282	0.991090	-8.99	2.133

At the time writing this paper, detailed calculations were performed with TRISUL code for only one of the seed zones, viz., 'eueut'. For this fuel, a complete two group database was generated as a function of burnup at four void fractions, three flux levels (0.5, 1.5 and 2.5 X  $10^{14}$  n/cm<sup>2</sup>/sec), and irradiation duration of 300 or 400 days. Pitch was increased to 32 cm. Fig.2 shows the reactivity variation with burnup for 40% void and different fluence levels of thoria rods. It is seen that one can gain reactivity either by having higher flux level in thoria rods or by having longer time of irradiation.

Table-4 Comparison of Fuel Requirement for 10 Gwe and 30 years Core Lifetime (Approximate Estimates for Relative Comparison)						
Parameter		eueut	npnpt	nunut	tptpt	tutut
Discharge Burnup	(MWD/T)	35000	35000	35000	35000	35000
Fissile feed per year	(kg)	388	727	349	689	378
Specific power	(kg/Mwe)	0.65	1.21	0.58	1.15	0.63
Fuel per 10Gwe, 30year	s (T)	4845 U	4845 U	4845 U		
300 efpd per year		3835 Th	3835 Th	3835 Th	7356 Th	8180 Th
Gross seed input	(T)	194	363	175	344	189
Gross Output Pu	(T)	39	198	39	117	
U-233	(T)	58	58	78	95	147
(U-234, U-235 & U-236	(T)	76	26	46	14	47
Gross Output	(T)	173	282	154	226	194

\* Seed includes <sup>235</sup>U or (<sup>233</sup>U, <sup>234</sup>U and U<sup>235</sup>U) or (<sup>235</sup>U and Pu isotopes), as applicable.

For the core analysis, several batch sizes were tried. Sample results are presented for the case of 78 fuel assemblies per batch. Table-3 gives the  $K_{eff}$ , overall power peak variation with burnup for an equilibrium core. It must be mentioned that that in this study all the thoria clusters were kept IN throughout the fuel cycle and no external reactivity control change was done, i.e. there was neither soluble boron adjustment nor any control device movement. It is seen that the fall in  $K_{eff}$  is only 0.54%, though the  $K_{eff}$  value is less than unity throughout the fuel cycle. Since the thoria clusters were kept IN all the time the small reactivity requirement of about 1% can be easily met by partial withdrawal of the thoria clusters. The latter can also be used for reducing the power peaking factors. Care must be taken to see that the integrated fluence in the thoria clusters remain adequate for adequate accumulation of <sup>233</sup>U in them to provide adequate reactivity later.

The average absolute flux level in thoria clusters was seen to be only about  $1\times10^{14}$  n/cm<sup>2</sup>/sec, though the flux level in surrounding zones is nearly double this value. This is due to the fact that fast flux drops by a factor of three in thoria cluster cells, while thermal flux is of similar magnitude in comparison to the neighboring cells. Thus it is necessary to enhance the fast or epithermal flux in thoria clusters. A small initial fissile content of 0.3% U<sup>233</sup> in all thoria rods, or better, 1.5-2% <sup>233</sup>U in just 6 of the 30 rods in pure thoria cluster, or an inner nat. UO<sub>2</sub> rods fuel ring cluster, can help to mitigate the (fast) flux depression in thoria clusters and thereby ensure accumulation of adequate <sup>233</sup>U in one year irradiation. Use of Be rods around thoria clusters can also augment the fast flux. The overall engineering feasibility has to be assessed. Pressurized D<sub>2</sub>O coolant can also result in higher flux level. Since burnup reactivity swing is small, there would be least movement of control and hence a deliberate higher power rating can also help in achieving higher flux level in thoria rods.

We compare in Table-4 the estimates of fuel requirement of the proposed reactor concept for installed capacity of 10 Gwe and a life-time of 30 years and 300 effective full power days (efpds) of operation per year. From Table-4 it is seen that use of  $^{233}$ U is superior to  $^{235}$ U as seed material as expected. This advantage will be slightly offset when  $^{234}$ U content increases after multi-recycling. When  $^{233}$ U is used along with natural UO<sub>2</sub> (nunut) one requires the minimum gross fissile input. When used with ThO<sub>2</sub> (tutut), slightly more fissile input is required, but the U<sup>233</sup> output is the largest in this case. If additional breeding in blanket type zone is also exploited, this option can become a net breeder of fissile material. When Pu is used as seed material, the gross seed input is nearly doubled. A large fraction of this material is discharged as unburnt Pu. This is due to large <sup>240</sup>Pu content in Pu and high capture to fission ratio of Pu isotopes. The fissile content in Pu output is 70% while in U output it is above 90% (<sup>233</sup>U).

## **3. UNCERTAINTIES IN THE CALCULATIONS**

The calculations presented in this paper are of indicative nature only. There is bound to be uncertainty in the calculations due to the basic nuclear data available with the author(s). Calculational tool like TRISUL and the burnup model for studying a dynamic mixture of fresh seed and irradiated fertile zones is novel and needs testing. Nonetheless the physics based ideas expressed here point to a definite feasibility of the proposed concept with some minor revision of the design parameters.

## 4. CONCLUSION

The proposed thorium breeder reactor opens up the possibility of inducting thorium into an otherwise enriched uranium reactor. The overall core characteristics with respect to safety, operational ease and economy are seen to be attractive. The seed material, 'tutut', is quite attractive to enable not only self-sustaining mode of Th-<sup>233</sup>U fuel cycle, but a steady growth would be feasible, if its breeding potential is properly exploited. However a proper mix of 'nunut', 'npnpt' and 'tutut' combinations should be worked out to exploit the energy potential uniformly from uranium and thorium. The overall Pu production and its accumulation can be minimised by using the 'npnpt' and 'tutut' seed types in the ratio in which these seed materials are regenerated.

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## MOX FUEL

## (Session 4)

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# AN OVERVIEW OF ECONOMIC AND TECHNICAL ISSUES RELATED TO LWR MOX FUEL USAGE



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## Abstract

This paper will present comparisons of the economics of MOX versus  $UO_2$  fuels. In addition to the economics of the front end, the scope of the comparison will include the back end of the fuel cycle. Management of spent MOX fuel assemblies presents utilities with some technical issues that can complicate spent fuel pool operation. Alternative spent fuel management methods, such as dry storage of spent MOX fuel assemblies, will also be discussed.

Differences in decay heat loads versus time for spent MOX and  $UO_2$  fuel assemblies will be presented. This difference is one of the main problems confronting spent fuel managers relative to MOX. The difference in decay heat loads will serve as the basis for a performance overview of the various spent fuel technologies available today.

The economics of the front end of MOX will be presented relative to  $UO_2$  fuel. Availability of MOX manufacturing capability will also be discussed, along with a discussion of its impact on future MOX fabrication prices.

The in-core performance of MOX will be compared to that of  $UO_2$  fuel with similar performance characteristics. The information will include highlights of nuclear design and related operational considerations such as:

- Reactivity reduction with burnup is slower for MOX fuel than for  $UO_2$  fuel;
- Spectral hardening resulting in lower control rod worths and a lower soluble boron worth; and
- More negative moderator, void and fuel temperature coefficients.

A comparison of Westinghouse and ABB-CE core designs for use on disposition of weapons MOX in 12- and 18-month cycles will be presented.

## 1. INTRODUCTION

Today there is increasing interest in use of MOX fuels as a component of a weapons plutonium disposition program. Though there are currently no reprocessing or MOX fabrication facilities in the United States, many utilities have indicated a willingness to participate in the MOX disposition program. Naturally, utilities expect economic benefits as a result of MOX fuel use. This is a commercial matter to be negotiated between the participating utilities and the government.

While economics is the driving force in the utility decision-making process, the technical aspects of MOX use, some of which will have economic consequences, cannot be ignored. Modifications to plant safety systems are among the more significant cost areas that must be investigated in order to obtain a complete picture of the true cost related to MOX utilization. Such modifications include, but are not limited to, use of enriched boron in the primary coolant, installing larger tanks for the boron reservoir, use of enriched hybrid  $B_4C$  control rods or additional standard control rods.

All technical information contained herein was obtained from reviews of nonproprietary published literature and prior discussions with vendors and utilities involved in MOX projects (done for other Stoller MOX studies).

## 2. TECHNICAL OVERVIEW

This section will focus on Pressurized Water Reactor (PWR) fuels. There are also issues related to fuels for Boiling Water Reactors (BWR); however, focusing on the PWR fuels will cover most of the issues. In addition, the main differences between recycle-derived plutonium and weapons-grade plutonium will be addressed.

## 2.1 INTRODUCTION

The general specifications used by fuel vendors for recycled MOX fuel and core designs are as follows:

- 1. MOX assemblies should be designed to minimize or eliminate local power peaking mismatches with co-resident and adjacently loaded UO<sub>2</sub> assemblies. Power peaking at the interfaces arises from different neutronic behavior between UO<sub>2</sub> and MOX assemblies.
- 2. A MOX core (MOX and UO<sub>2</sub> or all-MOX assemblies) should provide cycle energy equivalent to that of an all-UO<sub>2</sub> core.
- 3. The reactivity coefficients, kinetics data, power peaking and the worth of shutdown systems with MOX fuel and cores must be such to meet the design criteria and fulfill requirements for safe reactor operation.

While the first and third items are equally applicable to weapons-grade MOX fuel and core designs, the second item may not necessarily apply in these cases. The main objective when burning weapons-grade plutonium is rapid disposition as opposed to economics. Economics is a key element in the design process for recycled MOX assemblies and cores. Studies in the United States indicate that acceptable weapons-grade fuel/core designs can be achieved for up to 18-month fuel cycles with discharge burnups of up to 45GWD/MTHM.

Table 1 presents typical isotopic compositions of recycle and weapons-grade plutonium. The composition differences are sufficient to alter the operational behavior and safety analysis, primarily as related to temperature and void coefficients, as well as on-site handling.

# TABLE 1: TYPICAL ISOTOPIC COMPOSITIONS OF RECYCLE AND WEAPONS-GRADE PLUTONIUM

		IS	SOTOPE, WT.	%	
	<sup>238</sup> Pu	<sup>239</sup> Pu	<sup>240</sup> Pu	<sup>241</sup> Pu	<sup>242</sup> Pu
Recycle Pu ("Reactor Grade")	1	59	24	11	5
Weapons Grade	< 0.1	93	6	1	< 0.1

		COMMENT	up: e	On one-to-one basis l shim weapons-grade Pu sely core would behave more like UO <sub>2</sub> core sty, than recycle Pu core regarding affected items	ative: On one-to-one basis te coefficients ng somewhat more negative than tion equivalent recycled MOX core if hot-weapons-grade
IMPACT		NEGATIVE	At higher burnup: More fission gas release More clad corrosion	Lower, less efficient fixed shim Lower, adversely affects cold shutdown safety, post LOCA behavior	<ul> <li>1,3.More negative:</li> <li>Exacerbate over-cooling events, or pressurization transients</li> <li>Increases hot-</li> </ul>
IMI		POSITIVE	Higher local peaking at low burnup with no impact on margins	Lower, less oscillation hazard	<ol> <li>More negative:         <ul> <li>Improved cycle stretch</li> <li>More negative - better response to rod drop events</li> </ul> </li> </ol>
EFFECT OF MOX	WEAPONS-	<b>GRADE PU</b>	Same	Less of an impact due to the lower plutonium fissile loadings to compensate for <sup>240</sup> Pu neutron absorption	Some evidence of somewhat greater impact on coefficients
EFFEC		RECYCLE PU	<ul> <li>Flatter depletion:</li> <li>Less power fresh MOX, low burnup</li> <li>More power high burnup</li> </ul>	Higher cross- sections of plutonium give lower thermal flux harder spectrum and less efficient thermal region	More numerous resonances in epithermal range and fission/capture cross-section resonances lower than at thermal
	ITEM	AFFECTED	Power sharing MOX/UO <sub>2</sub>	<ol> <li>Xenon poisoning</li> <li>Control rod worth</li> <li>Soluble boron</li> <li>Assembly</li> <li>design</li> </ol>	<ol> <li>Moderator temperature coefficient</li> <li>Doppler coefficient</li> <li>Void coefficient</li> </ol>
		PARAMETER	Reactivity behavior with burnup	Absorption cross- section at thermal energies	Epithermal absorption resonances

TABLE 2 IMPACT OF MOX VERSUS UO<sub>2</sub> ON PARAMETERS

TABLE 2 (Continued) IMPACT OF MOX VERSUS UO2 ON PARAMETERS

		EFFECT	EFFECT OF MOX	VI	IMPACT	
	ITEM		WEAPONS-			
PARAMETER	AFFECTED	<b>RECYCLE PU</b>	<b>GRADE PU</b>	POSITIVE	NEGATIVE	COMMENT
Delayed neutron	Transient Power	Smaller	Same		Faster power/flux	Withdrawal event
Fraction	Increases				rises in	rise eased by lower
					uncontrolled rod	rod worth, more
					withdrawal,	negative Doppler
					overcooling events	
Decay of <sup>241</sup> Pu	Fuel reactivity	Important due to	Nil		Reactivity loss (less	
	(at longer than	relatively high Pu <sup>241</sup>			cycle energy) due to	
	anticipated times	content			decay of fissionable	
	between				Pu <sup>241</sup> for recycle Pu	
	manufacturing and				cases	
	operation)					
<b>Pressure vessel</b>	Vessel damage rate	Higher fast flux at	Same		Accelerated vessel	
fluence		low-moderate			damage if MOX	
		burnups			assemblies are	
					placed in critical	
					peripheral locations	
Batch isotopic	Manufacturing-	Relatively large	Little or no		Increased	
variability	related uncertainties	variability batch to	variability batch to		engineering	
		batch	batch		uncertainties built	
					into MOX fuel/core	
		-			for recycle Pu cases	

## TABLE 3 REACTOR SAFETY EVALUATION COMPARISON OF MOX AND UO2 IN PWRs

EVENT	SOURCE	EFFECT OF MOX IN CORES
Overcooling Transients (Breaks in Secondary Side) 1. Category 2: secondary valve opening	EdF	<ul> <li>Margins to recriticality lower</li> <li>No recriticality for 30% MOX core</li> <li>Major impact with MOX is lower boron worth</li> </ul>
<ol> <li>Category 4: double-ended main steam line break (consequences may be less severe with weapons-grade Pu due to more negative Doppler coefficient)</li> </ol>	Siemens	<ul> <li>Prompt critical in 30% MOX core (less favorable kinetics)</li> <li>More severe power rise (peak &lt;10 % rated power)</li> <li>DNBR below dryout conditions</li> <li>No system changes required</li> <li>Recriticality does not occur for envelope of MOX cores</li> <li>Margin less than for equivalent UO<sub>2</sub> cores</li> <li>No system changes required</li> <li>Problems with 16 x, 100% MOX, and 18x, 50% MOX <ul> <li>Recriticality can occur</li> <li>Larger tanks of enriched boron may be needed (avoid dryout)</li> </ul> </li> </ul>
Rod Ejection Accident (consequences may be less severe with weapons-grade Pu and higher fuel burnups)	EdF CABRI Test	<ul> <li>Margin to limits lower with 30% MOX core         <ul> <li>less favorable kinetics overshadows lower rod worth</li> </ul> </li> <li>Event terminated within limits (only modest fuel burnup)</li> <li>MOX rodlet at ~ 100 cal/gram failed violently at 50-55 GWD/MTHM</li> </ul>
LOCA (breaks in primary system)	EdF	<ul> <li>Minor changes for 30% MOX core case:         <ul> <li>initial boron reserve increased from 2000 to 2400 ppm</li> <li>minimum concentration in sump water raised from 1300-1500 ppm</li> <li>instruments and control of steam generator atmospheric relief system duplicated and emergency supplied</li> </ul> </li> </ul>
	Siemens	<ul> <li>German LOCA requirement &lt;10% rods fail in LOCA</li> <li>For MOX, failure threshold 10-20 watt/cm lower than UO<sub>2</sub> due to higher fission gas release (at modest fuel burnups)</li> </ul>

## 2.2 OPERATIONAL IMPACT OF MOX VERSUS UO2

Table 2 shows the operational impacts of MOX versus  $UO_2$ , as related to various neutronic parameters. Both recycle and weapons-grade plutonium are considered, and positive and negative impacts are given. Reactivity behavior with burnup as it adversely affects fission gas release, clad corrosion at high exposures, the high-thermal absorption cross sections that reduce shim worth, and the more negative moderator/void coefficients that adversely affect safety are particular concerns with MOX fuels.

COMMENTS	MOX should display higher corrosion at higher	burnups Higher MOX pellet creep during transients MOX rods release more power at higher burnups operate at 150 - 200 watts/centimeter in third	(last) annual cycle of irradiation in France Limited French data - only one leaking MOX rod to date
EFFECT OF MOX	No significant difference No significant difference	Better Higher - Accelerated release starts at $\sim 40$ GWD/MTHM (French and German data)	About same as $\mathrm{UO}_2$ (1.5 leakers per cycle)
ITEM	<ol> <li>Dimensional Behavior</li> <li>Corrosion Behavior</li> </ol>	<ul><li>3 Ramping Resistance to Failure (PCI Effect)</li><li>4 Fission Gas Release</li></ul>	5. Fuel Reliability

TABLE 4 FUEL PERFORMANCE ISSUES (MOX VERSUS UO<sub>2</sub>)

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## 2.3 SAFETY EVALUATIONS OF MOX VERSUS UO2

Table 3 contrasts MOX versus  $UO_2$  with respect to safety evaluations. Within the framework of current operations with MOX fuel in Europe, the consequences of limiting transients, while somewhat more severe with recycled MOX, are still acceptable with no system changes required. Rod ejection has adequate margin to current limits with MOX. This may become an issue if limits are reduced based on reactivity insertion accident experiments, particularly at higher burnups. The consequences of some transients/accidents may be less severe with the weapons-grade plutonium because of a more negative Doppler coefficient (relative to recycle plutonium).

## 2.4 FUEL PERFORMANCE ISSUES OF MOX VERSUS UO<sub>2</sub>

Table 4 compares fuel performance issues of MOX versus  $UO_2$ . The key concerns with MOX are apparently accelerated fission gas release starting at about 40 GWD/MTHM, and the potential for a higher level of clad waterside corrosion at elevated burnups. An important contributor to both of these effects is the higher power produced (and higher temperature) in MOX at higher burnups relative to  $UO_2$ . If the current information is borne out, fission gas release could be the limiting factor of MOX rods.

### 2.5 FRESH FUEL HANDLING

MOX assembly handling, particularly with MOX from recycled plutonium, raises some issues relative to UO<sub>2</sub> (see Table 5). One issue is radiation exposure during refueling, storage and general handling. For recycled plutonium MOX assemblies in particular, there is small measurable neutron and gamma radiation. Neutron radiation derives mainly from spontaneous fission and  $\alpha$ , n reactions in <sup>238</sup>Pu, <sup>240</sup>Pu and <sup>241</sup>Am, whereas most medium- to high-energy gamma radiation comes from <sup>237</sup>U (from  $\alpha$  decay of 2<sup>241</sup>Pu) and decay of a <sup>236</sup>Pu daughter. All of these isotopes are either absent or present in much lower quantities in weapons-grade plutonium. However, even with recycled plutonium MOX assemblies, analysis shows that during refueling operations the total exposure is only a small fraction of the annual overall dose commitment to the operations crew, and ICRP limits are not exceeded.

Fresh recycle plutonium dry storage may require shielding, as would general handling operations for personnel working closely to the fuel assembly surface for modestly long-time periods. Another issue related to storage of fresh recycled plutonium MOX assemblies is decay heat removal in dry storage. Decay heat is primarily from <sup>238</sup>Pu—thus, not an issue with MOX from weapons-grade plutonium.

## 2.6 SPENT FUEL MANAGEMENT ISSUES

Management of spent MOX fuel poses an interesting challenge to utilities due primarily to the much slower rate of reduction in the amount of decay heat. Figure 1 presents the decay heat load versus time for  $UO_2$  and MOX fuels. It is readily seen that the MOX fuel decays at a much slower rate than the uranium fuel.

As an example, consider a reactor with 193 assemblies in the core using one-third MOX loads 21 MOX assemblies per reload. If the spent fuel pool has a capacity of 5 reloads plus full-core reserve, it would be choked after 9 MOX discharges of 21 assemblies. At this time, this poses a difficult problem. The pool can be reracked, but there are significant issues related to dose that are not present for  $UO_2$  fuel. Another alternative is to place the MOX fuel into dry storage. This also poses a problem since the high decay heat load of the MOX fuel severely limits the number of assemblies that can be stored in a given storage unit. At this time, the economics of dry spent MOX storage are not very attractive.

In addition to the above, there is also the question of heat removal from the spent fuel pool. The MOX assemblies are about twice as hot as the  $UO_2$  assemblies after about 5 years of decay. Therefore, the load on the spent fuel pool heat exchangers can be significant and may ultimately exceed its capability.

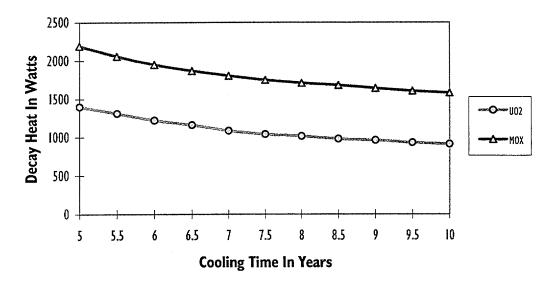
COMMENTS	Exposure is small fraction of annual overall dose commitment to operations crew	ICRP limits not exceeded	German regulations require local dose < 10 mSv/yr	May require shielding	Wet storage could introduce surface contamination to	fresh assemblies Shielding would be required for recycled MOX for personnel working closer than 1 meter to the fuel assembly surface for > 1 hour
EFFECT OF MOX Pu WEPONS GRADE Pu	Much less than recycle case - Essentially no <sup>241</sup> Pu, <sup>241</sup> Am, <sup>236</sup> Pu, - Low <sup>240</sup> Pu, <sup>238</sup> Pu	Not an issue - No <sup>238</sup> Pu	As under A1		Same	As under A1
EFFECT RECYCLE Pu	Small measurable radiation exposure to operations crew (neutron and gamma)	Dry Storage requires cooling of 20-25 watt/kg Pu decay heat	As under A1		Could require absorbers	As under A1
ISSUE	Radiation Exposure	Decay heat (mainly from <sup>238</sup> Pu	Radiation exposure		Criticality if wet storage used	Radiation Exposure
ITEM	<ul> <li>A. Normal Handling</li> <li>1. Refueling Operations</li> </ul>	2. Fresh MOX Storage				3. General Handling Operations

TABLE 5 FRESH FUEL HANDLING ISSUES AT THE NUCLEAR PLANT (MOX VERSUS UO<sub>2</sub>)

	EFFECT OF MOX COMMENTS	RECYCLE Pu WEPONS GRADE Pu		coil – . Enhanced fuel handling Event less severe than – Use of fire retardant	procedures/training recycle case (less $\alpha$	required inventory from long half-life	it (low – Additional care during <sup>239</sup> Pu event of a fire	assembly transport - Careful checking of all	equipment in transport	equipment in transport path
ON COONTA VOMI INTELIVERTOON TITL IN CROCELON	ISSUE EFFECT OF N					required	<ul> <li>Additional care during</li> </ul>	probability event) assembly transport		
TTANKII TAN'I WEAR'S CATAKI	ITEM		B. Abnormal Events	Spill and Dispersion of Rele		pelle	fresh	prob		

TABLE 5 FRESH FUEL HANDLING ISSUES AT THE NUCLEAR PLANT (MOX VERSUS UO,) (continued)

Decay Heat vs. Time UO<sub>2</sub> and MOX Fuels



## 2.7 COMPARISON OF WESTINGHOUSE AND ABB-CE CORE DESIGNS FOR USE ON WEAPONS PLUTONIUM DISPOSITION

## 2.7.1 WESTINGHOUSE ANNUAL CYCLE CORE DESIGN

Westinghouse developed a MOX assembly core intended to maximize plutonium disposition while requiring no additional control rods, control material changes or enriched <sup>10</sup>B in the cooling water.

The equilibrium cycle uses 64 feed assemblies in an out-in loading pattern. A small number of pyrex burnable absorber rods (288 per reload) is used to control power peaking. The design does not require any IFBAs. The assemblies use a total of plutonium fraction of 3.19 w/o. With this fuel the shutdown margin was the limiting factor. The shutdown margin can just be met with a 275 effective full power day (EFPD) cycle (annual cycle at 75% capacity factor) and a 3 batch refueling. The peaking factors meet all of the existing design limits. Further the moderator temperature coefficients are somewhat more negative than for a standard all UO<sub>2</sub> core, but were within the limits typically assumed in safety analyses.

## 2.7.2 WESTINGHOUSE 18-MONTH CYCLE CORE DESIGN

Although the annual cycle is capable of operating without any modifications to the plant's systems, an annual cycle is not very interesting to U.S. utilities. Westinghouse found that 18-month cycle designs could be achieved having capacity factors ranging between 80% (438 EFPD) and 94% (514 EFPD) and discharge burnup of ~ 45 GWD/MTHM. The designs use 84 and 92 MOX assembly reloads, respectively. Due to shutdown margin limitations, core configurations can be achieved by using hybrid control rods incorporating enriched B<sub>4</sub>C or increasing the number of control rods from 53 to 65. Many Westinghouse 4-loop plants have extra control positions that can be used to add these additional rod clusters without any modification to the upper head of the reactor vessel.

The 514 EFPD cycle design also used enriched boron (40 w/o) to maintain the critical boron concentrations at typical levels of equivalent all-UO<sub>2</sub> cores.

Analysis of the 493 EFPD equilibrium cycle showed:

- Power distributions are well behaved and comparable to UO<sub>2</sub> core power distributions
- $F_{\Delta H}$  and  $F_{O}$  limits are met
- Axial offsets are slightly more negative
- Shutdown margin can be met with an excess about equivalent to a full  $UO_2$  core if the enriched hybrid  $B_4C$  control rods are used (or additional standard control rods)
- The plutonium becomes slightly denatured (~ 30% <sup>240</sup>Pu), and 70% of the plutonium is burned.

## 2.7.3 ABB-CE 18-MONTH CYCLE CORE DESIGN

ABB-CE developed an 18-month cycle for a 100% MOX System 80, 241 assembly core using 16 x 16 fuel assemblies. The assembly design utilizes 1.6 w/o erbia as the burnable absorber. The preliminary fuel/core design is for a 449 EFPD cycle (18 months at 82% capacity factor), with a discharge burnup of 32.5 GWD/MTHM. Half of the core is reloaded at equilibrium with 80 assemblies containing erbia and 40 with no burnable absorber. The plutonium loading is 4.5 w/o plutonium (~4.2 w/o Pu fissile).

CE determined that the eight unused control element drive mechanism nozzles (CEDMs) in the reactor vessel top head would be needed to achieve adequate shutdown margin.

## 2.7.4 COMPARISON OF WESTINGHOUSE AND ABB-CE 18-MONTH CYCLE CORE DESIGNS

On the surface, there appears to be a significant dichotomy between the ABB-CE 18-month cycle and the Westinghouse 18-month cycle. Both cores employ 100% weapons grade MOX assemblies, but with the differences shown in Table 6.

## TABLE 6 CYCLE PARAMETER COMPARISON

	Westinghouse	ABB-CE
Cycle Energy (EFPD)	493	449
Cycle Burnup (GWD/MTHM)	21.6	16.3
Discharge burnup (GWD/MTHM)	45.1	32.5
Reload Fraction (% of core)	50	50
Average Reload Enrichment ( w/o	4.1	4.2
Pu fissile)		

The Westinghouse cycle has significantly higher duty than the ABB-CE cycle yet has a lower fissile plutonium content in the reload assemblies. This may be explained in part by the fact that Westinghouse has designed a more efficient fuel cycle than ABB-CE, but efficiency is not as important as rapid disposition in designing weapons-grade plutonium cycles.

The discharge burnup of the ABB-CE cycle is sufficient to achieve discharge <sup>240</sup>Pu concentrations of 23 w/o meeting the generally acceptable minimum denaturing limit (>20 w/o <sup>240</sup>Pu). Westinghouse fuel would have 30 w/o <sup>240</sup>Pu at discharge. It is important to note that the annual weapons throughput for the ABB-CE cycle is about 1.5 MT Pu versus about 1.1 MT Pu for Westinghouse. The ABB-CE assemblies would have a relatively high level of reactivity at discharge which could lead to criticality problems with current storage systems.

## 3.0 ECONOMIC OVERVIEW

### 3.1 INTRODUCTION

The economic comparison of  $UO_2$  and MOX assemblies is most straightforward if one assumes that the plutonium is available at no cost. The cost of MOX fuel then becomes the cost of fabrication. Other costs related to system modifications or equipment changes are one-time costs and can be included after the direct fuel cost fabrication is completed.

## 3.2 THE FABRICATION SITUATION

There is little excess MOX fabrication capacity available. In any event, none of the existing European plants is designed to produce MOX fuel from weapons plutonium on a routine basis. In light of this fact, one option is to develop a U.S. MOX facility. Another option is to augment or add to European MOX facilities and then fabricate the MOX fuel in Europe for subsequent delivery to U.S. utilities.

The Belgian, British and French MOX suppliers all had concepts that would provide MOX capacity to handle U.S. plutonium; however, they are all now concentrating their efforts within consortia planning to bid for work in connection with a U.S.-based MOX plant.

There are two options open in the United States: modification and completion of existing partially completed facilities at Hanford, or construction of a new plant, possibly with the support of European MOX suppliers. The option to upgrade the existing Fuel Materials Examination Facility (FMEF) at Hanford appears to be favored in the United States, although the technical, cost and institutional issues related to completion have not yet been fully evaluated.

## 3.2 MOX PRICES, PRODUCTION COSTS AND ECONOMICS

MOX fuel fabrication unit costs in Europe can be as much as 5 to 10 times higher than equivalent uranium fuel fabrication unit costs. The large range is due to many factors, including specific supplier, fuel type (BWR or PWR) and campaign size. Contracting for MOX business in the second half of the nineties has seen some sign prices for MOX fuel are being reduced.

BWR MOX prices are typically higher than PWR for two main reasons:

- Reduced throughput at the MOX rod stage, due to more plutonium assays
- Extra costs that are normally attributable to BWR UOX fuel, such as hardware, assembly and conversion costs

Long campaigns with a single, three-assay fuel design can reduce costs up to 20% based on cost estimates for a large MOX plant.

The economics of MOX and UOX fuels must be compared to determine the relative acceptability of MOX fuel derived from weapons material if a U.S. utility is going to agree to utilize the MOX fuel. At current market prices the cost of one kilogram of 4.2 w/o enriched uranium is between \$870 to 1050. The wide range is primarily due to differences in spot market and long-term contract prices for uranium and uranium enriching services. The total cost of fabricated fuel at 4.2 w/o in the United States for PWRs range from \$1070 to 1250 per kilogram. MOX prices in Europe to date have been substantially higher. By comparison the estimated prices for MOX fuel are in the range of \$1360 to \$2700 per KgHM.

The minimum difference is \$600/KgHM, which is not insignificant for a reload containing about 9 tons of MOX fuel (1/3 of a 27 MTHM reload) or about \$5.4 million.

There is clearly no direct financial incentive for U.S. utilities to use MOX fuel. For most utilities looking at the possible use of weapons plutonium in MOX fuel a substantial subsidy would be required to offset the higher costs of MOX fuel compared to UOX fuel; however, policy decisions must take into account the feasibility and cost uncertainties associated with the alternative of storage, conditioning and disposal. Depending on the results of such a comparison the requisite subsidies may present the less costly alternative.



## MOX FUEL DEVELOPMENT: EXPERIENCE IN ARGENTINA



XA9953264

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#### Abstract

Since 1973, when a laboratory conceived for the safe manipulation of a few hundred grams of plutonium was built, the CNEA (Argentinean Atomic Energy Commission) has been involved in the small-scale development of MOX fuel technology. The plutonium laboratory consists in a glove box facility (a Facility) featuring the necessary equipment to prepare MOX fuel rods for experimental irradiations and to carry out studies on preparative processes development and chemical and physical characterization. The irradiation of the first prototypes of (U,Pu)O2 fuels fabricated in Argentina began in 1986. These experiments were carried out in the HFR (High Flux Reactor)- Petten, Holland. The rods were prepared and controlled in the CNEA's  $\alpha$  Facility. The post-irradiation examinations (PIE) were performed in the KFK (Kernforschungszenttrum Karlsruhe), Germany and the JRC (Joint Research Center), Petten. In the period 1991-1995, the development of new laboratory methods of co-conversion of uranium and plutonium were carried out: reverse strike co-precipitation of ADU-Pu(OH)<sub>4</sub> and direct denitration using microwaves. The reverse strike process produced pellets with a high sintered density, excellent microhomogeneity and good solubility in nitric acid. Liquid wastes showed a very low content of actinides and the process is easy to operate in a glove box environment. The microwave direct denitration was optimized with uranium alone and the conditions to obtain high density pellets, with a good microstructure, without using a milling step, have been developed. At present, new experiments are being carried out to improve the reverse strike co-precipitation process and direct microwave denitration. A new glove box is being installed at the plutonium laboratory, this glove box has process equipment designed to recover scrap from previous fabrication campaigns, and to co-convert mixed U-Pu solutions by direct microwave denitration.

## 1. INTRODUCTION

A program for the small-scale development of MOX fuel technology at the CNEA-Argentina was initiated soon after commissioning of a plutonium laboratory ( $\alpha$  Facility) in 1973. The aim of this program was to study MOX fuel preparation processes; to produce prototype MOX fuel rods for experimental irradiations and to master safety issues related to plutonium handling.

Six rods containing (U,Pu)O<sub>2</sub> MOX fuel were fabricated and controlled in the CNEA's  $\alpha$  Facility [1], and the irradiation of these prototypes began in 1986 [2]in the HFR- Petten, Holland.

The first rod has been used for destructive pre-irradiation analysis. The second one was a pathfinder to adjust systems in the HFR [3]. Two additional rods including iodine doped pellets, were intended to simulate 15.000 MWd/t(M). The remaining two rods were irradiated until 15.000 MWd/t(M). One of them underwent a final ramp with the aim of verifying fabrication process and studying the behavior under power transients. The postirradiation examinations were performed in the Kernforschungszentrum Karlsruhe, Germany and in the JRC, Petten.

In the period 1991-1995, two processes of co-conversion of uranium and plutonium have been studied under IAEA's Research Contract N° 6742: reverse strike co-precipitation of ADU-Pu(OH)<sub>4</sub> and direct denitration using microwaves.

The selection of these processes was based in their potential advantages: relative simplicity of operation, feasibility to be adapted to glove box work, possibility of producing high density pellets with good microhomogeneity and nitric acid solubility.

At present, new experiments are being carried out to improve the reverse strike co-precipitation process and direct microwave denitration. A new glove box is being installed at the plutonium laboratory, this glove box has process equipment designed to recover scrap from previous fabrication campaigns, and to co-convert mixed U-Pu solutions by direct microwave denitration.

## 2. EXPERIMENTS AND RESULTS

## 2. 1. Irradiation test

The rods were originally designed for the MZFR (MehrZweckForschungsReactor) Karlsruhe, Germany. However, due do the decommissioning of that reactor, the experiments were performed in the HFR, Petten. Some rod dimensions (i.e. fuel length) and the plugs geometry have been modified to meet the specifications of HFR.

Three types of fuel rods were fabricated. Table I shows their main characteristics.

TABLE I. FUEL KUDS CHARACTE	KISTICS (U	ada he saqqa see sa	1 <b>2                                    </b>
(a) Fuel rod type	A.1	A.3	A.4
Length	24.1	17.9	17.9
Pellets number	21	16	16
Compensating pellets number	1	-	-
Doped pellets number	-	1	3
Doping material	-	Ι	CsI / Mo
Doping material (mg)	-	3	6.604±0.017 / 1.373±0.036
Simulated burnup (MWd/ton (M))	-	13761±4933	14800±390
Filling gas	He	He	He
Filling pressure (atmosphere)	1.15	1.15	1.15
(b) Pellets			
Density		10.52±0	$0.04 \text{ g/cm}^3$
Pellet height		1.12±	0.01 cm
Pellet diameter		$1.040 \pm$	0.001 cm
$Pu_{fiss}/U+Pu_{met}$		0.:	53%
Enrichment (U235+Pu)		1.2	25%
O/M relation		2	.00
Dishing volume		25±:	$5 \text{ mm}^3$
(c) Cladding			
Material		Zı	ry-4
Inner diameter		1.1	7 cm
Thickness		0.0	6 cm

TABLE I. FUEL RODS CHARACTERISTICS (U/PU MIXED OXIDE)

The first irradiation was with the A.1.4 rod to adjust systems in the HFR reactor instruments. The experiment verified the response of the HFR reactor control systems with our MOX rod.

The duration of the irradiation was about 100 h. It included a final tamp test. The maximum power level reached was 439 W/cm. [4]

A second experiment started with the irradiation of two fuel rods both containing iodine compounds as a dopant to simulate the effect of extending burnup [2].

Fuel rods A.3 and A.4 have been employed for this second experiment. The irradiation, carried out during 15 days consisted in a steady state condition including two powers cycling between 120 and 290 W/cm and a final ramp to reach 400 W/cm [5]. This final power level lead to a "hoop stress" of about 170 MPa which is likely to induce microcracks in the inner surface of the cladding, representing an incipient defect produced by stress corrosion cracking without reaching the failure threshold of the tubing.

CsI and Mo were mixed with  $UO_2$  (rod A.3), the mixture was introduced in central holes drilled in three pellets and hand pressed. Pure iodine was introduced in one pellet (rod A.4) with the aim of comparing its effect on the cladding with that of combined iodine (CsI in rod A.3).

Behavior of rods A.3 and A.4 was previously described [6-8]:

- there were no failures in the cladding
- microcracks in the inner surface of the cladding were detected during PIE.

The third irradiation experiment (BU15) was carried out with fuel rods A.1.2 and A.1.3, both similar to the pathfinder rod. The power history for this irradiation test was proposed upon calculations made with the BACO (BArra COmbustible) code. The target of the irradiation was an average burnup of 15000 MWd/t (M) (more or less twice the final burnup for Atucha I fuel), which was reached in two main steps:

(a) Up to a burnup of \$100 MWd/t (M) the rods were irradiated in different locations of the HFR core.

(b) Up to a burnup of 15000 MWd/t (M) both rods were irradiated in the Pulse Side Facility (PSF) of the reactor.

During the steady state irradiation both rods were assembled together through a threaded coupling and after termination of the bulk irradiation phase both fuel rods were disassembled in the Petten hot cells.

After the steady rate irradiation at an average power level of 230 W/cm one of the rods (A.1.3) was submitted to a power ramp which was divided in two parts:

- (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) a ramp test starting with a ramp rate of 50 W cm-1 min-1 from nearly zero power to maximum 420 W/cm linear fuel rod power and followed by a 6 h steady-state holding at this maximum power.

Both rods behaved during the stationary phase as it was expected:

- no rod failures were detected
- no fabrication defects were evident

During EOL (End of Life) power ramp, A.1.3 behaved as follows:

- a maximum power of 390 W/cm was reached due to the low quantity of remaining fissile material in the fuel rod.
- 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 [9].

Post-irradiation examinations of the BU15 experiment included: visual inspection, Eddy current check, neutron radiography, gamma scanning, dimensional control and microscopic examinations including  $\alpha$  and  $\beta$  autoradiography, pore and grain size, evaluation and identification of the failure in pin A.1.3. The main featuring of PIE and a comparison between the results of the PIE and the BACO code simulation have been presented previously [10,11]. The conclusions can be summarized as it follows:

- All the rods behaved as expected, including the defective rod.
- The maximum hoop stress and cladding radial contact pressure appeared in the axial section corresponding with the failure.

- There was a good agreement between experimental results and BACO code predictions, especially with respect to the relationship between the failure position and characteristics and the mechanical demands predicted.
- 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, the micro-structure observed and the grain size distribution indicated a good BACO code evaluation.
- Post irradiation examinations confirmed the calculated value of hoop stress (σv = 207 MPa). This value indicated that PCI (Pellet Cladding Interaction) - SCC (Stress Corrosion Cracking) was a mechanism likely to produce the failure.

## 2. 2 Co-conversion methods

Two co-conversion methods were selected: reverse strike co-precipitation and direct denitration using microwaves. They usually include several process steps shown in Fig. 1.

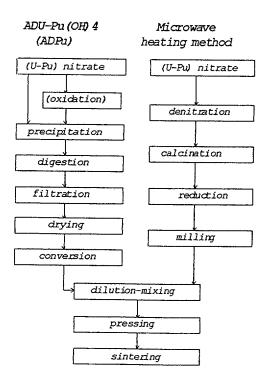


FIG. 1. Diagram of co-conversion methods

### 2. 2. 1. Reverse strike co-precipitation method

In this process, a nitric actinide solution and ammonia gas are continuously fed into a reactor which is thermostatized at the chosen temperature and having an initial volume of NH<sub>4</sub>OH solution at pH between 4 and 6. The pH is kept constant during all the process while the nitric actinide solution and the ammonia (gas) are injected in the reactor with continuous stirring to prevent preferential precipitation and assure a good precipitate filterability.

When the reaction is finished, the stirring of the slurry continues for one more hour, always at the same temperature. Afterwards the slurry is filtered and dried. The precipitation apparatus is shown in Fig. 2.

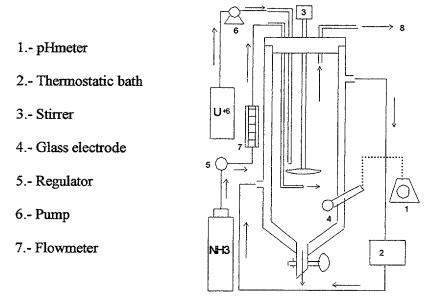


FIG. 2. Precipitation apparatus

The reverse strike process was first studied with uranium alone in the same conditions of the co-precipitation with plutonium.

Two parameters were specially considered in this step:

- Temperature (20, 40 and 60 °C)

- Actinide concentration in the uranyl-nitrate feeding solution (100, 200 and 300 g[U]/l)

The ADU (ammonium diuranate) obtained was converted to  $UO_2$  (uranium dioxide) in two steps:

- decomposition to UO<sub>3</sub> in N<sub>2</sub> atmosphere at 400 °C and

- reduction of UO<sub>3</sub> to UO<sub>2</sub> in N<sub>2</sub>/H<sub>2</sub>(8%) atmosphere at about 650 °C during one hour.

The following analytical determinations have been carried out on the powders prepared under each condition of precipitation: O/M (oxigen/metal ratio), SSA (specific surface area), particle size distribution, flowability and tap density ( $\delta_{tap}$ ).

Several milling, pressing and sintering conditions were studied.

Milling was carried out in a ball-mill with stainless steel jar and Cr-steel milling balls. Tests with different charge ratios (ratio between balls weight and powder weight), different ball size distributions and different milling times were performed.

In the pressing, two different kinds of lubrication were used:

Internal lubrication: zinc stearate was added in different percentages in some batches

- External lubrication: a vegetable oil was used to lubricate the press die without adding zinc stearate.

Finally, the sintering process was studied choosing two heating profiles shown in Fig. 3.

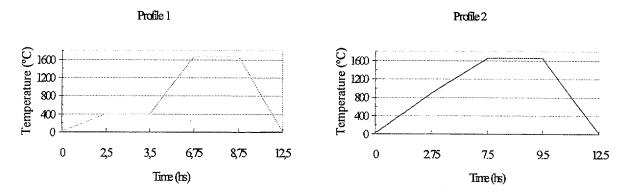


FIG. 3. Time-temperature schedule of the sintering cycles

The results of this step can be summarized as it follows:

- At 20 °C the precipitate was gelatinous and difficult to filtrate
- UO<sub>2</sub> powders obtained from solutions precipitated at 60 °C (with 200 g/l and 300 g/l of actinides in initial solution concentration) showed best sinterability.
- Low content of actinides in liquid wastes (lower than 3 µg/l) and easy operativity.
- UO<sub>2</sub> powders were free flowing, making suitable the automatic pressing.
- The different heating profiles used for sintering showed an important influence on the final density of the sintered pellets.

Typical characteristics of ADU and  $UO_2$  powders obtained from nitric solutions containing 200 and 300 g[U]/l which were precipitated at 60 °C (Types 200/60 and 300/60 respectively) are shown in Table II, and typical results obtained from the pressing and sintering tests on milled and unmilled  $UO_2$  are shown in Table III.

## TABLE II. CHARACTERISTICS OF BATCHES OBTAINED FROM ADU PRECIPITATED

ADU						U	O <sub>2</sub>			
Туре	¢ <sup>a</sup>	SSA	Unmilled				Milled			
	(µm)	(m <sup>2</sup> /g)	φ <sup>a</sup> (μm)	SSA (m²/g)	O/M	$\delta_{tap}$	φ <sup>a</sup> (μm)	SSA (m²/g)	O/M	$\delta_{tap}$
200/60	13.5	2.44	8.3	3.4	2.08	2.49	3.0	4.08	2.11	3.39
300/60	12	2.07	8.3	3.4	2.06	2.54	3.0	4.11	2.04	3.46

<sup>a</sup> Average particle diameter

## TABLE III. PRESSING AND SINTERING TESTS OF UO2 POWDERS (VIA ADU<sup>a</sup>)

		Compacting	Green density	Sinter G/cm <sup>3</sup> 9.85 9.95 10.26	red density	
		pressure <sup>b</sup> (MPa)	g/cm <sup>3</sup>	G/cm <sup>3</sup>	% Theoretica	
	Profile 1	500	6.54	9.85	89.9	
Unmilled	Prome 1	600	6.71	9.95	90.8	
Ummed	Profile 2	500	6.54	10.26	93.6	
	Pionie 2	600	6.71	G/cm <sup>3</sup> 9.85 9.95	94.3	
	Drafile 1	500	6.53	10.40	94.9	
	Profile 1	600	6.74	10.42	95.1	
Milled		500	6.53	G/cm <sup>3</sup> 9.85 9.95 10.26 10.33 10.40 10.42 10.55	96.3	
	Profile 2	600	6.74		96.4	

<sup>a</sup> 300/60 Type

<sup>b</sup> external lubrication

The ceramographic analysis of the sintered pellets revealed a homogeneous distribution of pores with average values of grain size between 6 and 7  $\mu$ m.

The equipment for mixed reverse strike coprecipitation with Pu<sup>+4</sup> was placed inside a glove box. Based on the results obtained in previous steps (with uranium alone), tests with a mixed (U,Pu) solution were carried out.

The conditions were:

_	Solution volume (ml)	474
-	Pu <sup>+4</sup> contents $(g/l)$	57.03
-	U <sup>+6</sup> contents $(g/l)$	228.12
-	Ratio (U/Pu+U)x100	20
-	pH of precipitation	4.5 - 5.5
-	Thermostatic bath temperature (°C)	60

These tests indicated:

- Low content of uranium and plutonium (< 3 µg/l, N.D.) in liquid wastes.
- Homogeneous green-yellowish color in the obtained product
- Excellent filterability of the precipitate.

Several tests of calcination-reduction were performed in order to reduce de SSA of the powders and to stabilize towards oxidation in air. At 700 °C during 3 hours the characteristics of the  $(U,Pu)O_2$  were: SSA 5.74 m<sup>2</sup>/g -  $\delta_{tap}$  1.46 g/cm<sup>3</sup>.

The pressing of these powders was difficult due to their low tap density. They had to be pressed between 110-170 MPa and sintered using the profile 2 (see Fig 3). Table IV shows the obtained results (average).

TABLE IV. PRESSING AND SINTERING TEST OF (U 0.8, Pu 0.2)O2 POWDERS

		والمحافظ المحافظ والمحافظ والمحافظ والمحافظ المحافظ والمحافظ والمحاف
Green density (g/cm <sup>3</sup> )	Sintered density (g/cm <sup>3</sup> )	% Theoretical Density
4.8	10.63	96.2

The sintered densities were very high and it can been adjusted to agree with the specified values for each reactor.

Ceramographic analysis showed:

- inhomogeneous pore distribution with a higher concentration of pores in some zones of the sintered pellets,
- homogeneous grain size distribution, and
- absence of zones with PuO<sub>2</sub> islands.

Nitric acid solubility tests were performed in order to verify the solubility of unirradiated sintered pellet. Pellets were attacked with boiling nitric acid 7 M during 6 hours using a reflux condenser. They were totally dissolved at the end of 3 hours' attack.

The result was S% = 99.7%, where the percentage solubility (S%) is defined as:

 $S = P \times (P+N)^{-1} \times 100$ 

where

- P is the mass of plutonium found by analysis in the dissolution mixture,
- N is the mass of plutonium found in the undissolved solid retained by filtration (with blue ribbon paper).

# 2. 2. 2. Direct denitration using microwaves

The direct denitration using microwaves heating method was already used in other countries for mixed oxides preparation [12, 13]. As in the case of coprecipitation, a preliminary set of tests with uranium was carried out.

The direct denitration of a solution consists in the evaporation of the nitric acid and the subsequent thermal decomposition of the solid formed.

The solutions used in different essays were prepared in batches of 2 liters with an uranium concentration of 280 g[U]/l in nitric acid 1 M. These levels of concentration and acidity were chosen in order to avoid polymerization and precipitation effects on plutonium [14].

The denitration was carried out in standard kitchen microwave ovens having the same power level but featuring different chamber capacity and geometric inlet of microwaves in the denitration chamber. It was necessary to make adaptations to prevent NO<sub>3</sub>H vapors and nitrogen oxides (NOX) corrosion on the structure and electronic circuits of the oven (see Fig. 4).

- 1. Denitration chamber
- 2. Pyrex glass vessel
- 3. Vacuum gauge
- 4. Refrigerator
- 5. Condenser 6. Scrubber
- 7. Vacuum pump
- 8. Microwave power unit
- 9. Microwave guide

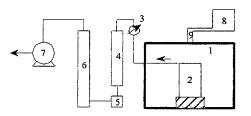


FIG. 4 Denitration device

The solutions were placed into Pyrex glass vessel. The orange product attained, easy to remove from its container, sponge-like, was crumbled with a hand mortar.

Different parameters of each step were studied:

- Based on results obtained in previous works [15], ammonium nitrate was added to the initial solution of one of the batches in order to study the influence of this salt on the final product quality.
- Different reduction and sintering profiles were tried.
- The influence of jar rotation speed, time and charge ratios (see Sec. 2. 2. 1.) was determinate in the milling process.
- Internal lubrication and external lubrication were performed in the pressing step.

The produced powders were inspected for:

- Particle size distribution by means X-ray monitored sedimentation technique.
- SSA by adsorption of helium method.
- O/M ratio by spectrophotometric technique.
- Tap density
- Morphology using a scanner electronic microscope (SEM).
- Composition by X-ray diffraction.

The following conclusions can be drawn:

- Significant improvements on the final pellet density were not observed with the addition of ammonium nitrate in the starting nitric solution.
- The obtained product from the denitration is a mixture of  $UO_3$ ,  $U_3O_8$  and  $UO_2(NO_3)_2$  xH<sub>2</sub>0.
- The low content of actinides in the liquid wastes generated during the process implies a recovery of about 99.996 % of the original actinide mass (uranium concentration in the distilled liquids less than 19 µg/ml).
- It is possible to control the SSA of UO<sub>2</sub> powders with different calcination-reduction cycles but this parameter has almost no influence on the sintered density of the pellets.
- The milling process reduce the average size of the particles and increases de SSA, the tap density and the green density. The final result is an important increase in the sintered density.
- The presence of a internal lubricant does not affect the pellet densities if the heating rate, and the dewaxing time and temperature are properly chosen for the lubricant that is used.
- Lower heating rates above 900 °C allow a better densification of the ceramic material during the sintering process. The best conditions can be obtained by dilatometry studies [16].

It was found that the powders prepared in microwave oven 1 reached higher green and ntered densities for the same pressing conditions than those produced in microwave oven 2 (see Fig. 5). Then, further studies were carried out to explain this phenomenon.

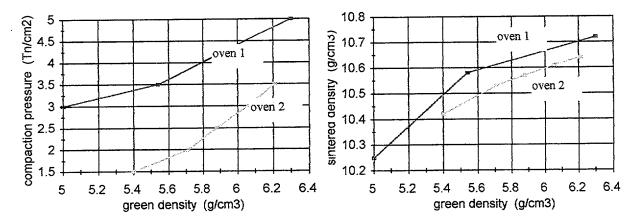


FIG. 5 Pressing a sintering test of  $UO_2$  powders obtained from different microwave ovens (1 Tn/cm<sup>2</sup> = 100 MPa)

Several tests under different pressures within the closed reactor vessel have been carried out. The results of these tests are shown in Table V.

TABLE V. CHARACTERISTICS OF UNMILLED UO2 POW	WDERS OBTAINED UNDER DIFFER-
ENT PRESSURE CONDITIONS.	

Pressure (mbar)	972 (A)	632 (B)	143 (C)
O/M	2.02	2.05	2.06
φ (μm)	0.5	0.45	3.7
SSA (m <sup>2</sup> /g)	2.10	2.29	1.90
Tap density (g/cm <sup>3</sup> )	2.4	2.35	2.24
Bulk density of denitrated product (g/cm <sup>3</sup> )	0.57	0.54	0.40

A reduction in the pressure within the reactor vessel causes a decrease on the bulk density of denitrated product and larger average particle size are attained.

The pressing and sintering results for these powders (without a milling process) are shown in Fig. 6. They indicate that the sintered density of the pellets can be controlled by adjusting the pressure in the reactor vessel during denitration. Higher pressures lead to smaller particle average size and consequently to higher sintered density. A patent for this procedure is being produced in Argentina.

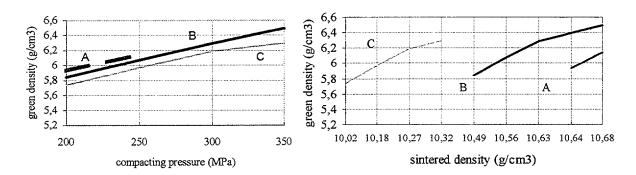


FIG.6 Pressing and sintering test of  $UO_2$  powders obtained from different denitration conditions

At present, a new glove box is being built. The equipment has been designed to recover scraps and to co-convert mixed U-Pu solutions by direct microwave denitration.

# 3. CONCLUSIONS

The continuous operation during more than 20 years of a small laboratory for the development of MOX fuel technology in Argentina produced the following achievements:

- The fabrication and control of MOX fuel rods for an experimental irradiation and a comparison between the experimental results of the postirradiation examinations and the BACO code calculations, producing an upgrade in the code.
- The development of co-conversion processes to produce MOX with significant improvements in the microwave direct denitration method.
- The training of the personnel in safe handling of a few grams of plutonium.
- The development of chemical and physical characterization techniques for MOX fuels.

At present, the research and development continue with the installation of a new glove box. This glove box is equipped to carry out experiments to prepare MOX fuel by direct microwave denitration.

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# PERFORMANCE TESTING OF CANDU MOX FUEL

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# Abstract

CANDU<sup>®</sup> fuel bundles containing 0.5 wt % plutonium in natural uranium were fabricated at Chalk River Laboratories and were successfully irradiated in the NRU reactor at powers up to 65 kW/m and to burnups ranging from 13 to 23 MW•d/kg HE. Two of the bundles experienced power histories that bound the normal powers and burnups of natural UO<sub>2</sub> CANDU fuel (<65 kW/m, to burnups of 13 to 15 MW•d/kg HE). These bundles exhibited sheath strain and fission-gas release (FGR) typical of those observed in similarly designed and operated UO<sub>2</sub> fuel. Significantly more grain growth was observed than that typically expected for UO<sub>2</sub> fuel; however, this increase in grain growth had no apparent effect on the overall performance of the fuel. Pellet-centre columnar grain growth was accompanied by plutonium homogenization. Two other MOX bundles operated to extended burnups of 19 to 23 MW•d/kg HE. Burnup extension above 15 MW•d/kg HE had no apparent effect on sheath strain or grain growth, and only a small effect on FGR and the amount of oxide observed on the inner surface of the sheath.

# 1. INTRODUCTION

The development of various types of plutonium-containing mixed-oxide (MOX) fuel by AECL has been on-going for over 30 years. Initially, AECL's MOX program consisted of tests conducted on individual elements, to study the behaviour of MOX pellets operating in conditions representative of CANDU fuel. More recently, the BDL-419 experiment has involved the fabrication, irradiation and post-irradiation examination (PIE) of MOX fuel bundles of the same 37-element geometry as is currently used in commercial CANDU power reactors.

The objectives of the BDL-419 experiment were to

- Demonstrate that (U, Pu)O<sub>2</sub> fuel fabricated in the Recycle Fuel Fabrication Laboratory (RFFL) at Chalk River Laboratories (CRL) is capable of sustaining powers and burnups typical of natural UO<sub>2</sub> fuel currently operating in CANDU power reactors. (Natural UO<sub>2</sub> CANDU fuel is typically irradiated at peak powers <65 kW/m to burnups of approximately 10 MW•d/kg HE.)</li>
- 2. Investigate the performance of CANDU MOX fuel at extended burnups (>18 MW•d/kg HE).

CANDU<sup>®</sup> is a registered trademark of Atomic Energy of Canada Limited (AECL).

This paper presents the results of the PIE of four BDL-419 bundles, designated as ABB, ABC, ABD and ABE, that were irradiated in the U1 and U2 loops of the National Research Universal (NRU) reactor at CRL. Bundles ABD and ABE operated at powers up to 65 kW/m and to burnups ranging from 13 to 15 MW•d/kg HE, thus bounding the normal powers and burnups of natural UO<sub>2</sub> CANDU fuel. Similar powers were observed in bundles ABB and ABC, but to extended burnups ranging from 19 to 23 MW•d/kg HE.

# 2. FUEL DESIGN AND FABRICATION

The fuel elements for bundles ABB, ABC, ABD and ABE were fabricated in the RFFL at CRL [1,2]. The (U, Pu)O<sub>2</sub> pellets for BDL-419 were produced from natural UO<sub>2</sub> blended with PuO<sub>2</sub>. The finished pellets contained 0.5 wt % fissile plutonium in heavy elements. The average density of the pellets was 10.54 g/cm<sup>3</sup>. The pellets were loaded into sheaths coated with DAG-154 CANLUB, a graphite coating used in CANDU fuel to improve power ramp performance. The elements were loaded into bundles that were essentially the same as those used in the Bruce commercial power reactors (operated by Ontario Hydro), except that the central element was eliminated to facilitate the insertion of a guide tube for vertical irradiation in the U1 and U2 loops of the NRU reactor (Figure 1). Fabrication data are summarized in Table I.

# 3. IRRADIATION OF BDL-419 MOX BUNDLES

Bundles ABB, ABC, ABD and ABE were irradiated in the U1 and U2 loops of the NRU reactor under conditions similar to those of commercial CANDU power reactors (Table II). A declining power history was observed in each of the bundles (Figure 2). Bundles ABD and ABE operated at beginning-of-life (BOL) powers of 54 to 65 kW/m to measured burnups ranging from 13 to 15 MW•d/kg HE. Bundles ABB and ABC experienced BOL powers of 50 to 62 kW/m to extended burnups ranging from 19 to 23 MW•d/kg HE. A summary of power and burnup data is shown in Table III.

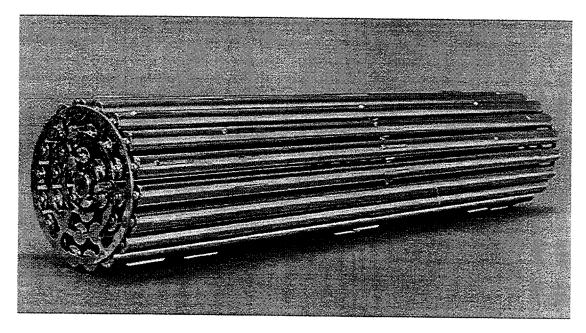


FIGURE 1. 37-Element Bruce CANDU Fuel Bundle Used in BDL-419 MOX Experiment. The Centre Element is Replaced with a Guide Tube to Facilitate Vertical Irradiation in the NRU Loops.

# TABLE I.SUMMARY OF FABRICATION DATA FOR BDL-419 MOX FUEL<br/>BUNDLES ABB, ABC, ABD AND ABE.

Parameter	Value	
Bundle Design	37-Element (Bruce) Geometry	
Overall Length	494 mm	
Heavy Element Weight	18 kg	
Element Data		
Stack Length	470 mm	
Sheath Outer Diameter	13 mm	
Sheathing Material	Cold-worked Zircaloy-4	
Pellet Data		
Manufacturer	RFFL, AECL	
Density	10.54 g/cm <sup>3</sup>	
Outside Diameter	12 mm	
Length	12 mm	
Enrichment	0.5 wt % fissile Pu in heavy elements (Pu + natural U)	

# TABLE II.TYPICAL COOLANT CONDITIONS FOR BDL-419 FUEL OPERATING IN<br/>THE U1 AND U2 LOOPS OF THE NRU REACTOR

Coolant Type	H <sub>2</sub> O
Temperature (°C)	260 - 305
Pressure (MPa)	9.5 - 10.9
Mass Flow (kg/s)	16 - 20
Outlet Quality	0 %
pH	9.5 - 10.5
O <sub>2</sub> Concentration (ppb)	<10
H <sub>2</sub> Concentration (cm <sup>3</sup> /kg)	3 - 15

# TABLE III.SUMMARY OF POWER HISTORY, STRAIN AND FISSION-GAS<br/>RELEASE (FGR) DATA FOR BDL-419 MOX FUEL

	Measured Outer-Element Burnup (MW•d/kg HE)	Outer-Element Power at BOL (kW/m)	Outer-Element Midpellet Strain (%)	Outer-Element Fission-Gas Release (%)
ABB	18.7 - 20.4	50 - 55	-0.1 to 0.0	4 - 5
ABC	22.1 - 22.8	60 - 62	-0.1 to +0.1	12 - 13
ABD	13.0 - 13.3	63 - 65	-0.2 to +0.2	10 - 11
ABE	14.4 - 15.0	54 - 56	-0.3 to 0.0	7 - 9

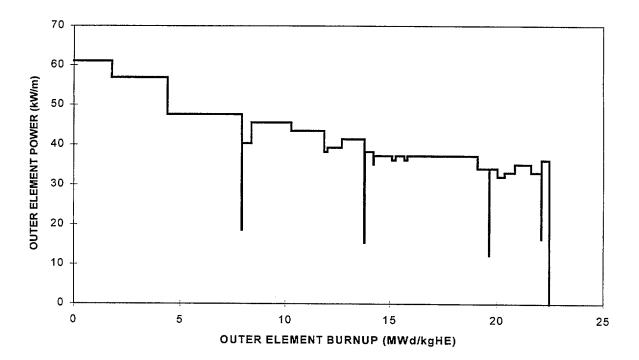


FIGURE 2. Typical Declining Power History for BDL-419 MOX Fuel (Bundle ABC is shown.)

# 4. PIE RESULTS

#### 4.1. Sheath Strain

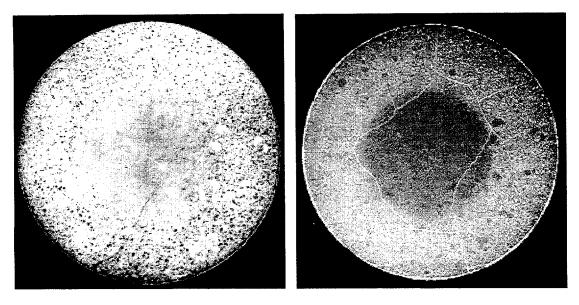
Midpellet residual sheath strains of -0.3% to +0.2% were observed in the outer elements of the four BDL-419 bundles (Table III). These strains are within the range that is normally observed in CANDU UO<sub>2</sub> fuel [3]. The progression in burnup from 13 to 23 MW•d/kg HE appeared to have little, if any, effect on the observed sheath strain. The effect of power was more pronounced; tensile sheath strains were observed in bundles ABC and ABD that operated at BOL powers of 60 to 65 kW/m, whereas bundles ABB and ABE, that operated at BOL powers of 50 to 56 kW/m, exhibited compressive sheath strains.

# 4.2 Fission-Gas Release

Fission-gas release of 4 to 13% was observed in the outer elements of the four BDL-419 bundles. This FGR is within the range that is expected for similarly designed and operated  $UO_2$  fuel [3-5]. As in the case of  $UO_2$  fuel [5], the MOX FGR dependence on burnup is weak relative to that of power. The strong dependence on peak power is observed by comparing the FGR of high-power bundles ABC and ABD (10 to 13% FGR at 60 to 65 kW/m) with the FGR of bundles ABB and ABE (4 to 9% FGR at 50 to 56 kW/m). The weak dependence on burnup is observed by comparing the FGR of bundles ABD and ABC, which had the greatest variance in burnup, but similar powers (10% FGR at 13 MW•d/kg HE vs. 13% FGR at 23 MW•d/kg HE, respectively).

# 4.3 Pellet Microstructural Changes

Figure 3a shows a post-irradiation alpha autoradiograph of an outer-element cross section from bundle ABC. The periphery of the fuel illustrates the as-fabricated inhomogeneous microstructure of the fuel, containing regions with plutonium-rich particles as well as regions







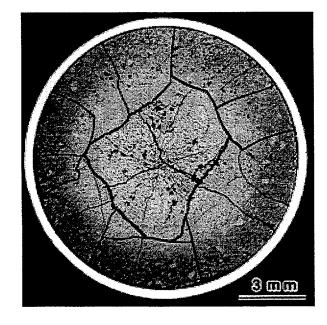




FIGURE 3. Transverse Section from Outer Element of Bundle ABC: (a) Alpha Autoradiograph - dark spots represent Pu-rich particles; lighter areas represent regions that are low in plutonium content. Note that the centre region has undergone Pu homogenization. (b) Beta-Gamma Autoradiograph white spots represent high-fission product regions (predominantly Cs-137) associated with Pu-rich particles. Note fission-product depletion at pellet centre. (c) Optical Micrograph - columnar grain growth is seen in the centre region. low in plutonium content. The central region of the fuel has undergone plutonium homogenization. This effect was observed at powers >55 kW/m and was accompanied by fission-product depletion (Figure 3b, beta-gamma autoradiography) and columnar grain growth (Figure 3c). Columnar grain growth is not normally observed in CANDU UO<sub>2</sub> fuel below powers of 65 kW/m. The progression in burnup from 13 to 23 MW•d/kg HE did not appear to have any effect on the observed microstructural changes; this finding indicates that the degree of grain growth observed was due to BOL (high power) operation, not burnup extension.

The degree of grain growth observed in the BDL-419 MOX fuel relative to the degree expected of  $UO_2$  fuel is indicative of higher operating temperatures or different grain-growth kinetics, or of both. Blanpain et.al. [6] have measured higher operating temperatures in lightwater reactor MOX fuel containing 4 to 5 wt % plutonium. They observed that the difference in MOX and  $UO_2$  centreline temperatures increases with increasing linear power, up to a difference of  $100^{\circ}C$  at 40 kW/m. Temperature measurements were not made on the BDL-419 fuel that contained only 0.5 wt % plutonium and operated at 50 to 65 kW/m at BOL. The columnar grain growth observed in the BDL-419 MOX fuel does not appear to have contributed to increased strain or FGR, which, as noted above, were in the normal range for comparable  $UO_2$  fuels.

# 4.4 Internal Sheath Oxidation

Very little oxide was observed on the inside sheath surface of bundles ABD and ABE that were irradiated to burnups ranging from 13 to 15 MW•d/kg HE. This finding is consistent with the amount of oxide observed in CANDU UO<sub>2</sub> fuel. At burnups ranging from 19 to 23 MW•d/kg HE, the outer elements of bundles ABB and ABC exhibited oxide films, 1 to 8 µm thick, on the inner sheath surface. Similarly designed and operated UO<sub>2</sub> fuel typically exhibits oxide films <1 µm thick [4]. Fissioning of Pu-239 produces less oxideforming elements than does the fissioning of U-235 [7]; consequently, more oxygen is liberated by fissioning in (U, Pu)O<sub>2</sub> than in conventional UO<sub>2</sub> fuels. The BDL-419 experiment suggests that this effect becomes significant at burnups in excess of 15 MW•d/kg HE; notwithstanding, this increase in oxide film on the inner sheath surface of the outer elements has no apparent effect on the overall performance of the CANDU MOX fuel.

# 5. SUMMARY AND CONCLUSIONS

MOX CANDU fuel bundles ABB, ABC, ABD and ABE that contained 0.5 wt % plutonium in natural uranium were successfully irradiated from outer element BOL powers of 50 to 65 kW/m to burnups ranging from 13 to 23 MW•d/kg HE.

The power histories observed in bundles ABD and ABE bound the normal powers and burnups of natural UO<sub>2</sub> CANDU fuel (up to 65 kW/m, to burnups ranging from 13 to 15 MW•d/kg HE). These bundles exhibited sheath strain and FGR typical of that observed in similarly designed and operated UO<sub>2</sub> fuel (up to +0.2% midpellet strain and 9% FGR). Significantly more grain growth was observed than that expected for UO<sub>2</sub> fuel; however, this increase in grain growth had no effect on the overall performance of the fuel. The onset of columnar grain growth at the pellet centre at a power of 56 kW/m was accompanied by plutonium homogenization.

Bundles ABB and ABC operated to extended burnups of 19 to 23 MW•d/kg HE at powers similar to those at which bundles ABD and ABE operated. Burnup extension beyond

15 MW•d/kg HE had no apparent effect on sheath strain and grain growth (similar to that observed in bundles ABD and ABE). The burnup extension had a small effect on FGR (maximum = 13%, similar to that observed in UO<sub>2</sub> fuel) and the amount of oxide observed on the inner surface of the sheath. Neither of these phenomena had any apparent negative effect on the performance of the extended-burnup bundles.

The BDL-419 tests demonstrate that MOX fuel produced in the RFFL at CRL is capable of sustaining powers and burnups typical of natural  $UO_2$  fuel currently operating in CANDU reactors, and that its performance at extended burnups is similar to that expected of  $UO_2$  fuel.

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# STUDY OF ADVANCED LWR CORES FOR EFFECTIVE USE OF PLUTONIUM AND MOX PHYSICS EXPERIMENTS



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#### Abstract

Advanced technologies of full MOX cores have been studied to obtain higher Pu consumption based on the advanced light water reactors (APWRs and ABWRs). For this aim, basic core designs of high moderation lattice (H/HM  $\sim$ 5) have been studied with reduced fuel diameters in fuel assemblies for APWRs and those of high moderation lattice (H/HM  $\sim$ 6) with addition of extra water rods in fuel assemblies for ABWRs. The analysis of equilibrium cores shows that nuclear and thermal hydraulic parameters satisfy the design criteria and the Pu consumption rate increases about 20 %. An experimental program has been carried out to obtain the core parameters of high moderation MOX cores in the EOLE critical facility at the Cadarache Centre as a joint study of NUPEC, CEA and CEA's industrial partners. The experiments include a uranium homogeneous core, two MOX homogeneous cores of different moderation and a PWR assembly mock up core of MOX fuel with high moderation. The program was started from 1996 and will be completed in 2000.

#### 1. INTRODUCTION

In Japan, the MOX fuel demonstration programs have been performed successfully and reload size use of MOX fuel has been prepared. The recycling of plutonium in light water reactors is expected to continue in several decades. As medium and long term development, core concepts have been studied for enhancing consumption of plutonium with the higher moderator to fuel ratio than the conventional fuel lattices in 100 % MOX fuel cores[1-2]. In order to measure the main core physics parameters of high moderation MOX fuel cores, an extensive experimental program, MISTRAL[3], was undertaken as a joint study of the Nuclear Power Engineering Corporation (NUPEC), CEA and its industrial partners. NUPEC has been conducting these studies on behalf of the Japanese Ministry of International Trade and Industry (MITI).

This paper presents some of results of the advanced LWR core design study on plutonium high consumption MOX cores, and the status of critical experiments for high moderation MOX lattice.

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# 2. ADVANCED LWR CORES FOR EFFECTIVE USE OF PLUTONIUM

#### 2.1. Design Conditions and Target of Study

The study has been performed based on cores of APWR[4] and ABWR[5]. The basic specifications of those plants are shown in TABLE I and these parameters including the power densities of cores have been conserved in this study.

As an index to measure the efficiency of plutonium use, "fissile plutonium consumption rate" was defined as follows:

Fissile Plutonium (Puf) Consumption rate = (Loaded Fissile Plutonium - Discharged Fissile Plutonium) / (Loaded Fissile Plutonium)

The target of the study is to increase this Puf consumption rate. Systematic sensitivity study of the Puf consumption rate has been done for main design features of LWR cores. The results have shown that increase of moderation ratio of the cores is most effective to increase the Puf consumption rate.

# 2.2 High Moderation PWR

Preliminary fuel design study has been performed to increase the core moderation ratio with two methods: (1) reducing the fuel pin diameter and (2) replacing fuel pins by water rods. The influence of

	APWR	ABWR
Rated Thermal Power	4100 MW	3926 MW
Effective Core Height	3.66 m	3.71 m
Operation Cycle Length	15.5 EFPM	15 EFPM
Number of Fuel Assemblies	257	872
Fuel Assembly Type	17x17	9x9
Maximum Burn-up	55 GWd/t	55GWd/t

Table IBasic Design Parameters

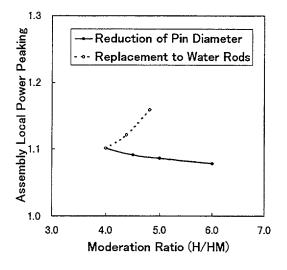
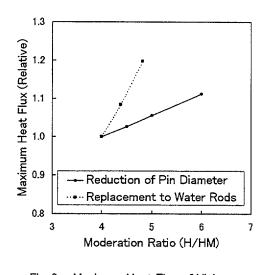
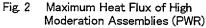


Fig. 1 Local Power Peaking of High Moderation Assemblies (PWR)





these two methods on in-assembly power distribution and thermal margin has been studied with 17 x 17 fuel assembly. Typical results, shown in Figs.1 and 2, show advantage in reducing fuel diameter for increasing moderation ratio in terms of thermal margin.

Thermal hydraulic margin decreases with increase of moderation ratio so that the moderation ratio should be selected properly to keep enough thermal margins. For the one of options to conserve adequate thermal hydraulic margins, a core design of the hydrogen to heavy metal atomic ratio (H/HM) of 5.0 has been studied with fuel rods of a reduced diameter. That H/HM was obtained by reducing the fuel diameter from 9.5 mm to 8.8 mm with the same fuel pin pitch. Full MOX core design with original 17 x 17 fuel assembly of the APWR has been also conducted for the reference of this advanced design. TABLE II shows the specifications of those fuel assemblies. The core performance is shown in TABLE III.

The effect of increasing H/HM is appeared in the necessary fissile plutonium enrichment for the same operation cycle length, which decreases by 1.4 wt % corresponding 19 % reduction. On the other hand, the number of refueling assemblies increased from 88 to 108 due to the decrease of fuel inventory of the core. The maximum assembly burn-up for both cores are less than the limitation (55 GWd/t).

Fig. 3 shows depletion behavior of power distribution in the core. Since full MOX core of PWRs requires no burnable absorber such as gadolinium and applies the same enrichment for all fuel pins, peaking factors of linear heat rate ( $F_Q$ ) and enthalpy rise ( $F_{\Delta H}^{N}$ ) are well suppressed and change

	Reference 17X17 Assembly	Highly Moderated Assembly
Moderation Ratio (H/HM*)	4.0	5.0
Fuel Pin Diameter	9.5 mm	8.8 mm
Assembly Lattice	17X17	17X17 (same pitch)
Number of Thimbles	25	25
Fissile Plutonium Enrichment**	7.2 wt%	5.8 wt%
Matrix	Depleted Uranium	Depleted Uranium
Burnable Absorber		

 Table II
 Specifications of Highly Moderated MOX Assembly (PWR)

\*: Atomic ratio of hydrogen to heavy metal.

\*\*: Enrichment is uniform in the assembly

Table IIHigh Moderation Core Specifications (PWR)

	Reference Core	High Moderation Core
Moderation Ratio (H/HM*)	4.0	5.0
Number of Refueling Assembly	88	108
Operation Cycle Length	15.5 EFPM***	15.5 EFPM***
Cycle Burnup	16.5 GWd/t	19.0 GWd/t
Maximum Assembly Burnup	52.7 GWd/t	53.5 GWd/t

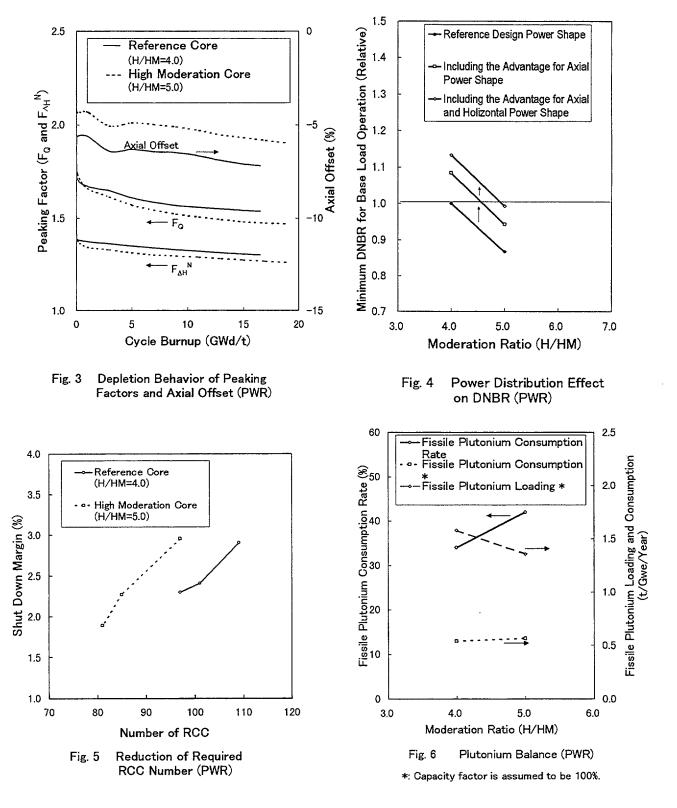
\*: Atomic ratio of hydrogen to heavy metal.

\*\*: Enrichment is uniform in the assembly

\*\*\*: Effective Full Power Months.

smoothly with depletion. The axial power distribution of full MOX cores are slightly shifted to the bottom of the cores compared with typical  $UO_2$  cores. The axial offset of full MOX cores is more negative than typical  $UO_2$  core. Such power distribution characteristics provide additional DNBR margin for the reference core and this high moderation core.

Fig. 4 shows that the DNBR analysis results for the reduced diameter fuel design with practical power shapes, which add excess margin. As the fuel pin diameter is decreased for high moderation ratio, DNBR margin becomes small because of increase of heat flux and decrease of coolant flow speed caused by increase of channel flow area. These analyses show that such DNBR decrease can be



recovered with these power distributions. Since  $F_{\Delta H}^{N}$  of the full MOX cores are 2-3 % less than typical  $UO_2$ , it provides about 5 % additional margin of DNBR. The axial power distribution deference also provides about 10 % additional DNBR margin. Those analyses indicate that this high moderation full MOX core has the same DNBR margin as the reference core.

For the reference full MOX core, the soluble boron worth is about 1/3 of the boron worth of typical UO<sub>2</sub> cores. The one of advantages of the high moderation MOX core is increase of soluble boron worth by 50 %. The control rod worth decrease in the reference full MOX core for the harder neutron spectrum so that it is necessary to add control rods to keep required shut down reactivity margin. The increase of H/HM has the beneficial effect to increase the rod worth and to reduce the additional number of control rods. Fig 5 shows the results of shutdown margin analysis. The required shutdown margin for a full MOX core is roughly estimated to be about 2 %. Then, the results show that the required number of control rod clusters (Ag-In-Cd) for the high moderation core is 85, and is about 12 clusters less than that for the reference core. Using higher worth absorber such as enriched B4C can reduce the required number of the clusters. Fig. 6 shows the fissile plutonium consumption rate and related parameters. With increasing H/HM from 4.0 to 5.0, the consumption rate increases 20 %.

Since these nuclear and thermal hydraulic characteristics of the high moderation core are acceptable, the studied high moderation core is verified to be feasible in APWRs and contributes to effective fissile plutonium consumption.

#### 2.3 High Moderation BWR

The BWR fuel design studies have been done based on the  $9 \times 9$  fuel assembly which was developed as the high burn-up "Step-III" fuel for Japanese BWR plants. Preliminary studies were carried out on the fuel design to increase the moderation ratio by reducing fuel pin diameter and by replacing fuel pins with water rods. Since there is no significant difference between these two designs for the BWR cores, the assembly with additional water rods was selected as the first step of high moderation fuel design for BWRs. The high moderation assembly design of reducing fuel pin diameter will be studied as the next step in this study.

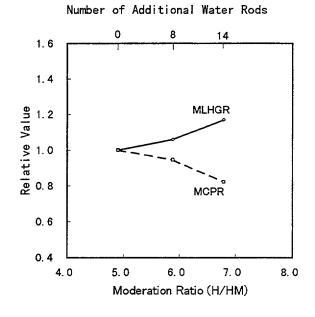


Fig.7 Variation of Maximum Linear Heat Generation Rate (MLHGR) and Minimum Critical Power Ratio (MCPR) with Moderation Ratio (BWR)

	Reference 9X9 Assembly	Highly Moderated Assembly
Moderation Ratio (H/HM*)	4.9	5.9
Number of Additional Water Rods	0	8
Assembly Lattice	9X9	9X9 (same pitch)
Fissile Material Concentration**	5.0 wt%	4.6 wt%
MOX Rod		
Assembly-averaged Enrichment	3.6 wt%	3.3 wt%
Matrix	Depleted Uranium	Depleted Uranium
Gadilinia Rod		
Gadolinia Concentration	2.5/1.5***wt%	3.0∕2.0 <del>***</del> wt%
Matrix	Enriched Uranium	Enriched Uranium

Table IV Specifications of Highly Moderated MOX Assembly (BWR)

\*: Atomic ratio of hydrogen to heavy metal.

\*\*: Assembly-averaged Concentration of (U235+Pu239+Pu241).

\*\*\*: Axial zoning of gadolinia concentration (Upper / Lower).

	Reference Core	High Moderation Core
Moderation Ratio (H/HM*)	4.9	5.9
Fissile Plutonium Enrichment**	3.6 wt%	3.3 wt%
Number of Refueling Assembly	232	264
Operation Cycle Length	15 EFPM <del>**</del> *	15 EFPM***
Cycle Burnup	12.0 GWd/t	13.6 GWd/t
Average Discharge Burnup	45.1 GWd/t	44.9 GWd/t
Maximum Assembly Burnup	49.7 GWd/t	47.3 GWd/t

 Table V
 High Moderation Core Specifications (BWR)

\*: Atomic ratio of hydrogen to heavy metal.

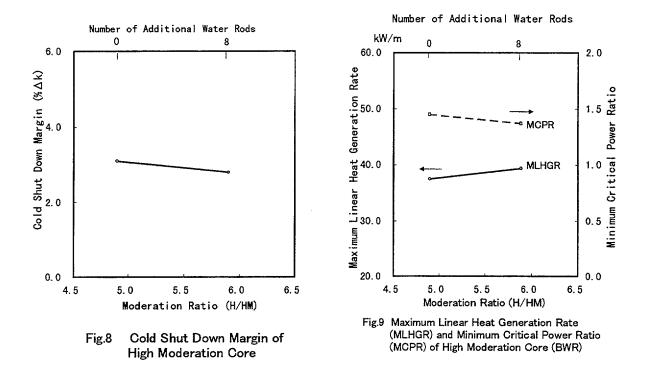
\*\*: Assembly-averaged initial enrichment.

\*\*\*: Effective Full Power Months.

While fissile plutonium consumption rate increases with moderation ratio, the number of additional water rods is limited by thermal margins. The variation of the maximum linear heat generation rate (MLHGR) and the minimum critical power ratio (MCPR) were evaluated with the number of additional water rods. The results are shown in Fig. 7. To maintain adequate thermal margins, the assembly design with 8 additional water rods was adopted as the first option of the high moderation fuel, for which the H/HM value is 5.9, while that of the original 9x9 assembly is 4.9.

The specifications of the fuel assembly are summarized in TABLEIV. The fissile enrichment and burnable poison design were determined to meet the discharge burn-up of 45GWd/t and the operation cycle length of 15 EFPM. Plutonium is blended in depleted uranium and gadolinium in enriched uranium. To simplify the MOX fuel fabrication, MOX fuel pins make use of uniform axial fissile distribution. The axial zoning of the gadolinium concentration was adopted to reduce the axial power peaking.

Under the condition to keep the number and the size of fuel assemblies and control rods in ABWRs, equilibrium core design with high moderation fuel was performed. Full MOX core design with original 9 x 9 fuel assembly of the ABWR has been also conducted for the reference of this advanced design.



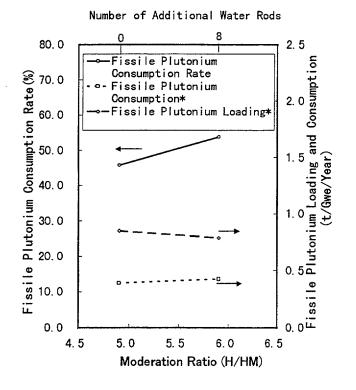


Fig. 10 Plutonium Balance (BWR) \*: Capacity factor is assumed to be 100%

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The specifications of the high moderation core are shown in TABLEV. The fissile plutonium enrichment can be reduced to 3.3 wt% for the high moderation core from 3.6 wt % for the reference core. The number of reload fuel assemblies per an operational cycle increases for high moderation core due to less fuel inventory. The average discharge burn-up is set to 45 GWd/t for both cores. The maximum assembly burn-ups for both cores are less than 55GWd/t, which is the same criteria to "Step-III" UO2 fuel. The nuclear and thermal-hydraulic performances were evaluated with a threedimensional nuclear thermal-hydraulic coupled core simulator. The calculated results on the cold shut down margin (CSDM) are shown in Fig.8. The CSDM values for both MOX cores are about  $3\%\Delta k$ and satisfy the design criteria without any modification on the control rods in the ABWR. The CSDM varies only  $0.3\%\Delta k$  with higher moderation ratio by adding 8 water rods. The evaluated values of MLHGR and MCPR during the equilibrium cycle are presented in Fig. 9. They show the satisfactory results comparing to the operational limits. The relative differences of MLHGR and MCPR between the two MOX cores are about 5%. The fissile plutonium consumption rate and related parameters for both MOX cores were evaluated and the results are presented in Fig. 10. With increasing H/HM from

## 4.9 to 5.9, the consumption rate increases by 20 %.

Since these nuclear and thermal hydraulic characteristics of the high moderation core are acceptable, the studied high moderation core is verified to be feasible in the ABWRs and contributes to effective fissile plutonium consumption.

# 3. MOX CORE PHYSICS EXPERIMENTS

# 3.1. Objectives

The objective of the MISTRAL program is to obtain the core physics parameters systematically in the high moderation MOX cores. The measured data will be analyzed with the core analysis systems and contribute to their verification and improvement.

	Core-1	Core-2	Core-3	Core-4
Volumetric Moderation Ratio	1.8	1.8	2.1	to be defined
Atomic Moderation Ratio	5.11	5.15	6.21	to be defined
Fuel	UO <sub>2</sub> -3.7 %	MOX-7.0 %	MOX-7.0%	MOX-7.0%
Critical Mass	0	0	0	0
Boron Concentration	0	0	0	0
Spectrum Indices	0	0	0	
Radial Power Distribution	0	0	0	0
Axial Power Distribution	0	0	0	0
Iso. Temp. Coef.	0	0	0	
Boron Worth	0	0	0	0
Absorber Worth	0	0	0	
Fuel Substitution	0	0	0	
Cluster Worth				0
2D Void			0	
β <sub>effective</sub>	0	0		

# TABLE VI Core Configurations and Measured Parameters in the MISTRAL Program

# 3.2 Content of program

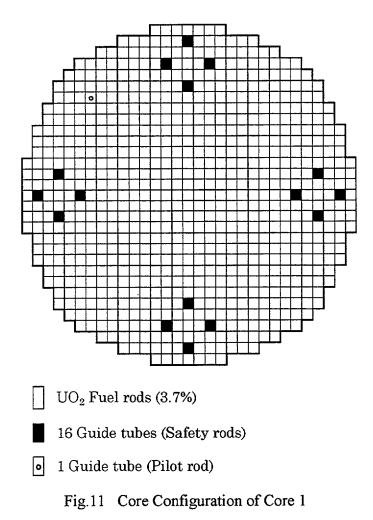
TABLE VI shows the four core configurations and the measurement parameters which have been decided to realize the objective of the program. The core configurations consist of three homogeneous cores and one mock up core. The measurement parameters consist of 1) basic core characteristics such as critical mass, buckling and spectrum indices, 2)reactivity worth of boron diluted in the moderator, absorber rods and water holes, and 3)core safety parameters such as moderator temperature coefficient, void reactivity and effective delayed neutron fraction.

- Core 1: a  $UO_2$  core of homogeneous square lattice with H/HM $\sim$ 5 which is a reference core for MOX cores,
- Core 2: a MOX core of homogeneous square lattice with H/HM~5 which is dedicated to obtain basic characteristics of the MOX core with high moderation,
- Core 3: a MOX core of homogeneous square lattice with H/HM~6 which is dedicated to obtain the effect of increasing moderation ratio,
- Core 4: a mock up core of PWR assembly loaded with MOX fuel

# 3.3 Experiment of Core 1

# 3.3.1 Critical mass

The fuel rods of the Core 1 have almost same specifications of standard 17x17 assemblies of PWR commercial plants except their heights. They consist of pellets of 3.7% enriched uranium and Zry4 fuel claddings. The fuel rods are covered by over claddings of AG3 to adjust the moderattion ratio.



The configuration of the Core 1 is shown in Fig. 11. The Core 1 has about 740 fuel rods located in a rod pitch of 1.32 cm, 16 guide tubes for safety rods and one guide tube for a pilot rod. The core has mirror symmetry of 1/4 except the guide tube of the pilot rod. The boron concentration was about 300 ppm and the core temperature was regulated to be 20 °C with a thermo-regulation system.

### 3.3.2 Fission rate distribution

The core radial and axial fission rate distributions were measured with miniature fission chambers of  $^{237}$ Np and  $^{235}$ U and integral gamma scanning technique of the fuel rods. Examples of radial distribution measured by the gamma scanning are shown in Fig. 12. The values of geometrical buckling, Br<sup>2</sup> and Bz<sup>2</sup>, were derived by the least square fitting of those measured fission rate distributions in the center region with J<sub>0</sub> and cosine functions for the radial and the axial distributions. The geometrical buckling was determined with the uncertainty of about 1 %.

#### 3.3.3 Spectrum index

#### 3.3.3.1 Fission spectrum index

The fission rate measurements with the miniature fission chambers,  ${}^{235}$ U,  ${}^{239}$ Pu,  ${}^{241}$ Pu were carried out to determine the fission spectrum indices,  $\sigma_{f}{}^{239}$ Pu/ $\sigma_{f}{}^{235}$ U,  $\sigma_{f}{}^{241}$ Pu/ $\sigma_{f}{}^{239}$ Pu. The derivation of the fission spectrum indices for fissile nuclides is shown in the following formula:

$$\frac{\overline{\mathrm{o}} f_{j}^{M1}}{\overline{\mathrm{o}} f_{i}^{M1}} = \left(\frac{\frac{C_{j}^{M1}}{C_{i}^{M1}}}{\frac{C_{j}^{Th}}{C_{i}^{Th}}}\right) \times \frac{\overline{\mathrm{o}} f_{j}^{Th}}{\overline{\mathrm{o}} f_{i}^{Th}}.$$

The fission spectrum index of the nuclide "j" to the nuclide "i" in the Core 1(the left side of the formula) is related to that in the thermal column(the second term of the right side of the formula) through the ratio of the counting rate ratios normalized by reactor power(the first term of the right side of the formula). In the derivation of the indices, necessary corrections were applied for the impurities and the decay of nuclides in the fissile deposit in the fission chambers.

#### 3.3.3.2 Modified conversion factor

The modified conversion factor defined as the ratio between capture of <sup>238</sup>U and total fission in the fuel rod was derived by measuring specific gamma rays from the fuel rod[6]. The measured gamma-rays are 293.27 keV of <sup>143</sup>Ce, 537.31 keV of <sup>140</sup>Ba in the decay of fission products and 277.60 keV of <sup>239</sup>Np in the chain of <sup>238</sup>U+n→<sup>239</sup>U→<sup>239</sup>Np( $\beta$ <sup>2.355days)→<sup>239</sup>Pu. A high purity Ge detector</sup>

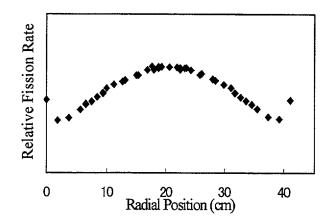


Fig. 12 Measured Radial Fission Distribution

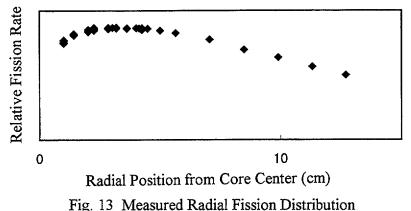


Fig. 13 Measured Radial Fission Distribution

used in the gamma ray measurement has the energy resolution of 0.9 keV for the gamma-rays less than 600 keV. The self-shielding effect of gamma-rays in the fuel was calculated with the Monte Carlo code, MCNP in a 3D geometry. The experimental uncertainty was about 3 % which was synthesized from each uncertainty of the self-shielding effect, the fission yields, the counting statistics of the photo peaks and the detector efficiencies.

# 3.3.4 Temperature coefficient

#### 3.3.4.1 Reactivity measurement

Criticality was reached at nine temperature points, 10, 20, 30, 40, 60, 65, 70, 75, and 80 °C with necessary compensation of reactivity by dilution of boron in the moderator, and the concentrations of boron and residual reactivity was measured by the reactor periods. The difference of reactivity between the two states of adjoining temperatures was derived with using the boron deferential coefficients and the relation between reactivity and reactor period (the inhour equation).

#### 3.3.4.2 Fission rate distribution

The core radial and axial fission rate distributions were measured at 65 °C and 80 °C with the gamma scanning of the fuel rods. From these distributions, the values of geometrical buckling were deduced.

#### 3.3.5 Differential boron efficiency

To obtain the differential boron efficiency, the reactivity measurements ware performed in the two core states with slightly different boron concentrations from that of critical core configuration at 20 °C. The reactivity were derived by the measurement of the reactor period. The differential boron efficiency was determined with the uncertainty of about 10 %.

#### 3.3.6 Absorber worth

One fuel rod in the center of the core was replaced by each absorber rod,  $UO_2$ -Gd<sub>2</sub>O<sub>3</sub>, Ag-In-Cd, natural B<sub>4</sub>C and enriched B<sub>4</sub>C. The reactivity change caused by the insertion of the absorber rod was compensated by the dilution of boron in the moderator to reach criticality and the residual reactivity was measured by the reactor period. The absorber worth was determined with the uncertainty less than 3 %. The radial fission rate distributions were also measured with the gamma scanning of the fuel rods. Fig. 13 shows measured fission rate distribution for the natural B<sub>4</sub>C.

#### 3.3.7 Water hole worth

Nine fuel rods (3x3) in the center of the core were withdrawn to be replaced by a large water hole. The reactivity change caused by this was compensated by the dilution of boron in the moderator to reach criticality and residual reactivity was measured by the reactor period. The worth of the water hole was determined with the uncertainty of about 7 %. The radial fission rate distributions were also measured with the gamma scanning of the fuel rods.

### **3.4 EXPERIMENT OF CORE 2**

The experiment of the Core 2 started in February, 1997. The loading of the MOX fuel rods was completed in the middle of February and the first critical was reached on February 21. The Core 2 in the critical mass measurement consists of about 1570 MOX fuel rods of 7 % Pu (total) and about 20 MOX fuel rods of 8.7 % Pu (total) located in a rod pitch of 1.32 cm, 16 guide tubes for the safety rods, and one guide tube for the pilot rod. The core has mirror symmetry of 1/4 except the guide tube for the pilot rod. The moderator was zero. The measurements following the critical measurements are progressing now and will be completed in May, 1998.

#### 4. CONCLUSION

A study have been performed to develop advanced technologies for full MOX cores with high Pu consumption rate based on APWRs and ABWRs. For this aim, basic core designs of high moderation (H/HM  $\sim$ 5) have been carried for APWRs with reduced fuel diameters in fuel assemblies and those of high moderation (H/HM  $\sim$ 6) for ABWRs with addition of extra water rods in fuel assemblies. The analysis of equilibrium cores shows that nuclear and thermal hydraulic parameters satisfy the design criteria and the Pu consumption rate increases by about 20 %.

The MISTRAL program was planned to obtain the main core physics parameters of the high moderation MOX cores. The program is scheduled to be carried out from 1996 to 2000. The experiment of the Core 1 has been completed by the end of November 1996 and that of Core 2 is ongoing now.

#### ACKNOWLEDGEMENTS

The authors thank Prof. Oka of Tokyo Univ., and other members of the implementation committee of "Study of Advanced LWR Cores for Effective Use of Plutonium," in NUPEC for their technical advice and suggestions.

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# **BILATERAL MOX PROJECTS FOR W-GRADE Pu DISPOSITION: SAFETY IMPLICATIONS**

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#### Abstract

Beginning from 1992 a number of studies were performed in Russia in the field of peaceful utilization of the excess Weapon-derived Plutonium (W-Pu). Very important results were obtained from Russian-German, Russian-French, Russian-Canadian Feasibility Studies and from the US-Russian Projects. These Studies have demonstrated technical feasibility of W-Pu conversion into MOX-fuel and its utilization in certain power reactors. Technical feasibility implies safety assurance for all stages of W-Pu management.

All safety-related aspects (e.g. reactor safety, nuclear criticality, fire and explosion safety, protective barriers reliability, etc.) have to be taken into account for implementation of W-Pu utilization through MOX-route. Safety has to be guarantied for such W-Pu-related activities as:

- Plutonium interim storage;
- Plutonium conversion from metal to oxide;
- MOX fuel fabrication and transportation;
- MOX irradiation in power reactor;
- MOX spent fuel management;
- Pu-bearing waste minimization and waste management;
- Radiological protection of the personnel and environment.

Paper addresses to safety-related issues of W-Pu conversion into oxide form and MOX- fuel fabrication. Some results of Russian-German and Russian-French (AIDA-MOX 1) Studies are discussed.

# 1. W-Pu conversion safety

Two primary objectives for conversion of metal Pu into oxide are:

- production of plutonium oxide suitable for MOX-fuel fabrication (sinterable oxide);
- removing of impurities (primarily Am and Ga), which could influence MOX-fuel manufacture.

Different technologies could be used for this purpose. As example, two of them are being developed in All-Russian Institute of Inorganic Materials (Moscow) and in Institute of Nuclear Reactors (Dimitrovgrad).

First process includes direct dissolution of metal Pu in HNO<sub>3</sub>+HF with further Pu purification by liquid extraction. Pu oxide could be produced through oxalate or hydroxide precipitation. The process is complicated by secondary waste management. Nuclear criticality is one of the most important issue in this case. It means strict control of W-Pu metal mass to be dissolved, limitation of Pu concentration in water solutions, use of equipment with nuclear safe geometry, etc. Nevertheless this process offers the best warranties of safety. It is based on pilot-scale experience and could be implemented without extensive R&D.

Second process is based on pyrometallurgy with direct dissolution of metal Pu in molten salt media (NaCl+KCl) and chemical precipitation of compact Pu oxide (or electrodeposition of  $UO_2$ +PuO<sub>2</sub>). This oxide is not suitable directly for MOX fuel fabrication and needs additional treatment. The process seems to generate relatively small amount of secondary wastes, but it was

not proven at industrial scale. Implementation of this technology needs further extensive R&D. Nuclear safety is not a major issue for this case.

Dry process like HYDOX process was also examined in AIDA-MOX 1 Study and finally has been turned down in view of additional work needed for qualification. Operational safety of the oxidation of plutonium hydride into oxide will be one of the most important issues in this technology. Another challenge is volatile Ga oxide waste formed during high temperature treatment of the initial plutonium dioxide.

# 2. Safety issues for MOX Fuel Fabrication from W-Pu

Different safety-related issues of MOX fabrication were considered under bilateral feasibility studies mentioned above [1, 2, 3]. Generally speaking the safety provisions and safety related design characteristics should be similar for MOX-fuel fabrication from civil as well as from weapon-derived plutonium. Some general safety issues of MOX fuel production are covered in NEA/OECD Reports [4,5].

To prevent risk of plutonium release a multi-barrier protective system is designed for MOX fabrication facility. A graduated low air pressure supports the system to avoid Pu contamination outside enclosure, which is provided by glove boxes and caissons or shielded hot cells. All initiating events (internal and external) which could bring the possibility to damage the multi-barrier system are to be taken into account at the design stage. Working personnel have to be protected from ionizing radiation. Doses to personnel are to be well under authorized limits.

Within the framework of Russian-German Feasibility Study experts from both sides developed safety criteria for MOX Pilot Plant designated for W-Pu processing into MOX fuel. Based on technical information from German side and Russian regulatory documents experts developed "Safety Concept for Pilot Plant for Uranium-Plutonium Fuel Manufacture from Weapons-Derived Plutonium". This document was approved by Russian Supervision Authorities (Ministry of Health and Gosatomnadzor) and by Russian Minatom. It was found that German equipment designed for the new Hanau MOX Plant offers unique opportunity to process W-Pu due to the fact that nuclear safety is ensured for PuO<sub>2</sub> with isotopic composition of 95% Pu-239 and 5% of Pu-240.

Low Pu-238 and Am-241 content in W-Pu make it possible to minimize radiation dozes for personnel (5 mSv per year) which is well below authorized limit of 20 mSv/year. This limit is established by Russian Radiation Safety Standard NRB-96. 4 stages of HEPA (high efficiency particulate) filters prevent any environmental effect for normal operation of the plant and for the design-based events.

For the MOX Pilot Plant preliminary design as well as for TOMOX-1300 Facility special containment building was considered as a necessary protection from external hazard such as airplane crash [1,2].

Design of VVER-1000 fuel is different from Western PWR Fuel Assembly designs. It means that a special equipment is to be developed for MOX-fuel rods testing and mounting in fuel assemblies for the VVER MOX-fabrication facility.

One of important issues is safe transportation of MOX fuel from MOX fabrication facility to the designated reactor site. For VVER MOX fuel transportation within Russian Federation a special transport container is to be designed. Extensive European experience in MOX-fuel transportation is of great value for that case. In bilateral Russian-Canadian Feasibility Study for overseas transportation of MOX CANDU fuel with W-Pu it was proposed special new safe package with 18 MOX fuel bundles capacity.

# **Conclusions:**

- 1. Nuclear and radiological safety are of major importance for all the stages of W-Pu utilization. Licensing of any Pu-disposition Project in Russian Federation implies in-depth study of all important safety issues.
- 2. All safety-related aspects of W-Pu conversion and MOX fuel fabrication (nuclear criticality, fire and explosion safety, protective barriers reliability, MOX-transportation, Pu-waste management, etc.) have to be taken into account for implementation of W-Pu utilization through MOX-route.
- 3. Accumulated experience and internationally adopted safety philosophy for civil Pu management are of great importance for future steps connected with W-Pu utilization.

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# **BILATERAL COOPERATION:**

MINATOM-CEA	MINATOM – GRS/SIEMENS
Intergovernmental Agreement -	Intergovernmental Agreement -
November 12, 1992	December 16, 1992
AIDA-MOX 1 PROGRAM:	Feasibility Study - First Phase: 1994
1993 -1997	Second Phase : May 1995 – Dec. 1996
Final Report on Feasibility	Final Report (in German and
Studies	Russian) – 1997
(in French, Russian and English)	Safety Concept for MOX-
– 1997	Fabrication Facility

1998-99 : Trilateral Russian-French-German Intergovernmental Agreement (to be signed):

- 1. Reactor Core calculations & Reactor Safety Studies for VVER-1000 and BN-600 with MOX-Fuel (MINATOM – GRS/ CEA/FRAMATOME)
- 2. Joint MINATOM-SIEMENS-COGEMA Basic Design of the MOX-fabrication Facility (DEMOX-facility) (1300 kg of W-Pu/year)
- 3. Study and Design of W-Pu Conversion Facility (MINATOM-CEA)
- 4. Demonstration Program of VVER-1000 MOX-fuel irradiation

# Russia-US and Russia-Canada Studies on W-Pu utilization

MINATOM - US DOE	MINATOM – AECL
<ul> <li>First Joint Study – 1995-1996</li> <li>Final Report issued in October 1996</li> </ul>	mid 1996 – Jan 1997: • First MOX-CANDU Feasibility Study
<ul> <li>1997-1998: Joint Studies on</li> <li>W-Pu Conversion</li> <li>Safety issues of W-Pu geologic disposal</li> <li>Reactor Safety with MOX – BN-600/BN-800</li> <li>VVER-1000 (from 1998)</li> <li>W-Pu Waste Solidification</li> </ul>	<ul> <li>1997:</li> <li>Second Feasibility Study DEMOX- 1300 + CANDU-MOX Line</li> <li>(Economics for Different MOX- Production Rate and CANDU Fuels)</li> </ul>
Bilateral Inter government Agreement is under preparation	Bilateral Inter government Agreement is signed in April 1996

**Parallex Project – Started September 1997** 

Fabrication of MOX-Fuel in Bochvar Inst. (MINATOM) and LANL (US DOE) for Irradiation in Canadian NRU Reactor

# INERT MATRIX (URANIUM-FREE) FUEL FOR ACTINIDE BURNING AND PLUTONIUM ANNIHILATION

(Session 5)

Chairperson

A. Stanculescu Switzerland





# SWISS R&D ON URANIUM-FREE LWR FUELS FOR PLUTONIUM INCINERATION

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#### Abstract

The most efficient way to enhance the plutonium consumption in LWRs is to eliminate plutonium production altogether. This requirement leads to fuel concepts in which the uranium is replaced by an inert matrix. The inert matrix material studied at PSI is zirconium oxide. For reactivity control reasons, adding a burnable poison to this fuel proves to be necessary. The studies performed at PSI have identified erbium oxide as the most suitable candidate for this purpose. With regard to material technology aspects, efforts have concentrated on the evaluation of fabrication feasibility and on the determination of the physicochemical properties of the chosen single phase zirconium/ erbium/plutonium oxide material stabilised as a cubic solution by yttrium. The results to-date, obtained for inert matrix samples containing thorium or cerium as plutonium substitute, confirm the robustness and stability of this material. With regard to reactor physics aspects, our studies indicate the feasibility of uranium-free, plutonium-fuelled cores having operational characteristics quite similar to those of conventional UO2-fuelled ones, and much higher plutonium consumption rates, as compared to 100% MOX loadings. The safety features of such cores, based on results obtained from static neutronics calculations, show no cliff edges. However, the need for further detailed transient analyses is clearly recognised. Summarising, PSI's studies indicate the feasibility of a uranium-free plutonium fuel to be considered in "maximum plutonium consumption LWRs" operating in a "oncethrough" mode. With regard to reactor physics, future efforts will concentrate on strengthening the safety case of uranium-free cores, as well as on improving the integral data base for validation of the neutronics calculations. Material technology studies will be continued to investigate the physicochemical properties of the inert matrix fuel containing plutonium and will focus on the planning and evaluation of results from appropriate irradiation experiments.

#### 1. INTRODUCTION

The Swiss R&D contributions on uranium-free LWR fuels are embedded within the framework of PSI's research efforts on advanced fuel cycles. Apart from the need to ensure analytical and experimental support on plutonium recycle issues to the utilities as well as to various federal agencies, PSI's efforts aim at strengthening the sustainability arguments for nuclear power as a very-close-to-zero-CO<sub>2</sub> emission primary energy source for electricity generation.

The strategy presently adopted by some utilities to deal with ever increasing plutonium inventories accumulating in the spent fuel generated by their nuclear power plants corresponds to the socalled "self-generated mode" which consists in reprocessing the uranium based LWR fuel, fabricating MOX fuel assemblies, and using these to load up to 1/3 of the core. This strategy is firmly based on industrial processes, but it permits only to stabilise the plutonium mass flow (over the life span of a core loading, as much plutonium is consumed as is produced through neutron captures in <sup>238</sup>U). To go beyond the "self-generated mode", full MOX core loadings are envisaged for advanced LWR core designs. A much more efficient way, however, to enhance the plutonium consumption in LWRs would be to eliminate plutonium production altogether. This requirement leads to fuel concepts in

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which the uranium is replaced by an inert matrix. The inert matrix fuel (IMF) studied at PSI is based on zirconium oxide Ref. [1-2]. This material is a promising inert matrix candidate due to the fact that it is stabilised by rare earth oxides in a single phase solid solution with a cubic structure. By the addition of yttrium, the zirconium oxide based inert matrix material becomes comparable to  $UO_2$  in MOX fuel, thus offering the sought after properties for hosting the plutonium, the other actinides, and the fission products. For reactivity control reasons, adding a burnable poison to this fuel proves to be necessary. The studies performed at PSI have identified erbium oxide as the most suitable candidate for this purpose.

Within the framework of the reactor physics studies, the main objectives have been to investigate the operational and safety-related parameters of uranium-free, plutonium-fuelled LWR cores, to identify and implement the minimum design modifications permitting a smooth transition from present-day  $UO_2$  and  $UO_2$ /MOX loadings to such plutonium-incinerating cores, and, last but not least, to strengthen the validation basis for the data and calculational methods employed.

With regard to material technology aspects, efforts have concentrated on the evaluation of fabrication feasibility and on the determination of the physicochemical properties of the chosen single phase zirconium/erbium/plutonium oxide material stabilised as a cubic solution by yttrium.

The present paper summarizes the results obtained to-date with respect to both material technology and reactor physics aspects of PSI's IMF studies.

# 2. FUEL MATRIX

Zirconium oxide is a promising candidate as inert matrix because it may be stabilised by rare earth oxides in a single phase solid solution which offers attractive properties for a nuclear fuel. In this material, rare earth oxides stabilise the phase in a cubic structure, and the stabilised zirconia is then comparable to  $UO_2$  in MOX fuel. In the suggested fuel material, the stabilised zirconia fluoritetype phase will be the host phase for plutonium, other actinide elements or fission products. Binary cubic mixtures  $ZrO_2$ -YO<sub>1.5</sub> form solid solutions with numerous dopants from room temperature to about 3000 K [3]. Little is known on the properties of zirconia based ceramics, and on their behaviour under irradiation. Some early studies reported data on zirconia lattice expansion [4], thermal conductivity changes [5], and mechanical property changes [6]. It was also observed that during neutron irradiation of uranium doped zirconia, the phase transforms to cubic when the neutron fluence increases [7]. More recently, Clinard [8] studied the behaviour of stabilised zirconia under neutron irradiation and showed the presence of ordered inclusion arrays. All of this information indicates that these zirconia based materials have a relatively stable behaviour under irradiation.

For estimating the in-pile behaviour of zirconia based fuel, the behaviour of both inert matrix and simulated fuel materials is being studied under specific types of irradiation. Since fission products such as Xe are known to cause considerable material damage, the stability of these ceramic materials has been assessed under Xe irradiation utilising both high and low energy particles.

#### 2.1 Material Preparation and Analysis

The selected route for material preparation is aqueous co-precipitation of the nitrate salts solution mixture by ammonia. This wet preparation method was adapted for the fabrication of a simulated fuel material  $(Zr_{0.95-x-y}Y_xEr_{0.05}M_y)O_{1.975-x/2}$  with M = Ce, U or Th as analogue of Pu (x= 0.10-0.15, y= 0.07-0.10). Subsequent drying, crushing, pelletising and sintering at 1875 K was carried out for pellet preparation. The relative density of these material samples was about 95%. X-ray diffraction was used for phase interpretation and determination of second phase formation. Fig. 1 illustrates that the quaternary material forms a single solid solution. Lattice parameter measurements where applied for characterisation and the determination of theoretical densities. Specific samples for measurements were obtained by cutting the pellet using a diamond saw.

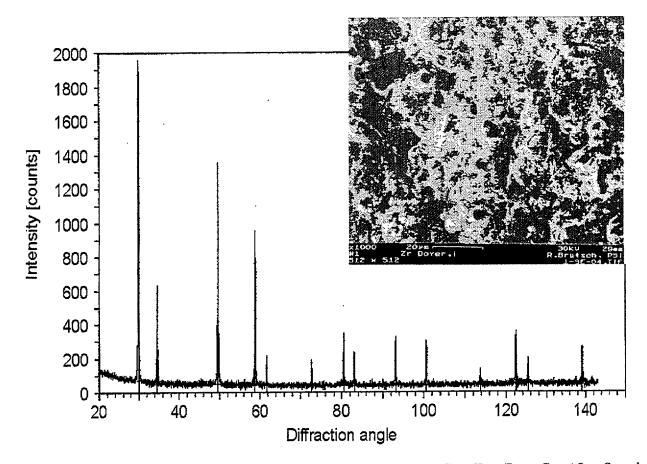


FIG. 1. XRD Spectrum and Structure of Pellet (SEM Micrograph) for a  $(Zr_{0.7}Y_{0.15}Er_{0.05}Ce_{0.1})O_{1.9}$  Sample

#### 2.2 Thermal Conductivity Analysis

Thermal conductivities of zirconia based inert matrix and analogous fuel materials were measured and modelled [9]. Measurements were performed using the laser flash method and systematically applied to binary, ternary and quaternary systems including zirconia, yttria, erbia and ceria or thoria. Measurements were carried out from room temperature up to 1300 K. Thermal diffusivity results for simulated fuels are given in Fig. 2 and compared to non-irradiated and irradiated  $UO_2$ . Thermal conductivity was calculated using theoretical values for specific heat capacity and was also modelled taking into account the effect of dopants on the lattice parameter of the cubic solid solution and the oxygen vacancy size and concentration [9]. Experimental and lattice parameter values are compared prior to full justification of the results. In the temperature range from 300 to 1000 K, the thermal conductivity of the single phase solid solution with yttria, erbia and ceria as analogues of fuel material was confirmed to be about 2 W·m<sup>-1</sup>·K<sup>-1</sup>, a value similar to stabilised zirconium oxide with similar dopant concentrations.

In addition, inert matrix sample conductivity was measured up to 2200 K. It was striking to note the large conductivity increase around 1500 K, reaching 3 W·m<sup>-1</sup>·K<sup>-1</sup> at 2200 K. These samples presented, however, lower conductivities at room temperature (about 1.5 W·m<sup>-1</sup>·K<sup>-1</sup>). Recently, comparable thermal conductivity values were obtained by oscillating scanning differential calorimetry for the same material (Fig. 3). Energy transfer in this transparent matrix must, however, be discussed on the basis of both photonic and phononic conductivities. Based on the phononic conductivity, an annular pellet design to reduce the centre line temperature has been proposed [2].

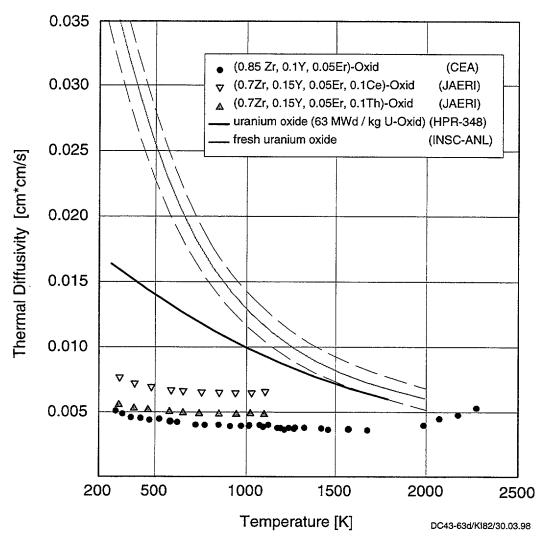


FIG. 2. Comparison of Thermal Diffusivity of Stabilized Zirconium Oxide and simulated IMF with Uranium Oxide, and with High Burn-up Uranium Fuel

#### 2.3. Behaviour under Irradiation

The inert matrix and simulated fuel materials were irradiated using an analytical electron microscope with a low energy Xe injector at the Japanese Atomic Energy Research Institute (JAERI) and using the HVEM-Tandem Facility at Argonne National Laboratory (ANL).

For electron microscopy, trepanning disks of 3 mm diameter with a thickness of 10-20 mm in the centre part were made by ultrasonic cutting and dimpling. Perforation was made by thinning with a 3 keV Ar<sup>+</sup> ion beam. With the electron microscope, amorphization and defect clusters due to ion thinning were not observed on the inert matrix samples. In all samples cubic solid solutions were determined. One as fabricated sample (with thoria included) presented locally amorphous phases (about 10%) produced in the tri-join boundary spaces.

Observations and irradiation experiments for  $(Zr_{0.85}Y_{0.10}Er_{0.05})O_{1.925}$  with and without 10% CeO<sub>2</sub> at JAERI were performed in a JEM electron microscope equipped with an ion accelerator. The beam of 60 keV Xe ions provided a flux of  $5 \cdot 10^{12}$  Xe·cm<sup>-2</sup>·s<sup>-1</sup>. The irradiations were carried out at room temperature and at 925 K accumulating relatively large doses, simulating fission damage under light water reactor conditions (Fig. 4).

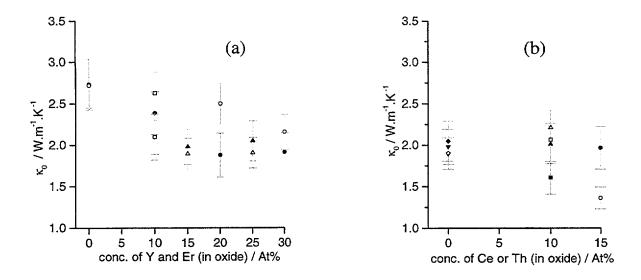


FIG. 3. Comparison of Thermal Conductivity Values. Determined by Oscillating Differential Scanning Calorimetry (solid symbols) and Laser Flash Methode (open symbols)

- Ternary systems:  $(Zr_{1-x-y}, Y_x, Er_y)O_{2-(x+y)/2}$  with an erbia part of (At%): •: 0,  $\blacksquare$ : 5,  $\clubsuit$ : 10. (a):
- $Quaternary \; systems: \; (Zr_{1-x-y-z}, Y_x, Er_y, Ce_z)O_{2-(x+y)/2} \; and \; (Zr_{1-x-y-z}, Y_x, Er_y, Th_z)O_{2-(x+y)/2}.$ (b):  $\checkmark$ : (Zr<sub>0.70</sub>, Y<sub>0.10</sub>, Er<sub>0.05</sub>)O<sub>1.925</sub>, The different material compositions were:
  - $\bullet: (Zr_{0.70}, Y_{0.20}, Er_{0.05})O_{1.875},$
  - $\blacksquare$ : (Zr<sub>0.70</sub>, Y<sub>0.15</sub>, Er<sub>0.05</sub>, Th<sub>0.10</sub>)O<sub>1.900</sub>.
- $\uparrow$ : (Zr<sub>0.70</sub>, Y<sub>0.15</sub>, Er<sub>0.05</sub>, Ce<sub>0.10</sub>)O<sub>1.900</sub>,
- •:  $(Zr_{0.68}, Y_{0.1}, Er_{0.07}, Ce_{0.15})O_{1.915}$ .

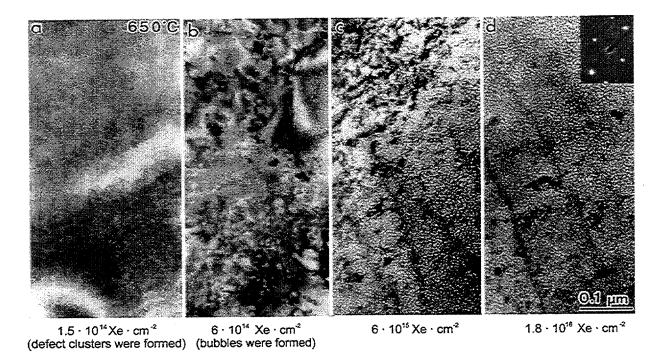


FIG. 4. Process of Formation and Growth of Defect and Bubbles in  $(Zr_{0.85}Y_{0.10}Er_{0.05})O_{1.925}$  During Irradiation with 60 keV Xe Ions of  $5 \cdot 10^{12} \text{Xe} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$  at  $650^{\circ} \text{C}$ .

The thickness of the sample observed with an electron microscope was about 50 nm. Irradiation performed at the two temperatures demonstrated that for a dose of  $1.8 \ 10^{16} \text{Xe} \cdot \text{cm}^{-2}$ , no amorphization was observed. The penetration depth of the Xe ions in the inert matrix irradiated with 60 keV Xe ion was estimated to be about 15 nm by the TRIM-95 code.

After ion irradiation, the volume swelling of the specimens was estimated from the total bubble volume which was measured from bubble densities and average bubble radius. The volume swelling was estimated to be about 0.19 % at room temperature and about 0.72 % at 925 K, respectively.

Simulated fuel samples  $(Zr_{0.75}Y_{0.10}Er_{0.05}Th_{0.10})O_{1.925})$  were prepared to investigate the microscopic behaviour of the material under high energy irradiation (1.5 MeV). TEM sub-samples were prepared by mechanical grinding and ion milling at the University of New Mexico. The thickness of those samples was of the order of 100 nm and the irradiation took place in the HVEM-Tandem Facility. The material was irradiated with an ion dose rate of  $3.4 \cdot 10^{12} Xe \cdot cm^{-2}s^{-1}$ , to doses of  $2.0 \cdot 10^{16} Xe \cdot cm^{-2}$  with high energy Xe ions. Irradiation was carried out at 20 K with a liquid helium stage, because previous experiences on  $ZrO_2$  have shown that the phase is very resistant to amorphization, but that amorphization might occur at sufficiently low temperatures at which defect recovery could be suppressed. Some observations were carried on-line (Fig. 5).

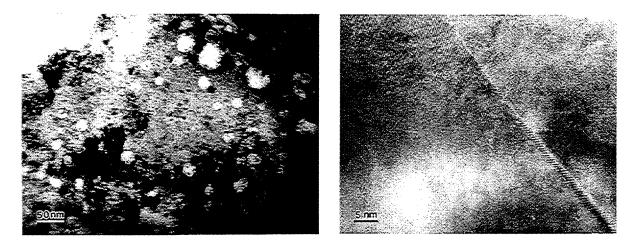


FIG. 5. Examination of Defects on a  $(Zr_{0.75}Y_{0.\ 10}Er_{0.05}Th_{0.10})O_{1.925}$  Sample during Irradiation with 1.5 MeV Xe Ions of  $2 \cdot 10^{16}$  Xe·cm<sup>-2</sup> at 20 K Courtesy Prof. Ewing, Uni New Mexico

At a fluence of  $2 \cdot 10^{16}$  Xe·cm<sup>-2</sup>, a high density of dislocation loops was observed, and the size of the loops ranged from 20 to 60 nm. Detailed TEM analysis after ion irradiation did not reveal any amorphization even near the grain boundaries. According to a full cascade TRIM-95 calculation (using  $E_d = 60$  eV), the damage in the middle of the electron transparent TEM foil (100 nm in depth) reached 25 dpa (displacement per atom) at full ion dose.

The results of all examinations indicate that the irradiation temperature (20 K) is still too high to destroy the crystal structure of stabilised zirconia [10]. Compared to the stability of  $UO_2$  under energetic irradiation conditions [11], the proposed IMF seems to be much more stable.

#### 3. FUEL ASSEMBLY DESIGN

Based on scoping cell calculations [1] addressing the basic reactor physics characteristics (e.g. depletion dependent reactivity variation, temperature and void coefficients, reactivity control) of IMF, a promising candidate was identified, with  $ZrO_2$  being the inert matrix in which the  $PuO_2$  fuel is embedded, and  $Er_2O_3$  the burnable poison. The plutonium isotopic composition considered corresponds to that obtained from LWR UO<sub>2</sub> fuel with a burnup of approximately 40 GWd/t and five years cooling time. This IMF was included, among others, in an international benchmark exercise aiming at data and methods validation [12]. Apart from PSI, participating with results based on the code BOXER using JEF-1 data, the other participants were CEA (APOLLO-2/JEF-2.2), ECN (SCALE/JEF-2.2), JAERI (SRAC-95/JENDL-3.2) and POLIMI (WIMS/WIMS-86). The main results with regard to the IMF studied at PSI can be summarized as follows:

- k-infinity results at beginning of life are in good agreement, the standard deviation with respect to all participants' average value being 400 pcm. The uncertainties observed at beginning of life are due to the different cross section libraries. At the end of life, the discrepancies are larger, with the standard deviation reaching 1000 pcm, and are mainly due to the different methods;
- the results obtained for the fuel temperature coefficients show the same trend as for k-infinity: satisfactory agreement at beginning of life, and larger discrepancies at end of life;
- there are significant discrepancies in the results obtained for the void coefficients if the void fraction exceeds 90%. These are mainly due to differences in the zirconium resonance cross sections.

The overall conclusion from these benchmark studies to-date is that there remain some physics characteristics of inert matrix fuels requiring further clarification (mainly with regard to the fuel temperature coefficients and to the void coefficients for void fractions larger than 90 %).

In a second step, whole-core three-dimensional neutronics analyses were performed [13-14] to assess the operational, safety-related, and - last but not least - plutonium consumption characteristics of present-day PWR core designs fuelled with the aforementioned uranium-free fuel. Within the scope of these studies (i.e. equilibrium cycle situations, homogeneous uranium-free assembly design), the results obtained have been very encouraging. They indicate, for a present-day 1 GWe PWR, the feasibility of a 100% uranium-free fuelled four-region core with a cycle length of over 300 efpd. Its plutonium consumption capabilities (in terms of total plutonium) are approximately twice as high as in the case of 100% MOX-fuelled cores, i.e. 60% of the initial plutonium inventory. At the same time, the operational characteristics (e.g. reactivity variation with burnup, and power peaking) of the uranium-free cores are very similar to those of conventional present-day UO<sub>2</sub> fuelled ones. A discussion of the safety-related parameters, on the other hand, asks for a more discerning approach. While the fuel temperature feedback reactivity effect (Doppler coefficient) in the uranium-free core is approximately half the value (in absolute terms) of that for the conventional UO<sub>2</sub>-core, the moderator temperature coefficient is very similar, and the shutdown margin (reactivity margin at hot zero-power conditions) is even larger - always when comparing the uranium-free with the conventional UO<sub>2</sub>-core. The bottom line to date is that, although no cliff edges are expected based on the results for safetyrelated parameters obtained from static neutronics analyses, detailed transient studies are necessary to strengthen the safety case for the uranium-free cores.

Finally, results are presented summarising the main findings of the third step in PSI's physics design efforts towards the goal of enhanced plutonium consumption in present-day PWRs fuelled with uranium-free plutonium fuel. In these studies, we have looked at "real-life situations" which ask for "transition configurations" in which loadings with mixed  $UO_2$  and uranium-free assemblies must be considered.

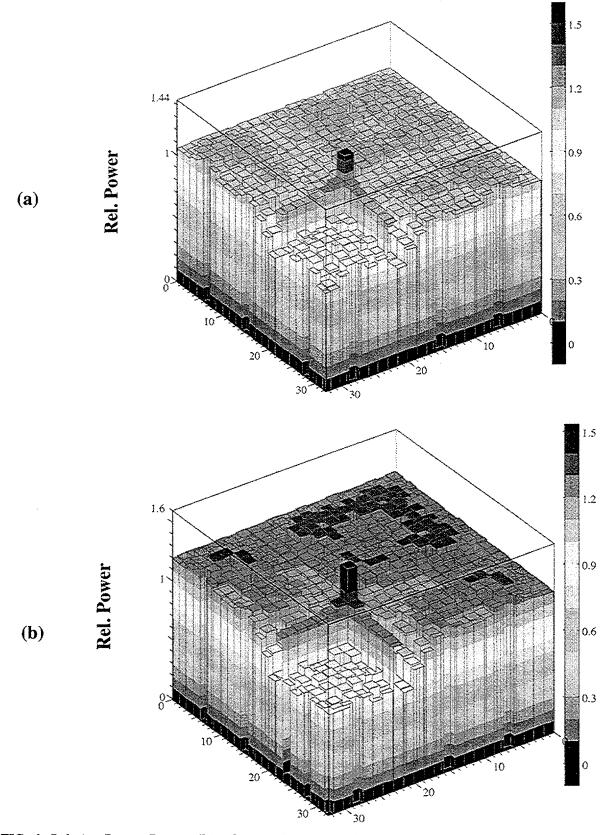
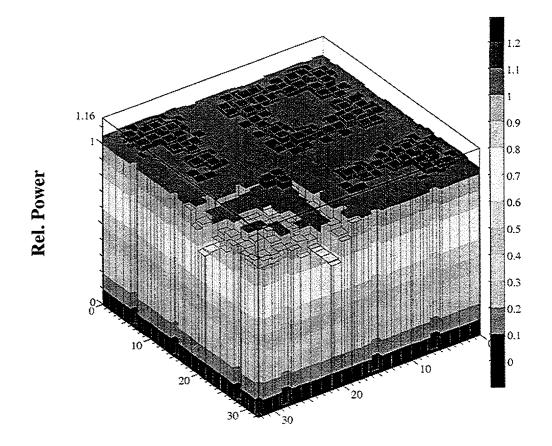


FIG. 6. Relative Power Density Distributions for a (a) MOX and (b) Uranium-Free Plutonium Fuel Assembly Surrounded by Standard Uranium oxide fuel.



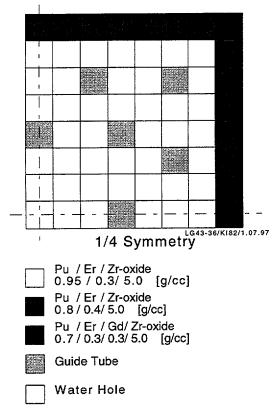


FIG. 7. Optimised Design and Related Power Density Distribution for a Uranium-Free Plutonium Fuel Assembly Surrounded by Standard Uranium Oxide Fuel

The problems faced when introducing uranium-free assemblies into a  $UO_2$ -assembly environment are similar to those encountered in the case of MOX fuel [15]. Fig. 6 shows the distribution of the relative power densities for both a MOX (Fig. 6 (a)), and a uranium-free fuel assembly (Fig. 6 (b)). In both cases, the special assemblies are homogeneous and surrounded by standard  $UO_2$ fuel. It is worthwhile mentioning that, with the exception of the outermost corner pin (for which the spectral effect of the neighbouring  $UO_2$ -elements is extreme), the spectral impact of the uranium zone on the plutonium bearing assembly is stronger in the case of the MOX fuel assembly than in the case of the uranium-free one.

As a matter of fact, the spectral effects are "felt" well into the MOX assembly, while, in the uranium-free case, they hardly influence the power distribution beyond the first pin row. This may be explained by the fact that, on the one hand, the plutonium content in the uranium-free fuel assembly is approximately 50% higher than in the MOX one (thus having a more determining influence on its spectral characteristics), and, on the other hand, the thermal absorption rates, at beginning of life, in the uranium-free assembly are enhanced due to the presence of the burnable poison.

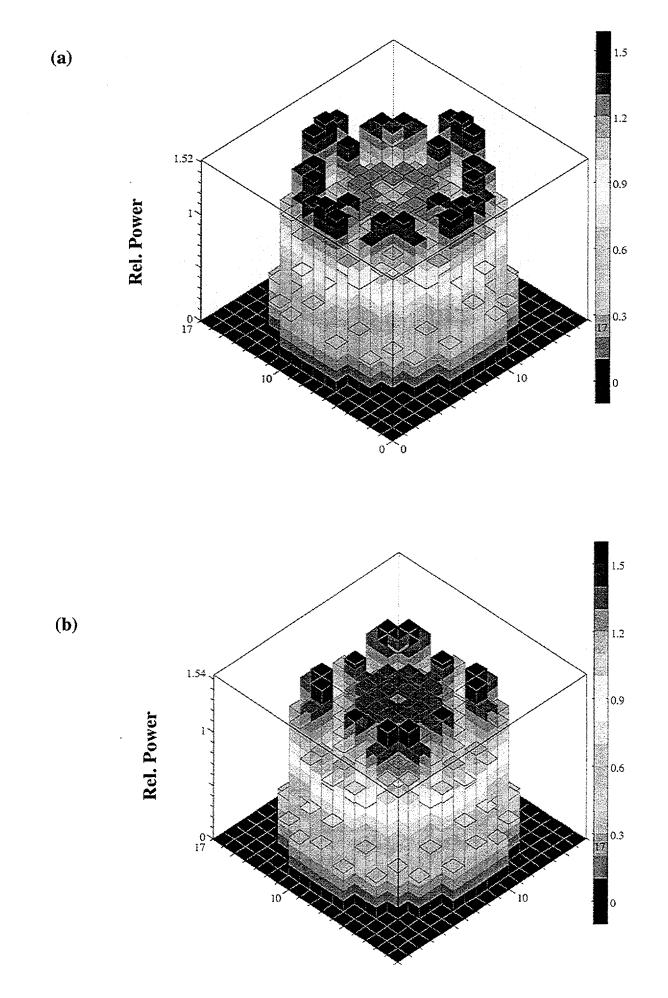
Based on the results presented in Fig. 6, an optimised heterogeneous design of a 15 x 15 uranium-free fuel assembly has been obtained (lower part of Fig. 7, <sup>1</sup>/<sub>4</sub>- symmetry). Its simplicity, when compared to advanced MOX fuel assemblies [15], is apparent. The distribution of the relative power densities for this subassembly surrounded by standard  $UO_2$ -assemblies at a depletion stage corresponding to 350 efpd is given in Fig. 7 (upper part). This burnup state - corresponding approximately to the end of the first cycle - has been chosen because it is the most demanding one (the supplementary gadolinia in the corresponding poisoned rod at the outermost corner position is depleted at this stage). As can be seen, the power distribution is very satisfactory (maximum form factor not exceeding 1.16).

To determine the influence of the introduction of the proposed uranium-free fuel assembly on the core parameters of a standard  $UO_2$ -fuelled PWR, three-dimensional full core analyses of a present-day 1 GW<sub>e</sub> PWR containing 4 and 12 such fuel assemblies have been performed. The main results are summarised below:

- loading 4 uranium-free assemblies does not alter any of the operational characteristics of the PWR core, as compared to the standard UO2 -fuelled case;
- in case 12 uranium-free assemblies are considered, the beginning-of-life boron concentration is lower than in the standard UO<sub>2</sub>-fuelled core.

The results summarised in Fig. 8 quantify the effects on the power distribution. In Fig. 8 (a), relative nodal power densities are indicated for the beginning of cycle situation of a standard  $UO_2$ -fuelled present-day 1 GWe PWR (maximum form factor not exceeding 1.52). The four uranium-free fuel assemblies have been loaded symmetrically into the peak power locations. The power distribution results for the beginning and end (gadolinia depleted) of cycle situations are shown in Fig. 8 (b) and Fig. 8 (c), respectively. As can be seen, satisfactory power distributions are attained, with form factors below 1.54 and 1.33 for the beginning and end of cycle, respectively.

The results presented so far strengthen the case for a gradual introduction of uranium-free fuel assemblies having the design proposed into a present-day 1 GW<sub>e</sub> PWR. They indicate that loading 4 to 12 such assemblies permits to envisage fuel management schemes compatible with current operational requirements (i.e. cycle length, boron concentration, etc.). However, when considering, in absolute terms, the plutonium consumption capabilities of such a core, caution is due with respect to using results obtained for 100% uranium-free PWR loadings [14]. Clearly, additional studies are necessary to define a detailed fuel management strategy beyond the 4-12 uranium-free fuel assemblies considered in the present study, and towards 100% uranium-free PWR-cores (about 430 kg/GW<sub>e</sub>a and 1090 kg/GW<sub>e</sub>a plutonium consumption rate, respectively [14]), conclusions with regard to the relative



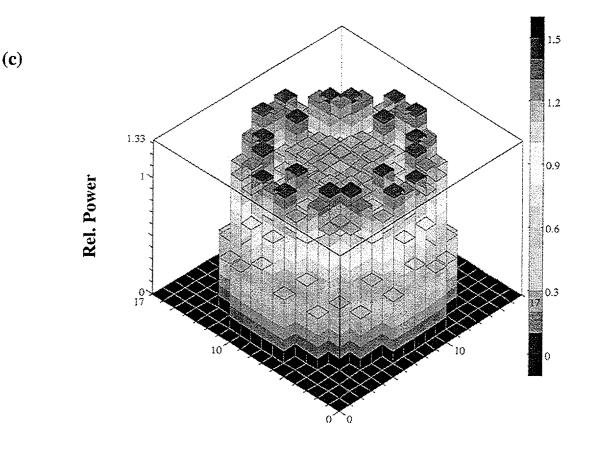


FIG. 8. Relative Power Distributions Derived from 3D Full-Core Calculations for (a) a Standard Uranium-Oxide Fuelled PWR at BOC, and for the Same PWR with 4 Uranium-Free Plutonium Assemblies Loaded into the Peak Power Location at (b) BOC and (c) EOC.

plutonium consumption capability of uranium-free fuelled PWRs as compared to MOX fuelled ones can be drawn.

The 1 GW<sub>e</sub> PWR considered consumes the same amount of plutonium when loaded with 4 or 12 uranium-free fuel assemblies, as it would with 10 or 30 MOX assemblies, respectively. In other terms, if a PWR loaded with a 1/3 loading of MOX assemblies can be considered as operating in a "self-generating mode", the same would be achieved with as little as a 1/8 loading of uranium-free fuel assemblies. The uranium-free fuelled PWR has an edge over the 100 % MOX core also when comparing the minor actinides (MA) production. Per unit amount of plutonium consumed, the MA production is almost a factor of 2 lower in the former case [14].

#### 4. THE FUEL ASSEMBLY AS WASTE FORM

An assessment of the radio-toxicity risk associated with the waste obtained from uranium-free fuelled PWRs and a comparison with MOX have been carried out on the basis of the nuclide inventories in the discharged fuel. No barrier effects of the waste repository are considered. The radiotoxicity risk values are thus obtained by multiplying the nuclide activities in the discharged fuel by the corresponding effective ingestion dose conversion factors derived from the ICRP Publication 61 data on the limits on intake of radionuclides by workers [16]. Table 1 summarises the core inventories of the considered PWR variants at beginning and end of life. TABLE 1. CORE INVENTORIES AT BEGINNING (BOL) AND END OF LIFE (EOL) OF THE DIFFERENT 1 GW<sub>E</sub> PWR (1300 EFPD RESIDENCE TIME)

· · · · · · · · · · · · · · · · · · ·	ore invento	e inventory [10 <sup>3</sup> kg]				
PWR		BOL			EOL	
fuelled with :	Pu	U	MA	Pu	U	MA
UO <sub>2</sub>	0	88.4	0	1.0	82.6	0.10
100 % MOX	5.3	81.6	0	3.5	78.5	0.27
100 % U-free	7.7	0	0	3.3	0	0.39

# TABLE 2. RADIO-TOXICITY HAZARD POTENTIAL FOR THE DIFFERENT 1 $\mathrm{GW}_\mathrm{E}$ PWR SCENARIOS CONSIDERED

PWR	Radio-Toxicity hazard given in [10 <sup>6</sup> ·Sv/GW <sub>e</sub> a] after				
fuelled with:	10 <sup>3</sup> a	10 <sup>4</sup> a	10 <sup>5</sup> a	10 <sup>6</sup> a	
UO <sub>2</sub>	1270	257	14.1	2.33	
100 % MOX	899	162	7.38	1.80	
100 % U-free	670	109	5.20	1.69	

A meaningful comparison of the radio-toxicity hazard potential has to include also the reactor systems needed to feed the plutonium-consuming reactor. On the basis of the results given in Table 1, it is concluded that 5.3 and 7.7 standard  $UO_2$ -fuelled 1 GW<sub>e</sub> PWR are necessary to feed one 100% MOX and one uranium-free 1 GW<sub>e</sub> PWR, respectively. Accordingly, the radio-toxicity hazard potential given in Table 2 and Fig. 9 considers the waste coming from 5.3 PWRs for the 100 % MOX, and from 7.7 PWRs for the uranium-free fuelled case, on top of the waste produced by the respective plutonium burning reactor. Obviously, the plutonium unloaded from the  $UO_2$ -PWRs to feed the respective burners is not to be directly considered as waste. It contributes to the radio-toxicity only indirectly, i.e. after being recycled in the burners.

To help put the radio-toxicity hazards into perspective, a line at  $5 \cdot 10^6$  Sv/GW<sub>e</sub>a is drawn in Fig. 9. This value corresponds to the asymptotic toxicity hazard of the natural uranium (0.7% <sup>235</sup>U) mass needed to generate 1 GW<sub>e</sub> per year. As can be seen, the additional radio-toxicity hazard due to the fission products from actually generating this power falls below this level after less than 400 years. The radio-toxicity associated with the actinides (plutonium and minor actinides), on the other hand, reaches this limit only after 100000 - 200000 years, depending upon the fuel type considered. When comparing the radio-toxicity hazard in the period from  $1 \cdot 10^3$  to  $1 \cdot 10^6$  years (Table 2), the uranium-free case is the most favourable (a factor of approximately 2 lower than for the UO<sub>2</sub>-fuelled core), with the MOX case indicating hazard potentials about 25 - 30% higher.

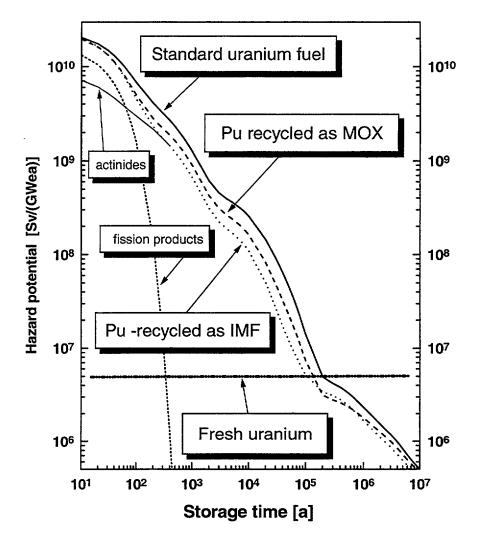


FIG. 9. Comparison of Hazard Potential

However, when assessing the potential of the proposed uranium-free fuelled PWRs, it must be borne in mind that the insolubility of this inert matrix fuel in water and acids, on the one hand, and the important reduction of the waste volumes by reprocessing with the utilisation of the chopped claddings as source of zirconium for the inert matrix material, on the other hand, offer considerable advantages over  $UO_2$ - and MOX-fuelled cores.

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## **ROX-LWR SYSTEM FOR ALMOST COMPLETE BURNING OF PLUTONIUM**

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#### Abstract

On the disposition of excess plutonium, a new system of Pu rock-like fuels (ROX) and their oncethrough burning in PWR has been studied. The features of the system are remarkable from the view point of proliferation resistance, environmental safety and plutonium annihilation rate. From the ROX fuel studies, twophase mixture of fluorite (yttria stabilized zirconia, SZR) and spinel (MgAl<sub>2</sub>O<sub>4</sub>) seems to be the most favorable candidate for ROX fuel material. Plutonium is solidified in the SZR by making solid solutions. Reactor safety analyses show that the ROX fueled PWR core has nearly the same performability as the existing UO<sub>2</sub> fueled core under both reactivity inserted and loss of coolant accidents. Plutonium annihilation rates of ROX fuels are found to be about twice as high as that of MOX fuels. From the results of environmental analyses, the committed individual doses from ROX spent fuels are lower than those from MOX/UO<sub>2</sub> spent fuels by two or more order of magnitudes.

#### 1. INTRODUCTION

The incineration of plutonium derived from spent fuels or from nuclear warheads, is one of the major challenges of the nuclear society in many countries around the world. The optional use of LWR and CANDU system is a conventional solution for disposition of excess plutonium, though the production of extra plutonium is its inherent disadvantage [1]. From the consideration of safeguards, safety, technological background and economy, however, it appears that the combination of conventional LWR and new inert matrix fuels would be an attractive option for the annihilation of the excess plutonium [2,3].

The inert matrix fuels are mixtures of fissile materials and nuclear stable ones. Two types of inert matrix fuel systems have been proposed and studied in the world: one is a Pu recycle system combined with reprocessing of spent fuels [4]. This type of fuels is designed to recover fissile materials by reprocessing and, therefore, needs to be soluble in diluted acids. In these fuels plutonium is dispersed in inert materials such as MgO,  $Al_2O_3$ ,  $MgAl_2O_4$  and so on. The other is a Pu once-through burning system combined with direct disposal of spent fuels after cooling [2,3]. This type of fuels is designed to be chemically, physically and geologically stable to improve proliferation resistance and environmental safety of the system. In these fuels plutonium is solidified into inert materials by forming a solid solution or a compound. Stabilized zirconia is the most hopeful candidate for the inert material solidifying plutonium in the latter system.

We proposed a once-through system of plutonium rock-like fuel (ROX) and LWR [2,3,5]. The schematic diagram of the ROX-LWR system is shown in Fig. 1. Plutonium recovered from spent fuels at reprocessing plants and/or taken out from dismantled warheads is converted into ROX fuels by mixing inert additives of  $Zr(Y)O_2$  (SZR), MgO and  $Al_2O_3$  in MOX fuel facilities. Fissile plutonium in ROX fuel is almost completely burned in conventional LWRs. After cooling for about 50 years the spent ROX fuels

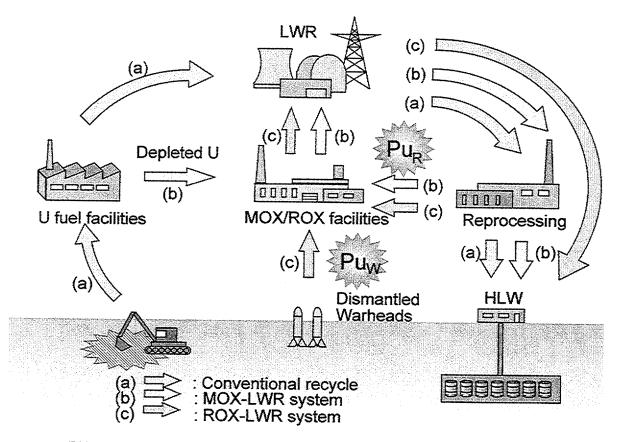


FIG. 1. Schematic diagram of ROX-LWR system and conventional fuel cycles Pu<sub>w</sub>: weapons grade plutonium; Pu<sub>R</sub>: reactor grade plutonium

are disposed as high level wastes (HLW) in repository sites under deep geological formations by conventional technologies. The ROX-LWR system is basically built on the conventional technologies and facilities.

In the present paper, current R&D activities on the ROX-LWR system are presented, which covers the fundamental aspects of the fuel materials, irradiation behavior, reactor design, accident safety analyses and environmental safety analysis. The outline of the R&D program in future is also included.

2. ROX FUEL STUDIES

2.1. Studies on basic properties of ROX materials

Through consideration of physico-chemical properties of stable minerals and ceramics, we expected that the optimum composition of fuel material candidates might be exist in the ternary SZR (or ThO<sub>2</sub>)-MgAl<sub>2</sub>O<sub>4</sub>-Al<sub>2</sub>O<sub>3</sub> system. Phase relations and element distributions over phases of simulated fuels and spent fuels were investigated by X-ray diffraction, scanning electron micro-analysis (SEM) and electron probe micro-analysis (EPMA) methods. These studies had been made systematically in the two-phase mixture of SZR (or ThO<sub>2</sub>)-Al<sub>2</sub>O<sub>3</sub> [2,5], the two-phase mixture of SZR (or ThO<sub>2</sub>)-MgAl<sub>2</sub>O<sub>4</sub> [6], and the three-phase mixture of SZR (or ThO<sub>2</sub>)-MgAl<sub>2</sub>O<sub>4</sub>-Al<sub>2</sub>O<sub>3</sub> [7] systems. From these studies, we obtained the following findings.

a) Plutonium, minor actinides and rare earth elements are distributed in fluorite phases (SZR and/or ThO<sub>2</sub>) by making solid solutions.

b) Most of solid fission products (FPs) are solidified into the fluorite and spinel phases by making solid solutions.

c) Noble metals are precipitated as alloys with molybdenum and long lived nuclides such as  $^{79}$ Se,  $^{99}$ Tc,  $^{107}$ Pd and  $^{126}$ Sn in the matrix.

d) With the presence of excess  $Al_2O_3$  in the fuel matrix, alkaline earth elements (Sr and Ba) and some rare earth elements form a hibonite-type phase (SrO·  $6Al_2O_3$  and/or  $M_2O_3$ ·  $11Al_2O_3$ ) with the alkali metals (Mg, Cs and Rb).

We also measured some basic thermo-physical properties of ROX fuels such as thermal conductivity, specific heat capacity, melting temperature and so on [7,8]. The typical values of ROX fuel with the composition of 44mol% fluorite ( $16\%UO_2+28\%SZR$ ) and 56mol% spinel (MgAl<sub>2</sub>O<sub>4</sub>) are shown in Table 1 together with those of UO<sub>2</sub> fuel for comparison. The thermal conductivity of ROX with fluorite and spinel phases is about 50% higher than that of UO<sub>2</sub> at high temperatures. Spinel phase markedly improves the thermal conductivity of ROX. The eutectic temperature of ROX is 2,210 K which is lower than the melting temperature of UO<sub>2</sub> by about 900 K. Addition of excess  $Al_2O_3$  also remarkably improves the thermal conductivity of ROX especially at low temperatures and, however, causes to lower the eutectic temperature by about 100 K. Therefore we concluded that the addition of excess  $Al_2O_3$  is not favorable for ROX fuel performance. So far, the best composition of ROX is considered to be about an equimolar mixture of fluorite and spinel phases.

Properties	ROX fuel	UO <sub>2</sub> fuel
density (g cm <sup>-3</sup> )	6.0	11.0
specific heat capacity (J g <sup>-1</sup> K <sup>-1</sup> )	0.58 (573 K) 0.74 (1,273 K)	0.29 (573 K) 0.32 (1,273 K)
melting temperature (K)	2,210	3,140
thermal conductivity (W m <sup>-1</sup> K <sup>-1</sup> )	6.5 (573 K) 4.5 (1,273 K)	5.3 (573 K) 3.0 (1,273 K)
nermal expansion coefficient (K <sup>-1</sup> )	9.5x10 <sup>-6</sup>	$10.0 \times 10^{-6}$

TABLE 1. THERMOPHYSICAL PTOPERTIES OF ROX AND UO<sub>2</sub> FUELS ROX: 44mol% fluorite (16%UO<sub>2</sub>+28%SZR)+56mol% spinel (MgAl<sub>2</sub>O<sub>4</sub>)

Irradiation damage of ROX matrix materials by a low energy Xe (80-100 keV) beam was studied by an electron microscope combined with a low energy ion beam apparatus[9,10]. Swelling rates of both SZR and MgAl<sub>2</sub>O<sub>4</sub> were estimated to about 0.1% by a Xe irradiation below 160 dpa at room temperature, and 0.24% at 920 K. No amorphousness was observed for SZR by Xe irradiation upto 160 dpa. These findings suggest that both SZR and MgAl<sub>2</sub>O<sub>4</sub> are suitable for the ROX matrix from the view point of the irradiation damage.

The chemical stability of ROX was examined using simulated spent ROX fuels, which consisted of five-phase mixture of fluorite, spinel, corundum, hibonite and alloy phases, by observing dissolution behavior in hot nitric acid solution, and leachability in hot water by analyzing the amount of leached elements, respectively [11]. Dissolution amounts of the test in 3 mol-HNO<sub>3</sub> at 420 K for 3 days were 2% for Zr, 30-50% for Al and Mg, and 60-98% for Ba. These facts show that the fluorite phase which is the host phase of plutonium is remarkably stable against the hot HNO<sub>3</sub>. Accordingly, it may be said that the

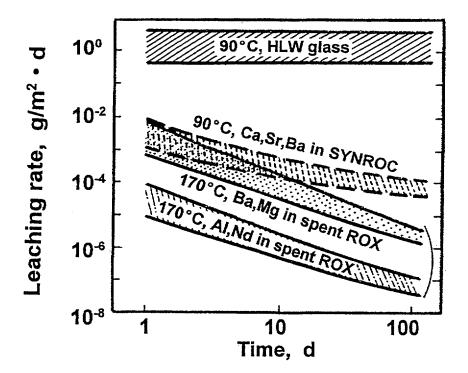


FIG.2. COMPARISON OF LEACHING RATES OF MAIN ELEMENTS IN SIMULATED SPENT ROX IN DEIONIZED HOT WATER WITH THOSE OF HLW GLASS AND SYNROC. HLW glass: total weight loss [12], SYNROC: titania waste form and Ti was below detection limit [13], Spent ROX: fluorite+spinel+corundum type fuel and Zr was below detection limit.

ROX fuel will become a chemical barrier for plutonium proliferation. Because the hibonite phase is the least unstable among the phases in the simulated spent fuel, it seems to be desirable to avoid the formation of this phase: that is the presence of excess  $Al_2O_3$  in the ROX fuel is not favorable. The leaching rates of main elements in simulated spent ROX in hot water are shown in Fig. 2, together with those of HLW glass [12] and SYNROC [13]. As can be seen easily, the leach rates of the elements in spent ROX are lower than those of HLW glass by about 4-5 orders of magnitude at the end of experiments, which is probably due to low solubilities of fuel matrix materials in hot water.

2.2. Irradiation behavior of ROX fuels

Irradiation and post-irradiation examination (PIE) of ROX fuels were performed to confirm the results obtained by simulated fuel and spent fuel studies, and to find out new phenomena associated with in-pile irradiation.

Two types of ROX fuel disks were fabricated and compositions and phases are summarized in Table 2. The formation of hibonite phase was not expected from the results of simulated fuel studies. The new hibonite phase contains Pu, Al and some Mg and may have a form of  $(Pu,Mg)_2O_3$ ·  $11Al_2O_3$ . Although we speculate that the fuel sintering in hydrogen atmosphere at 1,700 K might cause the formation of the Pu-hibonite phase due to a part of reduction of  $Pu^{4+}$  to  $Pu^{3+}$ , further studies require to clarify the point.

These fuel disks were encapsulated in a fuel pin and irradiated in Japan Research Reactor 3 (JRR-3) for 2,330 hrs at full power. Maximum neutron fluences were estimated to be  $8.1 \times 10^{24}$  and

 $3.4 \times 10^{24}$  m<sup>-2</sup> for thermal and fast neutrons, respectively [14]. Three fuels pins of each fuel type were properly located and the irradiation temperatures were controlled at 870, 1,270 and 1,070 K at the top, middle and bottom, respectively, by adjusting the flow rate and/or composition of cooling gas mixtures (He and N<sub>2</sub>). Burn-up rate was 27.9% FIMA for the fuel disk at the middle position and 20.7% FIMA for that at the top by the destructive chemical analysis combined with the <sup>148</sup>Nd method [15].

Materials	Zr-system (%)	Th-system (%)
PuO <sub>2</sub>	10	10
SZR*	15	-
ThO <sub>2</sub>	-	15
$Al_2O_3$	65	65
MgO	10	10
Phase	lattice parameter (nm)	lattice parameter (nm)
Fluorite	0.5180	0.5537
Corundum	a=0.4750	a=0.4760
	c=1.280	c=1.280
Hibonite	a=0.5586	a=0.5588
	c=2.192	<b>c=</b> 2.191
Spinel	tr.	tr.

TABLE 2. COMPOSITIONS AND PHASES OF FRESH ROX FUEL DISKS.

\* SZR: 88.8% ZrO<sub>2</sub> + 11.0% YO<sub>1.5</sub> + 0.2% GdO<sub>1.5</sub>

Post-irradiation examination was performed at hot cell laboratory in JAERI for items such as X-ray photography and gamma-ray scanning of fuel pins, FP gas release rate measurements, metallography, SEM & EPMA and X-ray diffraction analysis of fuel disks, dissolution of fuels and burn-up measurements, and others. Results of gamma scanning showed that a part of <sup>137</sup>Cs and <sup>134</sup>Cs moved from fuels disks to cooler parts of the fuel pin and, however, the rest of non-volatile FPs such as <sup>106</sup>Rh and <sup>144</sup>Pr remained in the fuel disks. Almost all of FP gas (Xe and Kr) were released from fuels disks to the plenum part of the pin probably due to the low densities of fresh fuel disks (about 80%TD) and to some swelling during irradiation (porosities of irradiated disks measured by metallography were in the range of 38-45%). Tentative swelling rate during irradiation was estimated to be about 10% at 10MWDd/kg for these fuel disks.

X-ray diffraction patterns of irradiated fuels of Zr-system were shown in Fig.3 with those of fresh and simulated spent fuels for comparison. As can be seen from the figure, as burn-up increases, the corundum phase disappears and, in turn, amounts of the hibonite and spinel phases increase. No appreciable peak broadening of the hibonite phases are observed compared to those of the fresh fuel suggesting that the hibonite phase has a relatively high stability against irradiation.

Element distributions over the phases in fresh and irradiated fuels were determined by SEM & EPMA. The results are summarized in Table 3. Distribution of most of elements are consistent with the results obtained by simulated spent fuel studies except for plutonium. Plutonium is expected to be in the fluorite phase making solid solutions with SZR or ThO<sub>2</sub> and, however, the observation results were quite different: that is, most of plutonium was observed with zirconium or thorium and the rest was observed with aluminum, in the fresh fuel. With increasing burn-up, almost all of plutonium was observed with aluminum and not with zirconium nor thorium. Because the formation of hibonite phase requires the

reduction of  $Pu^{4+}$  to  $Pu^{3+}$ , we estimated the oxygen potential around fuel disks during irradiation and found that the oxygen potential was well below -800kJ/mol at 1,273 K which was enough to reduce  $Pu^{4+}$  to  $Pu^{3+}$ [16]. To prevent the hibonite phase from forming, we must avoid adding excess  $Al_2O_3$  to fuel matrix as well as extremely low oxygen potentials during fuel fabrication and irradiation. Further studies are now underway to establish chemical and physical properties of the new Pu-hibonite compound.

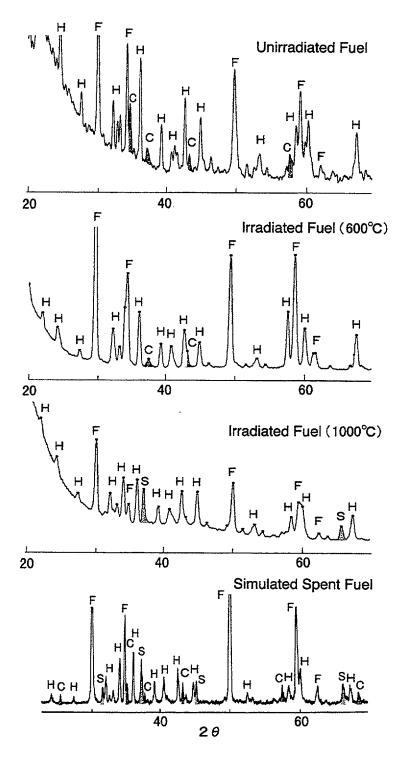


FIG. 3. X-RAY DIFFRACTION PATTERNS OF IRRADIATED ROX FUELS WITH THE SZR+SPINEL+CORUNDUM SYSTEM.

F: fluorite (ZrO<sub>2</sub>(Y,Gd)); H: hibonite (Pu<sub>2</sub>O<sub>3</sub>·11Al<sub>2</sub>O<sub>3</sub>); C:corundum (Al<sub>2</sub>O<sub>3</sub>); S: spinel (MgAl<sub>2</sub>O<sub>4</sub>)

	Phase					
	Fluorite	Spinel	Corundum	Hibonite	Alloys	
Zr-type fuel (1,300K)			<u></u>			
Pu	+		-	+++	-	
Lanthanides	++		_	+++	-	
Alkaline-earths		++	_	+++	-	
Mo, Tc, Pt group	-	-	-	-	+++	
Th-type fuel (1,300K)						
Pu	-	-	-	+++	-	
Lanthanides	++		_	+++	-	
Alkaline-earths	_	++	_	+++		
Mo, Tc, Pt group	_		-	_	+++	

#### TABLE 3. DISTRIBUTION OF ELEMENTS IN IRRADIATED ROX FUELS

+++: primary distribution, ++: secondary distribution, +: detected a little, -: not detected.

### 3. SAFETY ANALYSIS AND PLUTONIUM ANIHILATION RATE

#### 3.1. Safety analysis

Preliminary reactivity calculations for the ROX fueled PWR core showed that the ROX fuel had several disadvantages such as smaller negative Doppler and void reactivities, and larger power peaking factors compared to  $UO_2$  and MOX fuels. Partial loading of ROX fuels in PWRs is one way to improve these disadvantages but results in low plutonium annihilation rates. The other approach for improving them is addition of resonant nuclides such as <sup>232</sup>Th and <sup>238</sup>U. Parametric calculations were performed against one GWe class 17x17 type PWR with three batches refueling and fuel shuffling. The cycle length was about 450 effective full power days (EFPD). The calculated results are summarized in Table 4. It is clear that considerable improvements in the reactivities are achieved by adding 24mol% Th or 15mol% U.

The reactivity insertion event was analyzed with EUREKA-2 code [17]. The inserted reactivity values and scram conditions for ROX-PWR are the same as those of general PWRs. The main results of reactor safety analysis on ROX-PWR system are shown in Table 5 together with those of UO<sub>2</sub> fuel [18]. The temperature of the ROX fuel pellet could be reduced below that of UO<sub>2</sub> by adding Th (24%) and U (15%). The maximum temperature of heterogeneous  $1/3ROX+2/3UO_2$  fueled core is comparable to that of UO<sub>2</sub>. Because of smaller densities of ROX fuels, which are around  $6g/cm^3$ , the fuel enthalpies should be compared with that of UO<sub>2</sub> fuel by energy deposited per unit volume. As can be seen from Table 5, they are comparable to that of UO<sub>2</sub> fuel. The fundamental behavior of ROX fuels under reactivity initiated accident conditions and the fuel failure energy deposition need to be clarified by reactivity accident experiments, and theses studies are in progress using Nuclear Safety Research Reactor (NSRR) in JAERI. Preliminary results of the second pulse irradiation of ROX fuel test in NSRR, where the inserted fuel energy deposition was 380 cal/g·UO<sub>2</sub> (or 146 J/cm<sup>3</sup>) that is about twice as large as the limited value for UO<sub>2</sub> fuels, showed that no failure of the fuel pin was observed although the maximum temperature of cladding surface reached to 1,470 K and partial melt of the ROX fuel was observed [19].

# TABLE 4. VOID AND DOPPLER REACTIVITIES AND POWER PEAKING FACTORS IN THE ROX-FUELED PWR AT BOC.

ROX is the full ROX fueled core PWR with fuel composition of 5.2%PuO<sub>2</sub>+38.8%SZR+56%MgAl<sub>2</sub>O<sub>4</sub>; ROX-15%U and ROX-24%Th are U and Th added partially in place of SZR in the ROX fuel, respectively; ROX(Er) means Gd and Er added in SZR; 1/3ROX+2/3UO<sub>2</sub> is a heterogeneous core loading 1/3ROX in the conventional UO<sub>2</sub> fueled assemblies. Plutonium is the weapons grade.

Reactivity and peaking factor	ROX	ROX(Er)- 15% U	ROX- 24% Th	1/3ROX +2/3UO <sub>2</sub>
Void reactivity				
0 to 40% void	-0.32	-6.0	-5.6	-3.4
0 to 95% void	-17.7	-21.0	-42.0	-40.0
Doppler reactivity				
900 to 1,200K	-0.098	-0.61	-0.56	-0.48
600 to 900 K	-0.132	-0.72	-0.70	-0.58
maximum peaking factor	3.0	2.1	2.4	2.3

The loss of coolant accident (LOCA) event of ROX fueled PWR was analyzed by using the best estimate code RETRAN2 (MOD3) [20]. The analyzed system and event were a 4-loop type one GWe class PWR and a cold leg large break LOCA event at BOC and EOC. The calculated results are listed in the last column of Table 5. Contrary to the ROX fuel with no additives of resonant nuclides where the peak cladding temperature rises more than 1,500 K, the highest cladding temperature would be controlled lower than the limited value of 1,373 K by addition of such resonant nuclide as U and Th. This is due to flattening the power distribution and decreasing Doppler coefficient by adding Th of U.

	Reactivity	accident	Loss-of coolant accident
Fuel type	Max. fuel temp. (K)	Enthalpy (kJ/cm <sup>3</sup> )	Max cladding temp. (K)
ROX -24%Th	1,960	5.23	1,240 (p.f.=2.4)
ROX -15% U	1,700	4.52	1,090 (p.f.=2.1)
1/3ROX+2/3UO <sub>2</sub>	2,120	5.69	1,240(p.f.=2.8)
$UO_2$	2,090	4.27	1,080 (p.f.=2.0)

## TABLE 5. RESULTS OF ACCIDENT ANALYSES OF ROX-PWR SYSTEM

#### 3.2. Plutonium annihilation rates

Core burn-up calculation was made by using SRAC-COREBN code system on 2-dimensional X-Y core model of ROX-PWR [21]. Table 6 shows the calculated plutonium annihilation rates for some fuel types [22]. In the case uranium was contained in the fuel, plutonium generated from the uranium was included in the Pu-input. As for the annihilation of weapons grade plutonium, the ROX-24% Th fuel shows the highest transmutation rates of <sup>239</sup>Pu and total Pu, respectively. The transmutation rate for MOX fuel case becomes about 30% of total Pu, and 1/3MOX+2/3UO<sub>2</sub> fuel case shows the increase of total Pu. In the MOX fuel case, though the amount of input plutonium is similar to those of ROX fuels, the plutonium transmutation rates of total and <sup>239</sup>Pu are about a half of those of ROX fuels, respectively.

## TABLE 6. COMPARISON OF ANNIHILATION RATES OF PU IN ROX-PWR SYSTEM (Ton/GWe/300EFPD)

		Weapons-grade Pu			Reactor-grade Pu		
Loading/F	uel type	input	output	transmutation	input	output	transmutation
ROX-	<sup>239</sup> Pu	0.89	0.03	0.86 (97)	0.86	0.09	0.77 (90)
24% Th	total Pu	0.96	0.22	0.74 (77)	1.47	0.56	0.90 (62)
ROX-	<sup>239</sup> Pu	0.84	0.07	0.77 (92)	0.88	0.18	0.70 (80)
15% U	total Pu	0.90	0.21	0.69 (77)	1.50	0.72	0.78 (52)
мох	<sup>239</sup> Pu	0.81	0.30	0.51 (63)	0.87	0.47	0.40 (46)
	total Pu	0.86	0.60	0.26 (30)	1.49	1.12	0.37 (25)
1/3MOX+	<sup>239</sup> Pu	0.27	0.18	0.09 (33)	0.29	0.24	0.05 (17)
2/3UO <sub>2</sub>	total Pu	0.29	0.35	-0.06 (-20)	0.50	0.53	-0.03 (-6)

(2-dimensional core calculation and cell calculation)

The transmutation rate (%) of Pu is shown in ().

### 4. ENVIRONMENTAL SAFETY ANALYSIS

Based of the findings described in section 2.1, a comparative study of environmental safety was carried out for the direct disposal of ROX and MOX spent fuels. Safety analyses were performed on the case that one MTU of spent fuels are disposed into a deep granite rock formation, and the individual dose equivalent due to the exposure pathway ingesting drinking water was calculated by GSRW code[23] based on the groundwater migration scenario. Figure 4 shows the committed dose equivalent originated from the 4n+1 decay chain. <sup>229</sup>Th gives the maximum dose equivalent among the daughter nuclides both in ROX and MOX fuels, and the values are  $1.9 \times 10^{-12}$  and  $6.0 \times 10^{-10}$  Sv for reactor grade-ROX and for reactor grade-MOX fuels by two or more order of magnitude. It is also found that the dose equivalents from weapons grade-ROX is less than that of reactor grade-ROX. Similar results are also obtained for the other decay chains of 4n, 4n+2 and 4n+3 [24].

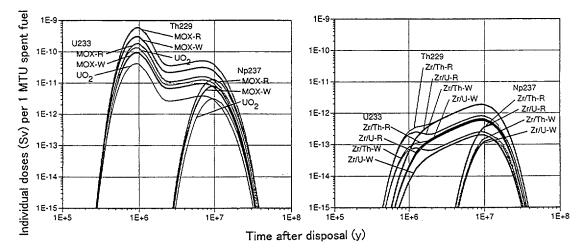


FIG.4. INDIVIDUAL DOSES FROM THE 4N+1 DECAY CHAIN OF DISPOSED SPENT ROX AND MOX FUELS.

### 5. FUTURE R&D PROGRAM

Based on the achievements obtained from the feasibility study of ROX-LWR system during 1994-1996, we are developing the second phase study to establish the basic technologies for the ROX-LWR system by the end of 2001. The main R&D items are as follows:

- (1) Fundamental studies of ROX fuels
  - $\cdot$  Research for new/advanced ROX fuels with higher thermal conductivity, melting temperature and stability against irradiation damage
  - · Measurements and evaluation of physico-chemical properties of ROX fuels
- (2) Engineering studies of ROX fuels
  - · Establishment of fuel fabrication process
  - · Establishment of irradiation behavior of ROX fuels
- (3) Reactor core design and safety analyses
  - · Measurements and evaluation of Doppler reactivity for resonant nuclides
  - · Measurements and evaluation of nuclear data for Pu and minor actinides
  - · Establishment of burn-up calculation methods for ROX fuels
- (4) Total assessments of ROX-LWR system
  - · Evaluation of environmental safety based on the second phase results
  - · Evaluation of cost of ROX-LWR system based on the second phase results

Among these R&D items, we focus on the irradiation behavior of ROX fuels in the second phase research. Two irradiation programs are in progress using pellet type fuels with SUS or Zircaloy claddings. Irradiation will be performed under real reactor operating conditions. Irradiation schedule is shown in Table 7.

## TABLE 7. SCHEDULE FOR ROX FUEL IRRADIATION AND PIE

	Year	1997	1998	1999	2000	2001
ROX(U) fuel irradi	ation at	JRR-3 in JA	AERI			
Fuel design	-					
Fuel fabrication	n	····				
Irradiation						
PIE						
ROX(Pu) fuel irrad	iation at	HFR in EC	N Petten, Net	herlands		
Fuel design		-				
Fuel fabrication	ı		<del></del>			
Irradiation						
PIE						

On the annihilation of excess plutonium, feasibility of a new system of plutonium rock-like fuels (ROX) and their once-though burning in PWRs has been investigated in these 4 years. The R&D activities have been carried out in such areas as fundamental studies on ROX fuel materials, irradiation behavior, reactor core design and burn-up calculations, reactor safety analyses, environmental analyses and cost estimation of the ROX-LWR system. From these activities, it is found that the ROX-LWR system has remarkable features as follows:

#### (1) Proliferation resistance based on ROX fuels

As the chemical stability of ROX fuels is excellent in hot nitric acid solution, they become themselves bearers of proliferation resistance. In the ROX-PWR system, about 0.9 ton of Pu a year will be burned in one GWe PRW, and about 95% of <sup>239</sup>Pu and 80% of total Pu in ROX fuels will be transmuted. As a result, the spent fuels do not contain valuable Pu nuclides.

#### (2) Environmental safety of ROX spent fuels

The leach rates of spent ROX fuels are remarkably low. Their geological stabilities are supported by those of stable natural minerals and rocks (natural analogues). Therefore, the committed individual doses from direct disposal of the ROX spent fuels are lower than those from MOX/UO2 ones by two or more order of magnitudes.

(3) Economical by use of conventional facilities

The system consists of conventional facilities with normal equipments based on the present technological backgrounds such as MOX fuel facilities, PWR and HLW (spent fuel) repository.

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#### CANDU INERT-MATRIX FUEL

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#### Abstract

Significant effort is underway internationally to develop a non-UO<sub>2</sub>-based inert-matrix fuel capable of hosting fissile isotopes without generating new plutonium or high-mass carcinogenic actinides. The intention is to be able to dispose of military or reactor-grade plutonium, or eliminate carcinogenic actinides. AECL has a program that focuses on the development of SiC as the candidate material for this new form of fuel; the program includes accelerator-simulation studies of in-reactor behaviour of SiC, fabrication of SiC, compatibility of SiC with water and Zircaloy, waste disposal of SiC, as well as examining reactor physics issues related to SiC. CANDU reactors have been shown to be uniquely suited for this purpose. Results of this AECL program are described.

#### 1. INTRODUCTION

A worldwide effort is underway to find the best ways to dispose of the military plutonium that is being freed by dismantling of nuclear weapons from the United States and Russia. The United States Department of Energy (USDOE) has set this as one of its primary goals for the near and intermediate future. Towards this end, Canada is investigating the potential of burning mixed-oxide (MOX) fuel in one or two of its CANDU reactors [1]. In the United States, underground disposal of the military plutonium as a waste-glass is also under serious consideration. And, a number of institutions, including AECL, are investigating a new form of non-uranium-based fuel, termed *inert-matrix* fuel, as a longer-term option for burning the plutonium without generating new plutonium at the same time as MOX or any uranium-based fuel does. This paper addresses this last topic—inert-matrix fuel—with emphasis on silicon carbide.

In Europe, a significant program is underway, not on the disposition of military plutonium, but on the annihilation of certain long-lived, carcinogenic actinides produced during normal reactor operation, and concentrated by reprocessing of the fuel<sup>4</sup>. Here, the intent is to make an inert-matrix fuel incorporating these actinides (<sup>237</sup>Np, <sup>241</sup>Am, <sup>244</sup>Cm) together with plutonium that is usually separated with these actinides. The plutonium provides a major fissile component and the neptunium, americium and curium are transmuted or fissioned to shorter-lived, less carcinogenic nuclides. Without <sup>238</sup>U in the fuel matrix, further production of these actinides does not occur. The challenge to develop an inert-matrix material is the same as for plutonium annihilation, above. Thus there are two separate possible applications for a proven inert-matrix fuel material—plutonium annihilation and actinide-waste destruction.

There is also a third potential application—burning reactor-grade plutonium. The US efforts on developing an inert matrix is for this application. The amount of plutonium produced in the next 10 to 20 years, intended for use in MOX fuel or fast-reactor fuel, may necessitate the use of inert-matrix fuel to help dispose of the plutonium.

<sup>&</sup>lt;sup>1</sup> Now at Bubble Technologies Industries, Hwy 17, Chalk River, ON K0J 1J0

<sup>&</sup>lt;sup>2</sup> Now at ACERAM, Hwy 17, Chalk River, ON K0J 1J0

<sup>&</sup>lt;sup>3</sup> Now at Chalk River Laboratories, Chalk River, ON K0J 1J0

<sup>&</sup>lt;sup>4</sup> For deep geological disposal of spent fuel in an underground repository, Canadian assessments have shown that the radiological risk from the actinides is negligible [2]

CANDU reactors are particularly suited for all three of these applications [3] because no major modifications to reactor design are required and because the neutron economy of CANDU reactors means the annihilation of plutonium/actinide waste can be more complete.

This paper surveys briefly programs that are underway worldwide and then describes AECL's program and progress. This material was presented at the last CANDU fuel conference [4].

#### 2. PROGRAMS AND CANDIDATE MATERIALS

The major European effort is a multi-national program called EFTTRA (Experimental Feasibility of <u>Targets for Transmutation</u>) [5] which is directed towards actinide-waste annihilation. The French fastreactor programs CAPRA and SPIN [6]<sup>1</sup> address annihilation of plutonium and recycling and burning of minor actinides, and include fast-reactor irradiations of candidate materials. Japan has a program [7] directed towards annihilation of military plutonium (particularly Russian military plutonium), which they call PROFIT (Plutonium Rock-like Fuel In-Reactor Technology). Their idea is to use a combination of materials to produce a multi-phase inert-matrix material that will resemble ordinary rock, even after irradiation, and thus facilitate underground burial as a waste.

The US program on inert-matrix material development, smaller than EFTTRA and PROFIT, focuses on zirconia  $(ZrO_2)$  [8]. Zirconia is one of the few materials that has already been shown to behave acceptably as a fuel material (with the addition of <sup>235</sup>U). Its major disadvantage is its poor thermal conductivity (slightly lower than UO<sub>2</sub>), which means that fuel temperatures will be as high as, or higher than, those in today's fuels. Other researchers are searching for an inert-matrix material that, besides its ability to function acceptably as fuel host in-reactor, has a higher thermal conductivity than UO<sub>2</sub> does so that fuel temperatures will be lower, fission-gas release from the fuel will be lower, and safety margins will be increased.

Many of the candidate materials and the properties that make them suitable as inert-matrix candidates can be found in Ref. 2. Those that we have worked with include  $ZrSiO_4$  (zircon), MgAl<sub>2</sub>O<sub>4</sub> (spinel), CeO<sub>2</sub>, CePO<sub>4</sub>, ZrO<sub>2</sub>-doped with Ca, Ce, Er or Y, and SiC. Cerium oxide (CeO<sub>2</sub>) has the same crystal structure as UO<sub>2</sub>, and was thought therefore that it would behave well in-reactor. Zircon is a stable mineral found in the earth's crust, and therefore at least should be a stable waste form. It is being evaluated as a candidate for hosting military plutonium for direct burial in the ground. Cerium phosphate is a candidate supplied to us from the Netherlands (via the Transuranium Institute in Germany).

Many papers are being written on these subjects. In addition to those discussed above, Burghartz et al. [9] presented information on the fabrication, thermal properties and stability to alpha radiation on five materials –  $MgAl_2O_4$ ,  $ZrSiO_4$ ,  $CeO_2$ , SiC and  $Si_3N_4$ . At the International Conference on Future Nuclear Systems (Global '97) in Yokohama, 1997 Oct. 5-10, at least eleven papers were presented on various aspects of, and choices for, inert matrix fuels, including, for example [5,7,8,10,11]. These references list many more on this subject.

#### 3. AECL'S PROGRAM

Silicon carbide stood out early as a promising candidate material because of its high melting temperature, very high thermal conductivity and known resistance to attack by many corrosive agents, including oxygen, even at high temperatures. Because no other laboratory in the world was examining SiC for this purpose, AECL focused its efforts on SiC, although the Tandem Accelerator studies described in the next paragraph were used for many candidate materials. The overall program was aimed at examining all issues that would be important in the selection of SiC as an inert-matrix fuel candidate.

Because of the large number of candidate materials, we utilized the Tandem Accelerator to simulate in-reactor conditions to help sort out those candidate materials that would behave well as a fuel. The

<sup>&</sup>lt;sup>1</sup> acronyms not given

critical unknown issue for candidate materials is how they will behave in-reactor under the severe damage inflicted by fission fragments blasting through the matrix at energies greater than 70 MeV. Especially, will the candidate materials become amorphous or undergo an unacceptable volume-change? Producing fuel from each candidate material for in-reactor tests is expensive, particularly with plutonium, so the Tandem Accelerator at AECL was used to bombard the candidate materials with a beam of 72 MeV iodine ions. This beam substitutes for fission fragments, and the bombarded areas were examined for damage, especially swelling. To increase the number of candidate materials available for testing, and to foster international collaboration, AECL tested candidate materials from the countries of the European Union and the United States, as well as its own.

Besides these accelerator-simulation tests, the program on SiC consisted of assessing:

- reactor-physics aspects of SiC fuels in CANDU reactors
- fabrication issues associated with SiC high sintering temperatures, handling plutonium, and characterization of microstructures that form with the addition of plutonium and several sintering aids
- compatibility of SiC with water at reactor coolant conditions and with groundwater for eventual storage
- compatibility of SiC with Zr-based sheath materials under accident temperatures
- waste-disposal aspects.

#### 4. REACTOR PHYSICS

Detailed fuel-management simulations demonstrate that an existing CANDU reactor can burn inertmatrix fuel containing plutonium or actinides without major modification. Fuelling rate, channel power, bundle power and element power ratings are all within current limits. The reference fuel, described in the next paragraph, would destroy 94% of the fissile plutonium.

The reference Pu-SiC fuel for Pu-annihilation contains 250 g of weapons-derived plutonium mixed uniformly with SiC in the outer 30 elements of a standard 37-element bundle. The central element contains 20 g of gadolinium. Another 40 g of gadolinium are distributed uniformly over the 6 elements in the next ring. The distribution of the gadolinium, which has a very high depletion rate, was optimized to suppress the excess reactivity of the fresh fuel and to minimize the power ripple that is due to refuelling.

Detailed fuel-management simulations using a uniform 2-bundle-shift refuelling scheme in a Bruce A reactor indicated that the maximum instantaneous channel power is below 7.1 MW and the maximum instantaneous bundle power is below 970 kW. Both are within the current licensing limits. The fuelling rate is about 15 bundles per full-power day of operation, which is within the current fuelling machine capability.

For this Pu-SiC fuel, as well as for all the conventional MOX options, void reactivity is negative. Hence there would be no power pulse in a postulated LOCA, and the safety and licensing analyses would be greatly simplified.

The fuel temperature coefficient is very slightly positive, about 12  $\mu$ k/°C; however, this is irrelevant in the safety analysis since any increase in heat in the fuel would immediately be transferred to the coolant because of the high thermal conductivity of the SiC, thus reducing the coolant density and producing a negative reactivity feedback because of the negative void-reactivity coefficient.

For the actinide-waste-burning option, results are similar for reactor-physics assessments of SiC. The actinide additive to the SiC consists of plutonium and minor actinides recovered from spent LWR fuel. As in the Pu-burning option, gadolinium is also added. Several combinations of actinide inventory and gadolinium loading were considered. The best result in a CANDU 6 type reactor would be achieved using a full-core loading of actinide-SiC fuel containing 400 g of actinides in the outer 30 elements of the standard 37-element bundle and 60 g of gadolinium in the inner 7 elements. The actinide mix consists of

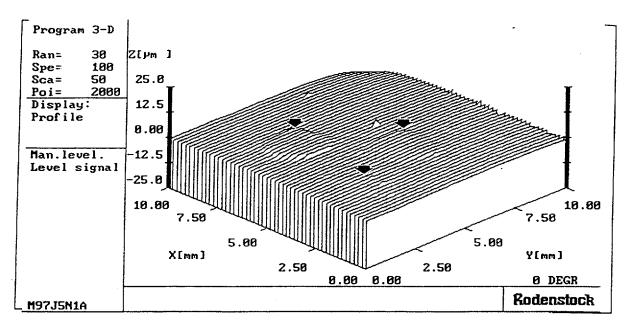


FIGURE 1. SINGLE-CRYSTAL SPINEL (100) WITH THREE IMPLANTATION DOSES AT 100°C OF IODINE AT 72 MeV (5 x 10<sup>14</sup>, 1 x 10<sup>15</sup>, AND 5 x 10<sup>15</sup> IONS/CM<sup>2</sup>).

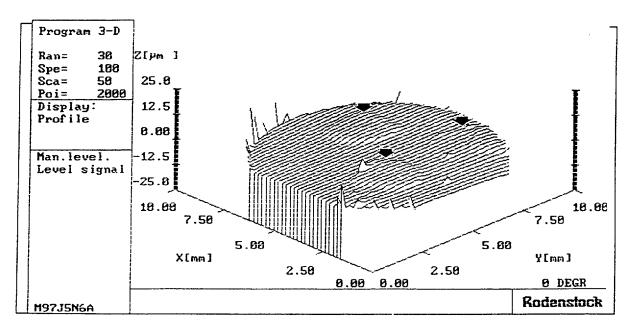


FIGURE 2. SPINEL AT 900°C, IMPLANTED WITH IODINE TO 5 x  $10^{14}$ , 1 x  $10^{15}$ , AND 5 x  $10^{15}$  IONS/CM<sup>2</sup>.

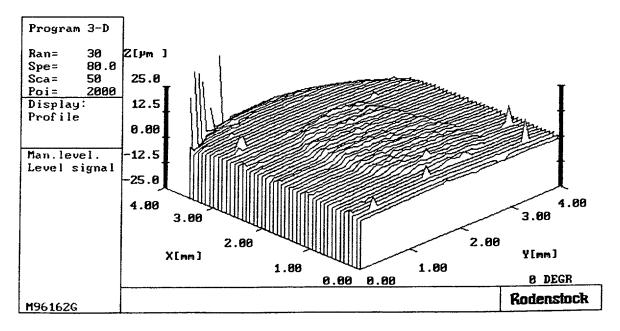


FIGURE 3. ZIRCON AT 1200°C, IMPLANTED WITH IODINE TO 1 x 1015 IONS/CM2.

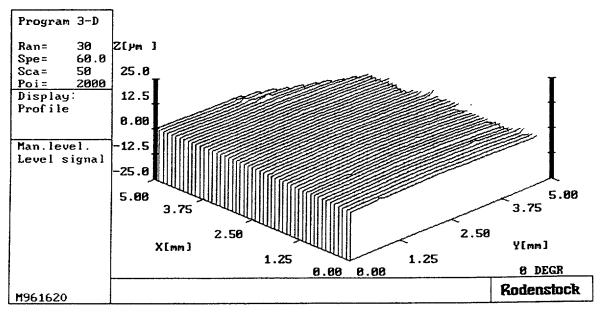


FIGURE 4. SINGLE-CRYSTAL SiC, WITH THREE DOSES OF IODINE: 1 x 10<sup>15</sup> AT 1200°C; 1.3 x 10<sup>14</sup> AND 1 x 10<sup>15</sup> IONS/CM<sup>2</sup> AT AMBIENT TEMPERATURE.

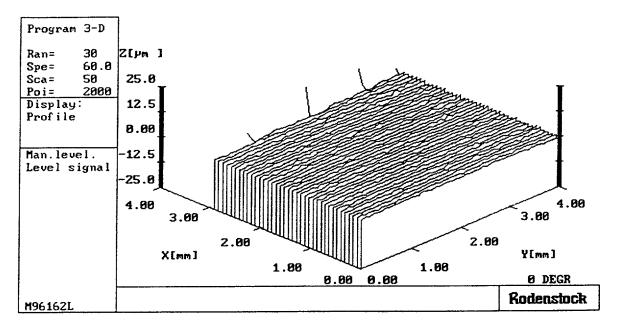


FIGURE 5. SiC AT 200°C WITH IODINE AT 72 MeV TO A DOSE OF 1 x  $10^{16}$  IONS/CM<sup>2</sup>.

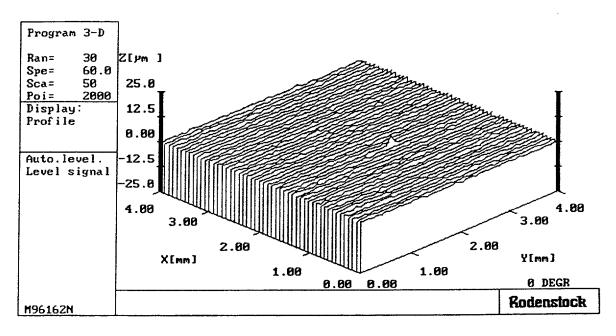


FIGURE 6. SIC AT 900°C WITH IODINE AT 72 MeV TO A DOSE OF 1x10<sup>16</sup> IONS/CM<sup>2</sup>.

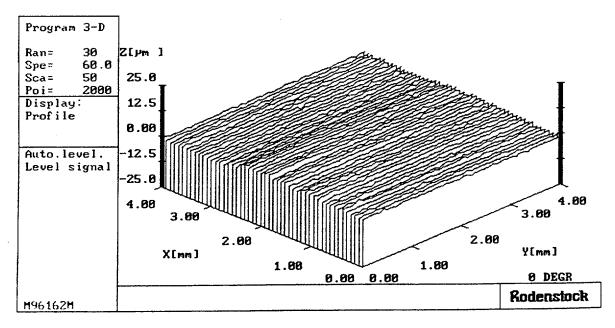


FIGURE 7. SIC AT 1150°C WITH IODINE AT 72 MeV TO A DOSE OF 1 x 10<sup>16</sup> IONS/CM<sup>2</sup>

the minor actinides and plutonium from spent PWR fuel; specifically, 356 g of plutonium and 44 g of neptunium, americium, and curium. About 60% of the initial actinide inventory is destroyed (as is 90% of the initial fissile plutonium inventory). The fuelling rate is 9.2 bundles per full-power day, resulting in the destruction of 0.68 Mg of actinides in a CANDU 6 reactor per year (assuming an 80% capacity factor). Maximum instantaneous channel power was less than 7.0 MW, and maximum instantaneous bundle power was less than 1000 kW. Fuel element power is expected to be below the current safety limit because of the high thermal conductivity of the SiC matrix. Fuel temperature coefficient is slightly positive, 12.2  $\mu$ k/°C, and the coolant-void reactivity is -4.2 mk. The power coefficient is expected to be significantly negative because of the negative coolant-void reactivity and the high heat conductivity of the SiC matrix.

#### 5. TANDEM ACCELERATOR TESTS

Since preliminary results were reported in 1995 [4], we have exhaustively tested many candidate materials,  $ZrSiO_4$  (zircon), MgAl<sub>2</sub>O<sub>4</sub> (spinel), CeO<sub>2</sub> (ceria), CePO<sub>4</sub>,  $ZrO_2$ -doped with Ca, Ce, Er or Y, as well as SiC, from ambient temperature to 1200°C, and over a wide range of doses  $(10^{14}-10^{17} \text{ ions/cm}^2)^2$ . After accelerator bombardment of each sample, the surface relief of the 3-mm-diameter beam spot was measured. Significant height of the spot above the original surface was taken as an indication that in-reactor swelling would occur. In general, results were not strongly dependent on dose—if a candidate material showed swelling at high dose, it also showed swelling at low dose. The results were also reasonably independent of temperature. The materials that showed the best results (least swelling) were SiC and  $ZrO_2$  with any of its dopants. These candidate materials did not show *any* swelling; i.e., laser profilometry could not detect any surface relief at the implantation spots. Ceria turned out to be one of the poorest candidate materials on the basis of these tests. All other candidate materials showed some surface relief, implying that swelling could be expected when used in-reactor as a fuel.

Figs. 1-7 show typical results. Figs. 1 and 2 are for MgAl<sub>2</sub>O<sub>4</sub> (spinel) at 100°C and 900°C, respectively, each showing the results for three doses  $(5 \times 10^{14}, 1 \times 10^{15} \text{ and } 5 \times 10^{15} \text{ ions/cm}^2)$ . Only the

<sup>&</sup>lt;sup>2</sup> 10<sup>17</sup>ions/cm<sup>2</sup> is approximately equivalent to a CANDU fuel burnup of 20 MW•h/kg U.

lowest dose spot (5 x  $10^{14}$ ) at 100°C shows little relief; the same dose at 900°C is clearly visible. Relief is about 1 to 2 µm. However, recent measurements with a new and recently calibrated stylus profilometer indicates that the calibration for the laser profilometer is about a factor of 5 too high. Thus relief for spinel is actually only 0.2 to 0.4 µm. Figure 3 shows a ZrSiO<sub>4</sub> (zircon) run at 1200°C to a dose of 1 x 10<sup>15</sup> ions/cm<sup>2</sup>; relief of a few micrometres is again clearly visible. Figure 4 shows a SiC single-crystal sample with 3 beam spots, 1 x 10<sup>15</sup> at 1200°C, and 1.3 x 10<sup>14</sup> and 1 x 10<sup>15</sup> at ambient temperature. There is no sign of any of the spots even though mild discolouration at the spot sites can be seen on the sample surface by eye<sup>3</sup>. Figures 5 to 7 show sintered SiC, prepared at Queen's University, at 200°C, 900°C, and 1150°C, all taken to a relatively high dose of 1 x 10<sup>16</sup> ions/cm<sup>2</sup>. Again, there is no sign of the beam spots.

#### 6. MANUFACTURING

Pressureless sintering of SiC normally requires temperatures of 1900°C and higher; we have been working with Queen's University to develop methods of fabrication at lower temperatures, more representative of  $UO_2$  sintering. In addition, fabrication tests were performed with Ce as an additive, Ce being generally considered, worldwide, as a reasonable chemical substitute for plutonium. No SiO<sub>2</sub> was used in these tests; the objective was only to determine what microstructures would develop.

Because of the largely covalent nature of the bonding in SiC, diffusion and, therefore, sintering do not occur easily. Usually, up to 5 wt %  $Al_2O_3$  and 8 wt %  $Y_2O_3$  is added to SiC powder to aid in sintering and reduce the temperatures required to achieve densities <90% TD to about 1900°C. Because this is still high, additional additives were examined in an attempt to reduce sintering temperatures even further. In particular, SiO<sub>2</sub> was examined since it was thought that this would melt and assist by *liquid-phase sintering*.

Reasonably good results were achieved with  $SiO_2$  additive; densities of >95% were achieved at a sintering temperature of 1770°C (and 87% at 1730°C).

Unexpectedly, when fabrication tests with  $CeO_2$  were done, not with the intent of lowering sintering temperatures, but rather to produce samples and observe microstructures, the  $CeO_2$  proved a better sintering aid than SiO<sub>2</sub>. At 1670°C, with the lowest amount of CeO<sub>2</sub> tested, 5 wt.%, a density of 96.5% was achieved; at 1700°C, 97%.

In the samples with 5 to 10% CeO<sub>2</sub>, a garnet phase  $(3Y_2O_3 \cdot 5Al_2O_3)$  and an oxide phase (CeAlO<sub>3</sub>) formed. At sintering temperatures  $\leq 1700^{\circ}$ C, these were very finely distributed among the 2 to 3-µm-sized SiC grains.

#### 7. COMPATIBILITY WITH WATER

Previous results [4] showed that SiC had negligible interaction with water at typical CANDU coolant conditions, 300°C and a pH of 10.3. To determine the robustness of SiC, tests were performed under stagnant acidic conditions, pH of 3 and 300°C, for 32 d, even though it is not expected that CANDU coolant would ever exist under such conditions. Eleven samples, weighing 75 to 400 mg were tested. Some interaction with the water occurred; visibly, a gray oxide layer formed and some samples broke into two or more pieces. Weight changes were small, however; samples decreased in weight by 1 to 1.5 mg.

Additional tests were performed to examine the stability of SiC as a waste under attack by groundwater. The SiC samples used were ceria-doped, also with alumina sintering aid, prepared at Queen's University. The 20 mL water sample was at 2.9 pH at 96°C, and test duration was 119 h. (Groundwater in a fuel disposal vault is not expected to have such low pH, but is a check under "extreme" conditions.) Because attack at these low temperatures was so small, weight measurements were not used; rather, the chemistry of the water was examined for dissolved components, and the surface of the sample was examined by X-ray photoelectron spectroscopy (XPS).

<sup>&</sup>lt;sup>3</sup> indicating that nothing had accidentally prevented the beam from bombarding the sample.

After the test, the water analysis indicated 0.53 mg/L Al, 11.8 mg/L Ce, and 11.3 mg/L Si. Since the water volume was 20 mL, total dissolved amounts were 0.011 mg Al, 0.24 mg Ce, and 0.23 mg Si. A similar test with  $UO_2$  has not been performed under these conditions, so direct comparison is not possible, at present. However, these amounts are very low, especially considering the high acidity level of the water, and indicate that dissolution rates are not likely to be a problem for SiC waste. The XPS analysis of the surface (2-3 monolayers) showed a change—the concentration of cerium was lower than the concentration of cerium in an unused sample.

#### 8. HIGH-TEMPERATURE CHEMICAL REACTIVITY BETWEEN SIC AND ZIRCALOY-4

Tests were performed by holding polished disc specimens of SiC and Zircaloy-4 under a light pressure and an argon atmosphere in a molybdenum cell at 1000°C, 1500°C and 1700°C. Temperatures were maintained for 1 h at 1000°C and 1500°C and for 15 min at 1700°C. Two types of SiC specimens, fabricated at Queen's University, were used; both contained alumina as a sintering aid, and one contained titania, the other ceria. Extra carbon had been added to all specimens during sintering to ensure that no free Si was present in the final products. After cooling and sectioning, the specimens were examined by optical and scanning electron microscopy to study the extent of interaction.

Although no significant interaction occurred at 1000°C, at 1500°C there was clear evidence of a diffusion-based reaction to form ZrC and free Si. Diffusion of free Si into the Zircaloy disc led to the formation of a molten Zr-Si-rich eutectic phase. This reaction was more pronounced in the 1700°C test specimens, where the amount of molten eutectic phase was sufficient to cause partial dissolution of the Mocell sidewalls.

The test results indicate that formation of a molten Zr-rich phase could conceivably occur during a hypothetical reactor accident at temperatures significantly lower than the melting point of unoxidized Zircaloy cladding (1760°C). This possibility is of concern because it would allow further accelerated attack on the SiC carrier material, thereby allowing any fission products that had exsolved from the plutonium phase to be released. However, UO<sub>2</sub> interaction with Zircaloy begins at about 1200°C [12], and it dissolves into molten Zircaloy at 1760°C. The rate and extent of UO<sub>2</sub>-Zircaloy interaction depends on the amount of oxygen present; however, it seems that the SiC interaction with sheath is, at least, no worse than the UO<sub>2</sub> interaction. Tests using SiC pellets clad with Zircaloy-4 sheathing, for more direct comparison are being evaluated.

#### 9. WASTE-DISPOSAL

Considerations of SiC-based fuel as a waste form can be summarized:

- The SiC matrix itself, with sintering aids used to date will not generate long-lived activation products in-reactor. Care needs to be taken to minimize the impurity levels of N and Cl to eliminate concerns about their activation products <sup>14</sup>C and <sup>36</sup>Cl.
- Silicon carbide itself is extremely stable and resistant to corrosion. Its performance as a waste form
  will depend on its ability to encapsulate and retain high-burnup plutonium-containing particles. It is
  likely that the plutonium-rich phase will remain encapsulated, and hence be protected from leaching.
  This needs to be verified by post-irradiation examination and leach-testing of irradiated inert-matrix
  fuel materials.

#### 10. CONCLUSION

Practical realization of an inert-matrix fuel for burning plutonium or actinide waste is 10 to 20 years off, at least partly because the incentive, while real, is not urgent. Silicon carbide and doped  $ZrO_2$  are the 2 candidate materials that most clearly pass the accelerator-simulation test, but tests in-reactor are the final arbitrator. Compatibility of SiC with water does not seem to be an issue, either as a fuel for compatibility with coolant or as a waste form for compatibility with groundwater. Compatibility of SiC with Zircaloy

does not seem to be worse than compatibility of SiC with  $UO_2$ -Zircaloy interactions. A small amount of melt phase above 1700°C may be acceptable in safety evaluations. A concern for the actinide burning application may be that SiC is a carbide rather than an oxide, and americium carbide, which could form when the actinide waste is incorporated in the SiC, is unstudied, and mounting a program to investigate it will be expensive because americium is highly alpha-active (MATZKE Hj., private communication, 1996). Americium oxide, on the contrary, has already been studied. For plutonium burning, and for actinide burning if the americium carbide issue can be resolved, SiC appears to be an excellent candidate material.

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## INERT MATRIX — CERMET FUELS: TOWARD OPTIMIZED UTILISATION OF Pu IN A PWR NEUTRONIC STUDIES — THE TANOX EXPERIMENT

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#### Abstract

In order to address the new issues linked to the checking of the nuclear programme and the abandonment of the fast reactor system in France, the Nuclear Reactor Division (DRN) of the CEA launched a study programme with the aim of increasing the plutonium load in pressurized water reactors.

## **INTRODUCTION**

The nuclear industry landscape has considerably changed since the 1960s: the development of the fast breeder reactor system was first curtailed, then progressively left in abeyance, which resulted in the accumulation of large amounts of plutonium. A temporary solution for these stocks of plutonium consisted in using them in pressurized water reactors (PWRs). However, for safety reasons, essentially related to the great sensitivity of the cores of these reactors - due to a very negative moderating coefficient that makes cold hazards particularly difficult to control -, the safety authorities decided to limit core loading of Pu in its conventional, MOX (Mixed Oxide) form to only 30 %.

On the other hand, for reasons linked to proliferation resistance and economics, increased burnup of these fuels is highly desirable.

Although 20 reactors of the French nuclear power plants are authorized to operate with a load of 30 % MOX, only 17 currently use this fuel, while eight units are soon to receive authorization to operate using this fuel.

100 % MOX core loading could obviously be envisaged [1], [2]; however, this would mean evolving towards higher moderation ratios of nearly 4 or 5 to enable efficiently controlling the reactor core, and this would entail making considerable changes to both the core array and more generally to the whole architecture of the core. Another way of considering the problem of increasing plutonium consumption and eliminating the conversion of U 238 to Pu 239 would be to use plutonium not in MOX form, but on a neutronically inert support, otherwise known as an inert matrix.

Very rapidly, analyses revealed the importance of microstructures in the nature of the fuel. Theoretical development, supported by experiments performed in the SILOE reactor at Grenoble, France, resulted in a better determination of the role played by the size of the grains and their interlinks in the retention or release of fission gases (3, 4, 5, 6)

Obtaining high burnups also poses numerous problems with respect to the resistance of the Zircalloy alloys used for fuel cladding: corrosion, thermomechanics, aging under irradiation, etc.

An experimental tool that would permit rapidly achieving high burnups was required to enable testing various microstructures and different compounds. This is why the TANOX device was designed and installed in the SILOE reactor at Grenoble.

## I. <u>CHOICE OF MATRICES</u>

Studies were oriented towards two types of composite fuels, CERCER in which the ceramic fuel (UO<sub>2</sub>, MOX, PuO<sub>2</sub>) is embedded in a ceramic matrix (spinel, magnesia), or CERMET, in which the fuel is dispersed in a metallic matrix.

From the neutronic point of view, the ceramics implemented are transparent to neutrons and do not require particular analysis. This, however, does not hold true of the metals and, as can be observed in Figure 1 and Table 1, their responses to reactivity can vary widely.

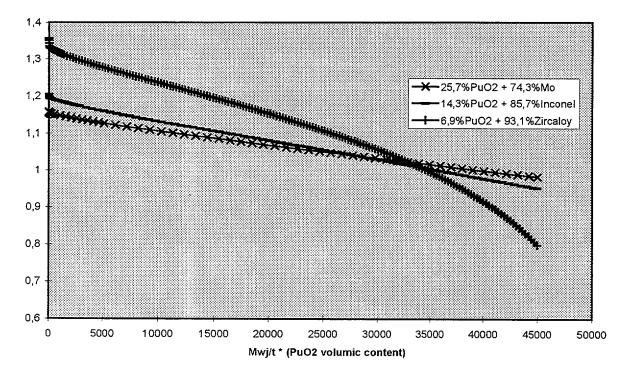


Figure 1 : PuO<sub>2</sub>-based CERMET fuels - Reactivity swing for different metals

## Table 1 : Kinetic coefficients for several fuels

Fuel	B	OL	E	OL
	Doppler pcm/°c /	Drain. coefpcm	Doppler pcm/°c /	Drain. coef pcm
UO <sub>2</sub>	-2.00	-61429	-3.39	-94613
UO <sub>2</sub> +INCONEL	-1.99	-33029	-3.29	-63650
UO <sub>2</sub> +MOLYBDEN	-1.68	-45168	-2.59	-75969
UO <sub>2</sub> +Zry	-1.72	-53536	-3.55	-181167
MOX+INCONEL	-2.7	+4953	-2.98	+3281
MOX+MOLYBDEN	-1.93	+11618	-1.99	+13434
MOX+Zry	-2.97	-13471	-3.42	-28877
PuO <sub>2</sub> +INCONEL	-1.04	+4183	-1.10	-3841
PuO <sub>2</sub> +MOLYBDEN	-0.71	+9481	-0.57	-8922
PuO <sub>2</sub> +Zry	-1.09	-9873	-1.80	-142821
PuO <sub>2</sub> +ThO <sub>2</sub>	-3.30	-16690	-3.47	-31704

# Table 2 : metal volumic fraction=0.5, cycle lenght=18 months, fissile material content(%)

Matrix	UO <sub>2</sub>	CERMET	CERMET	CERMET	CERCER
	_	UO <sub>2</sub>	MOX	PuO <sub>2</sub>	Pu/Th
**UO2**	3.7%				
Inconel		14.1%	32.5%	14.3%	
Molyb.		18.7%	50.5%	25.7%	
Zry		7.5%	18%	6.9%	
**Th**					14%

Table 2, in particular, gives an overview of the contents in fissile material required by the various composites to ensure a burnup (BU) of 120 Gwd/t up to unloading.

The logical choice should be directed towards a metal that is transparent to neutrons, such as Zircaloy, which is able to assure the desired cycle with the lowest plutonium content and the highest potential reactivity value (Figure 1).

However, for reasons of availability and know-how, molybdenum was selected for the first experiments in the TANOX device on CERMETs and the spinel MgAl<sub>2</sub>O<sub>4</sub> for those on CERCERs.

## **II. <u>THE TANOX DEVICE</u>**

The TANOX device (Figure 2) is a rotating cylinder that has six positions. The fuel rods are about six millimeters in diameter and are pierced by a central hole to allow the passage of the thermocouples that enable controlling the experiment through the core temperature of the fuel. The device turns regularly to ensure that all the rods have the same combustion rate.

Table 3 : Release fraction of 85Kr for the TANOX pins

Fuel	BU GWd/t	85Kr release 1580°C 30 mn	85Kr release 1700°C 30 mn
U02	19.58	0.1	0.4
CERCER	40.3	0.25	0.51
CERMET	55.4	0.12	0.17

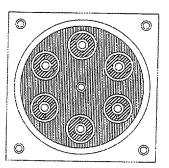
The device is contained in a box that can be moved from actual contact with the core of the SILOE reactor to a distance of little less than twenty centimeters away from it (Figure 2). The device permits achieving burnups in the composite pellets of about 0.5 Gwd/t per day of operation at full power of the SILOE reactor.

## III. FIRST TANOX EXPERIMENTS USING CERCER AND CERMET RODS

The TANOX 2 experiment was performed on spinel rods (64 %  $MgAl_2O_4 - 36$  %  $UO_2$ ) (% in volume) with 19.6 % U235 enrichment.

The CERMET used was 64 % Mo, 36 % UO<sub>2</sub>, also with 19.6 % U235 enrichment.

## Fuel Pin Location



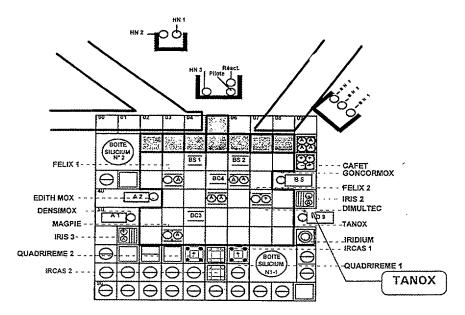


Figure 2 : SILOE core and TANOX location

# Table 4 Some characteristic of mass balance

Month	UO <sub>2</sub>	MOX	CERMET 40% UO <sub>2</sub>	CERMET 40% MOX	CERMET Pu
12	1	0.54	1.44	0.81	1.17
18	1.19	0.62	1059	0.87	1.17
24	1.31	0.69	1.67	0.93	1.19

## Initial Reactivity

## Enrichment

Month	$UO_2$	MOX	CERMET 40% UO <sub>2</sub>	CERMET 40% MOX	CERMET Pu
12	3,19	8.26	9.49	17.65	6.74
18	4.49	11.28	14.15	27.38	10.39
24	5.88	14.14	19.02	37.21	14.22

## Initial Fissile content (kg)

Month	UO <sub>2</sub>	MOX	CERMET 40% UO <sub>2</sub>	CERMET 40% MOX	CERMET Pu
12	750	1938	732	1262	1063
18	1056	2647	1091	1958	1661
24	1382	3318	1466	2660	2304

## BU in at%

Month	UO <sub>2</sub>	MOX	CERMET 40% UO <sub>2</sub>	CERMET 40% MOX	CERMET Pu
12	67.4	34.8	88.9	71.2	97.5
18	76.1	41.8	95.4	74.8	97.9
24	80.8	46.7	98	77	97.4

BU in weight (kg)

Month	UO <sub>2</sub>	MOX	CERMET	CERMET	CERMET Pu
			40% UO <sub>2</sub>	40% MOX	
12	505.3	378(Pu9)	650	542	653(Pu9)
18	803.5	620.5	1041	885	1018
24	1117	869.5	1438	1238	1399

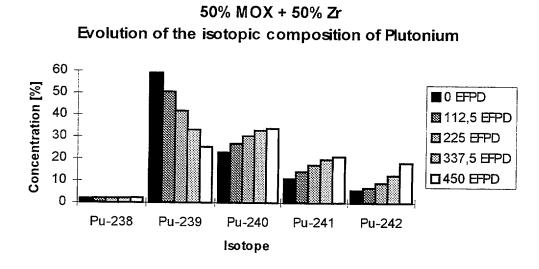


Figure 3 : Plutonium consumption in a MOX CERMET

Table 3 shows the excellent behavior of the CERMET as regards the release of fission gases.

This was confirmed when the cladding was opened (Figures 4 and 5). The behavior of the CERCER was disappointing: the composite evidenced considerable swelling and generalized pellet restructuring, whereas the integrity of the CERMET was preserved. This is why a second series of irradiation tests was launched, using a CERMET composite: the aim of TANOX CCE, (a composite fuel comprising erbium) was to reach, or even exceed, a burnup of 100 Gwd/t, to examine the behavior of the materials, the fuel, the matrix, and the absorbers, and to validate the consumption kinetics of erbium.

The composite was formed of 80 % Mo, 20 % UO<sub>2</sub>, with 40 % U235 enrichment; the ceramic also included 2 %  $Er_2O_3$  in weight.

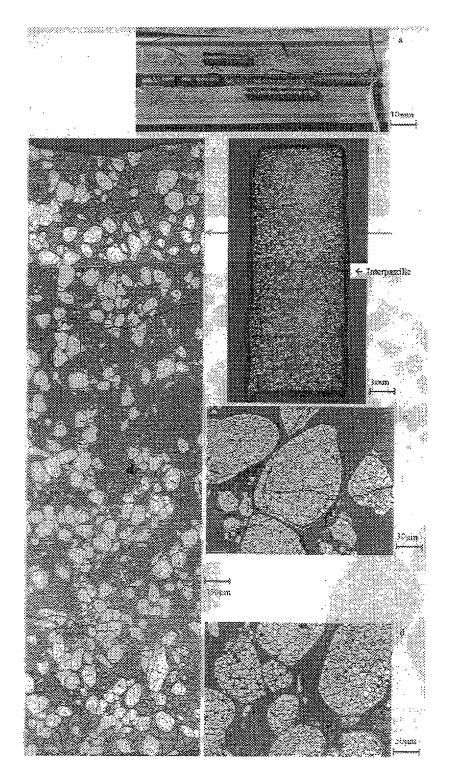
The test series ended in December 1997 and the results obtained at 136 Gwd/t confirmed the following points:

- The excellent dimensional stability of the CERMET under irradiation,
- The very slight degassing revealed in the TANOX 2 experiment,
- The excellent thermal conductivity of the composite which permits core temperatures under PWR conditions of about 420° C (the TANOX CCE irradiation test was performed at 350 W/cm, i.e., a temperature of about 530° C),
- The good agreement of the theoretical models provided by calculation of the composites' thermal conductivity (Maxwell-Euken) with the experimental results.

# IV. EVALUATION OF THE CYCLE

Using the simplified model of a PWR-type assembly, we determined various parameters, such as enrichment, initial fissile material balance, etc. (Table 4) for different CERMET (Zircaloy) composites for cycles of 12, 18, and 24 months.

This clearly showed that Pu CERMETs, as well as MOX CERMETs, are excellent plutonium consumers and that, in addition (Figure 3), good compution of Pu 239 is achieved without too high an increase of the Pu 240 concentration.



# Figure 4 : CERCER fuel after irradiation

a - aspect of the pellets after opening of the pin

b - macrography of two stuck pellets

c - fuel microstructure: transversal section of a pellet

d - presence of a strip surrounding the  $UO_2$  nodules at the pellet edge (after chemical attack)

e - absence of strips in the pellet center (after chemical attack)

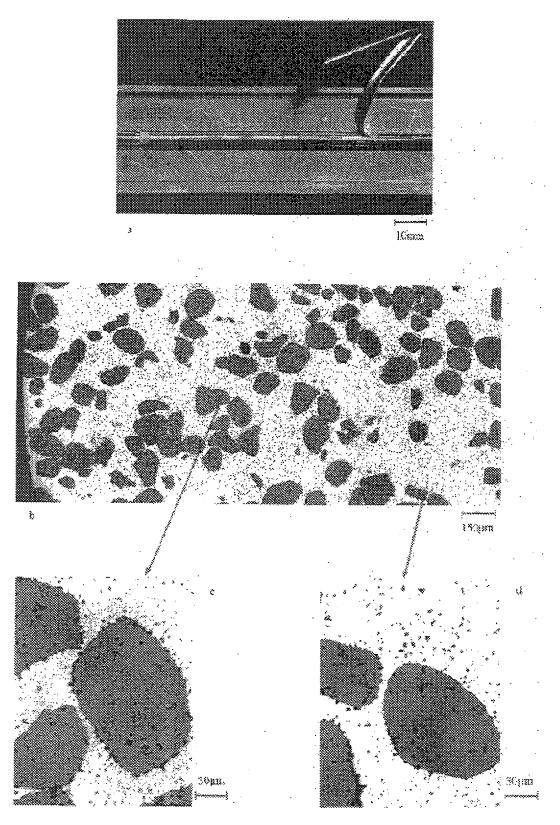


Figure 5 : CERMET fuel after irradiation

- a aspect of the pellets after opening of the pin
- b fuel microstructure along a radius
- c & d fuel microstructure with  $UO_2$  nodules in the Molybdenum matrix

# **CONCLUSION**

CERMET composites have numerous advantages not only as regards their behavior under irradiation and their behavior as cold fuels, but also as Pu consumers. However, their low  $\beta$ eff will make these cores very sensitive to accidents such as rod ejection; in addition, their very hard spectrum reduces the efficiency of soluble boron and their low Doppler coefficients are liable to be of little effect in countering power excursions.

As these 100 % Pu CERMET assemblies are too delicate to be used from the standpoints of reactivity control and safety coefficients, studies should be oriented towards heterogeneous assemblies, in which conventional UO<sub>2</sub> fuels would provide the necessary margins for the  $\beta$ eff and safety coefficients. It is why the APA (Advanced Pu fuel Assembly) which is presented in a companion paper (7, 8, 9, 10) was defined

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# APA: U FREE PU PIN IN A HETEROGENEOUS ASSEMBLY TO IMPROVE PU LOADING IN A PWR — NEUTRONIC, THERMO-HYDRAULIC AND MANUFACTURING STUDIES

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## Abstract

After having presented the specific context of France with respect to the fuel cycle and reprocessing, the problem of plutonium fuel utilization is posed.

If one of the solutions, a pressurized water reactor (PWR) with an increased moderation ratio seems possible, it entails making excessive changes to the reactor, the control systems, and the general architecture of the steam supply system.

Another solution consists in modifying the fuel itself so as to eliminate conversion on  $^{238}$ U by using plutonium (Pu) in a neutronically inert matrix. However, the disadvantage of this type of fuel is that it has very low Doppler and draining coefficients and a very small delayed neutron fraction. To enable using these fuels, a heterogeneous assembly has to be defined, in which standard UO<sub>2</sub> rods provide the physical properties required to ensure acceptable safety coefficients.

# **Introduction**

In view of the many changes that have occurred in the world nuclear landscape, the problem of managing our stocks of plutonium and irradiated fuels has become acute.

The following notions have been highlighted by the international trends expressed in numerous forums, particularly at the IAEA:

- Resistance to proliferation,
- Preservation of natural resources,
- Minimization of the volumes, quantities, and radiotoxicity of wastes arising from the nuclear industry.

All these points are underpinned by the crucial concept of public acceptance.

It is obvious, however, that a nuclear industry can only continue to exist if it produces a kWh that is competitive compared to other sources of energy.

In France, a plutonium reprocessing and recycling policy was set up very early to prepare fuel for the future breeder reactor system. However, a certain number of socio-economic factors led first to the checking of the development of the breeder system, then to its being placed in abeyance.

The reprocessing facilities that were constructed permit producing a high-quality mixed uranium-plutonium fuel (MOX), and were therefore oriented towards the production of a fuel intended for the cores of PWRs.

However, using plutonium in a conventional core poses numerous problems, such as, in particular, the great affinity of Pu isotopes for thermal neutrons, which results in a hardening of the neutron spectrum and considerably decreases the efficiency of control systems.

In addition, the moderator coefficient becomes very negative and this, in certain classes of accidents, such as cold hazards, steam line break at full power, or spurious secondary valve opening on shutdown at hot zero power, leads to reactivity consequences that are difficult to control. This is why the safety authorities have limited core loading of MOX fuel to 30 %.

Twenty reactors have been granted authorization to use MOX fuel, although only 17 currently do so, and this number will soon be increased to 28. However, given the fact that the French utility, EdF, has reprocessed only the amount of irradiated fuel needed for the fabrication of the MOX fuel authorized in these reactors, it is obvious that the stocks of plutonium and non-reprocessed fuels will continue to increase.

Various solutions have been proposed, notably highly moderated reactors: these would permit better thermalization of the neutrons, resulting in increased control system efficiency and a considerable decrease in the absolute value of the moderator coefficient, and would therefore allow 100 % MOX loading (1, 2).

However, this solution would entail making significant changes to the architecture of the core and control systems, and would lead to a considerable reduction of the power density of the core.

Another solution would mean modifying the fuel and using plutonium or MOX combined with neutronically inert material, so as to consume the plutonium while reducing or eliminating  $^{238}$ U to  $^{239}$ Pu conversion (3).

Unfortunately, it was demonstrated that homogeneous assemblies using this fuel made the core uncontrollable, because of a small  $\beta$ eff, a very low Doppler coefficient and, occasionally, a draining coefficient liable to be positive. The solution would be to supply the complement to these safety parameters by means of a heterogeneous load in which standard UO<sub>2</sub> fuel would provide the kinetic coefficients required for safety. This is the APA or Advanced Pu fuel Assembly (4,5,6).

# The APA Assembly

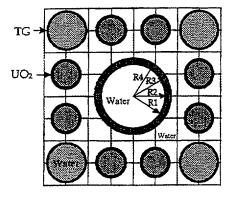
Externally, the APA assembly is strictly identical to a conventional assembly and can replace a standard assembly in a load.

It comprises 120 standard  $UO_2$  rods, 24 guide tubes, one instrumentation tube and 36 APA rods (Figure 1). The latter are big, very thin annular rods, 25.2 mm in diameter and 1.26 mm

thick, with inner and outer cladding to permit inside circulation of water. The material used is a 32.6 %  $PuO_2$ , 63.9 % cerium (CeO<sub>2</sub>), and 3.5 %  $Er_2O_3$  ceramic-ceramic composite (CERCER) (Table 1).

The advantage of the slight thickness of the ceramic is to increase the "apparent" thermal conductivity of the rod.

In addition, the inner water provides both an increase in the local moderating ratio ( $\sim 6$ ) and significant internal cooling.



R4 (mm)	=	11.5	
R3 (mm)	Ξ	11.0	] 1.261 mm
R2 (mm)	=	9.739	J 1.201 mm
R1 (mm)	=	9.239	
VpuO <sub>2</sub> /Vcomb	=	0.23	

Local moderation ratio = 5.96 (annular pin) Global moderation ratio = 3.47

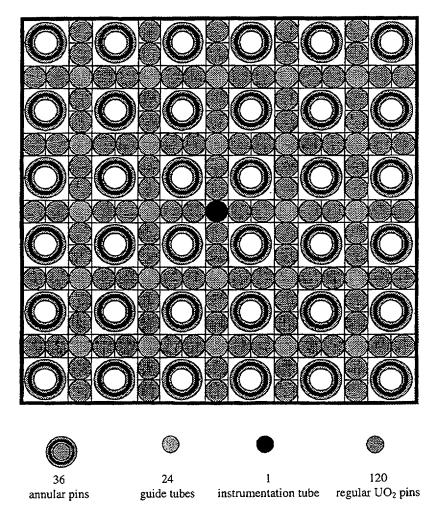


Figure 1 - The APA assembly

Table 1 : fuel composition

		$= 10.21 \text{ g/cm}^3$	120 standa	rd rods		
Uranium Percent mass	<sup>235</sup> U 2.2	<sup>238</sup> U 97.8	Fuel assembl	y $\frac{Pu}{U+F}$	$+^{241}Am$ $Pu+^{241}Am =$	0.1037
	APA d	$= 7.85 \text{ g/cm}^3$	36 annula	ır rods		
Material	PuO <sub>2</sub>	CeO <sub>2</sub>	Er <sub>2</sub> O <sub>3</sub>			
Percent mass	32.6	63.9	3.5			
Plutonium	<sup>238</sup> Pu	<sup>239</sup> Pu	<sup>240</sup> Pu	<sup>241</sup> Pu	<sup>242</sup> Pu	<sup>241</sup> Am
Percent mass	3.3	41.7	28.7	14.5	10.7	1.1
Cerium	<sup>140</sup> Ce	<sup>142</sup> Ce				
Percent mass	88.8	11.2				
Erbium	<sup>162</sup> Er	<sup>164</sup> Er	<sup>166</sup> Er	<sup>167</sup> Er	<sup>168</sup> Er	<sup>170</sup> Er
Percent mass	0.1	1.5	22.9	22.9	27.2	15.1

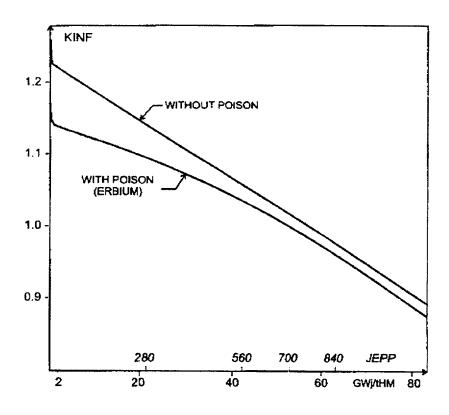


Figure 2 : reactivity swing with and without poison

Figure 2 shows evolution calculations for constant boron (600 ppm) made using the APOLLO 2 computer code. A considerable increase in erbium reactivity control can be observed, even if the residual penalty remains significant<sup>\*</sup>. The Xe effect at saturation is about 1760 percent milli k (pcm) ( $10^{-5} \Delta k/k$ ).

<sup>\*</sup> This is not a major economic factor insofar as the cost of Pu is already included in the cost of uranium reprocessing.

	BOL	EOL
		1120 EFPD
Annular rod (36)		
Linear power density (W/cm)		
Average	1010	664
Maximum	1010	681
Waxinum	1004	001
Power density (W/cm <sup>3</sup> )		
Average	1230	809
Maximum	1320	829
Surface heat flux (W/cm <sup>2</sup> )		
Avearge	77	51
Maximum	83	52
$UO_2$ standard rods (120)		
Linear power density (W/cm)		
Average	87	191
Maximum	91	196
Power density (W/cm <sup>3</sup> )		
Average	162	356
Maximum	170	366
Surface heat flux (W/cm <sup>2</sup> )		
Average	33	76
Maximum	35	76

# Table 2 : averaged and maximum integrated power levels

# **Temperature Assessment**

As the FLICA IV code was modified to take internal and external cooling of the rod into account, it can be observed (Table 2) that power densities are very high in the APA rods, but that, as a result of their extensive contact surface, the temperatures of both the fuel and the cladding remain very cold (Table 3).

Table 3 : Temperature distribution - power density 1320W/cm<sup>3</sup>

	Interna	l Clad		Fuel	· · · · · · · · · · · · · · · · · · ·	Extern	al clad	
Internal	Coolant	Fuel	Internal	Maximum	External	Fuel	Coolant	External
Water	Side	Side	Rod Side		Rod Side	Side	Side	Water
T <sub>W,i</sub>	T <sub>C,1</sub>	T <sub>C,2</sub>	T <sub>F,2</sub>	T <sub>FM</sub>	T <sub>F,3</sub>	T <sub>C,3</sub>	T <sub>C,4</sub>	T <sub>W,e</sub>
299°C	322°C	345°C	470°C	544°C	513°C	363°C	337°C	309°C

 Table 4 : Heavy Nuclei Balance (kg) for 1 TWh

	UO2 (3.7%)	APA
235U	-84.74	-22.18
238U	-94.41	-46.94
Total plutonium	+32.67	-76.52
Minor Actinides	+2.73	+9.04
Plutonium + Americium	+35.40	-67.40
SWU	14816	3054

# Material Aspects

The 120 UO<sub>2</sub> rods are enriched 2.2 % with <sup>235</sup>U. The APA rods contain plutonium known as 2G, which comes from MOX reprocessing in PWRs, discharged at 33 GWd/t, cooled three years and stored two years after reprocessing for a cycle of four times 280 EFPD in a 900 MWe PWR. Plutonium consumption is 48.6 % with about 80 % consumption for <sup>239</sup>Pu and 36 % consumption for <sup>241</sup>Pu, offset, however, by the production of + 36% of <sup>242</sup>Pu. Table 4 shows for an equivalent produced energy, the mass balances of a PWR using UO<sub>2</sub> fuel and one using APA fuel.

# Kinetic Coefficients

Table 5 also gives a comparison of the kinetic coefficients of conventional loading and APA loading.

The role of plutonium can be clearly observed to affect the efficiency of the boron, the moderator coefficient and the  $\beta$ eff. However, the values obtained remain within an acceptable range.

# **Fabrication Materials**

The first fabrication tests using classic powder metallurgy processes gave excellent results and rings meeting specifications were manufactured in the laboratory.

However, the slight thickness involved required very precise fabrication in view of the fact that grinding on 1.2 mm thick rings is extremely costly and results in numerous rejections. Vibrocompacting techniques should be able to be implemented to improve fabrication (7, 8).

# Possible Scenario

Considering the situation in France after the year 2000, twenty-eight (63 GWe) PWRs, recycling 30 % 1G MOX (4 batches), 12 MOX and 18  $UO_2$ , and four 1450 MWe APWRs using APA assemblies would be enough to absorb the second generation of Pu (2G). These new reactors (APWRs) would replace 6 decomisionned 900 MWe.

Fuel assembly parameters	APA	UO <sub>2</sub> (3.7%) PWR
Boron worth (pcm/ppm)	,	
BOL, full power, xenon = 0, $C_B = 550$ ppm	- 4.9	- 8.9
$C_{\rm B}$ = 1450 ppm	- 4.7	
Moderator temperature coefficient (pcm/°C)		
BOL, T = 296°C, $C_B = 1500 \text{ ppm}$	- 17.3	+ 6.4
$C_B = 600 \text{ ppm}$	- 31.9	- 15.4
EOL, T = 296°C, $C_B$ = 10 ppm	- 36.1	
Doppler coefficient (pcm/°C)		
BOL, 650 to 286°C, $C_B = 1500 \text{ ppm}$	- 2.0	- 2.6
Overall draining (pcm)		
BOL, water at 1500 ppm to without water	- 13 630	- 70 000
Core parameters		
Moderator temperature coefficient (pcm/°C)		
BOL, 1450 ppm	- 24.9	- 21.0
EOL, 10 ppm	- 57.8	- 62.0
Effective $\beta$ (pcm)		
BOL	370	595
EOL	400	522

# Table 5 : Comparison of the physical parameters

 Table 6 : Medium Term operating Scenario for the French Reactors

Type of PWR	Fuel	Number	Type of	Enricment	Tonnage in	Unloading Burnup
[Net Power		of	Managemen	with <sup>235</sup> U	HM/yr unit (t-1)	(GWd/tonne HM)
MW (electric)]		Units	t	[Total Pu	(Number of Fuel	
			(Annual	Content (%)]	Assemblies)	
			Cycles)			
APWR (1450)	APA	4	1/4	1.0 [10.4%]	14.763 t U - 1.708 t Pu	72
					(60)	
CPY (920)	UO <sub>2</sub> -MOX	28	1/4	3.7 [6.5%]	12.923 t U - 5.538 t	43
					UPu	
P4 (1300)	UO <sub>2</sub>	20	1/3	3.1%	(28) (12)	33
					34.387 tonne U	
N4 (1450)	UO <sub>2</sub>	4	1/3	3.25%	(64)	33
					36.525 tonne U	
					(68)	

.

 Table 7 : Annual balance of HM in the reactors

	Loading	Plutonium Production			
1. Uranium metal:	1254.7 tonne	1. 12.73 tonne of first			
2. U-Pu metal:	155.1 or10.08 tonne of first generation	generation			
Pu		2. 7.05 tonne of second			
	at 70% of fissile Pu (176 tonne of	generation			
MOX)		3. 3.00 tonne of "third"			
3. Plutonium metal:	6.83 tonne of second generation at 55%	generation			
of fissile Pu					
Theorically, when in	equilibrium, there remains (no losses):	L			
12.73 - 10.08 = 2.65 tonne of plutonium of first generation (70% fissile Pu)					
7.05 - 6.83 = 0.3	7.05 - 6.83 = 0.22 tonne of plutonium of second generation (55% fissile Pu) $\Rightarrow$ Total				
mixing 5.87 tonne					
3	.00 tonne of plutonium of "third" generation	on (70% fissile Pu)			
(48% fissile Pu)					

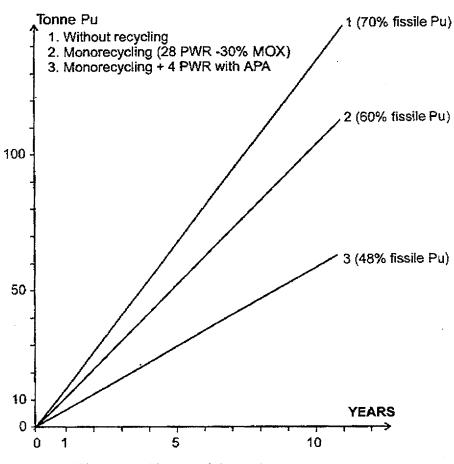


Figure 3 : Change of the Pu inventory (63 GWe)

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The results of this scenario are reported in Table 6. It should be noted that the Pu from APA cycling is very degraded (28 % fissile Pu). The combination of these three restricted qualities (Table 7) results in 48 % fissile Pu. This quality can be used in another APA cycle provided 235U enrichment is increased to something up to 2.5 % in the UO<sub>2</sub> rods (instead of 2.2 % in the example selected here);

The results of the various scenarios, once-through, single cycling, single cycling + APA are reported in Figure 3. The advantage of an APA solution is apparent.

# **Conclusion**

Obviously, a great many studies have yet to be performed in all domains -mechanics, neutronics, thermal-hydraulics, safety, etc.- to enable completely defining an industrial assembly. However, the concept presented here has the generic advantage of featuring a significant degree of freedom with respect to uranium and plutonium for adjusting control and kinetic coefficients to acceptable values, while preserving high plutonium utilization potential. This type of solution, possible in the relatively short term, could, in addition, provide end of cycle Pu for CAPRA type reactors.

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# LWR/HWR SYNERGISTIC FUEL CYCLES

(Session 6)

# Chairperson

H.S. Park Republic of Korea



#### THE DUPIC FUEL CYCLE SYNERGISM BETWEEN LWR AND HWR

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#### Abstract

The DUPIC fuel cycle can be developed as an alternative to the conventional spent fuel management options of direct disposal or plutonium recycle. Spent LWR fuel can be burned again in a HWR by direct refabrication into CANDU-compatible DUPIC fuel bundles. Such a linkage between LWR and HWR can result in a multitude of synergistic effects, ranging from savings of natural uranium to reductions in the amount of spent fuel to be buried in the earth, for a given amount of nuclear electricity generated. A special feature of the DUPIC fuel cycle is its compliance with the "Spent Fuel Standard" criteria for diversion resistance, throughout the entire fuel cycle. The DUPIC cycle thus has a very high degree of proliferation resistance. The cost penalty due to this technical factor needs to be considered in balance with the overall benefits of the DUPIC fuel cycle. The DUPIC alternative may be able to make a significant contribution to reducing spent nuclear fuel burial in the geosphere, in a manner similar to the contribution of the nuclear energy alternative in reducing atmospheric pollution from fossil fuel combustion.

#### 1. INTRODUCTION

The predominant nuclear reactor types in the current world market are LWR and HWR, and this trend will likely continue for the foreseeable future. Under such a trend, the question of "how to manage the spent fuel discharged from those reactors" will remain a key factor to be considered, as part of the sustainable supply of nuclear energy in the future. The conventional backend fuel cycle has evolved into two different options, depending upon national policies: either direct disposal in deep geological formations, or reprocessing of spent LWR fuel for MOX fuel recycle in LWR (or FBR). However, the decision on which of these two options to pursue is still pending for many other countries, which have a "wait and see" position.

The DUPIC fuel cycle offers an alternative to these conventional options when looking at the possibility of burning the spent LWR fuel again in HWR, by taking advantage of the high neutron economy of CANDU-type heavy water reactors [1]. The spent LWR fuel is transformed into HWR-type DUPIC fuel by direct refabrication, without any separation of nuclear materials. The technical implications associated with the DUPIC fuel fabrication result in some special features: the fabrication processes are highly radioactive requiring remote systems for operation and maintenance; therefore, these processes are in natural compliance with the "Spent Fuel Standard" for proliferation resistance [2]. It is to be noted that the basic premise of the DUPIC fuel development is to make the DUPIC fuel compatible with the existing HWR system. This approach is consistent with the reality that the LWR and HWR will remain as the major reactor types in the world, for a substantial time into the future. Most of the R&D efforts for the DUPIC fuel cycle are directed therefore, to fuel refabrication and verification of compatibility with the CANDU design. The DUPIC technology program, which is now in full swing at KAERI in cooperation with several international partners, can thus become a useful test-bed, toward a potential synergism between LWR and HWR.

To assess the effects of synergism from the DUPIC linkage of LWR to HWR, a multitude of factors will have to be taken into account: natural uranium saving for the HWR, removal of spent LWR fuel, environmental impacts, etc., as well as the additional burnup achievable using DUPIC fuel in HWR [3]. A key question in terms of economics will be whether these synergistic effects could outweigh the costs required for implementing the DUPIC fuel cycle.

#### 2. DUPIC SYSTEM ANALYSIS

#### 2.1. Parameters for DUPIC Linkage between LWR and HWR

The residual fissile content of spent LWR fuel is slightly over 1.5 wt. %, depending on the burnup achieved in the reactor operation. It is a somewhat higher value at lower LWR burnups, and it can vary by a few percent between 35 MWd/kgU and 50 MWd/kgU. This level of residual fissile enrichment represents more than two times that of natural uranium, which is 0.71%. According to physics calculations done at KAERI with this residual fissile content, the discharge burnup attainable in a HWR could reach 18 MWd/kgU or 13 MWd/kgU respectively, for the corresponding LWR burnups [4]. For an average LWR burnup of 42.5 MWd/kgU, an additional 15.5 MWd/kgU can be expected using the DUPIC synergism, or 37% more power than the once-through case. After burning the DUPIC fuel in the HWR, the fissile content of the spent fuel decreases to 0.63% and 0.78% respectively, for the low- and high-burnup LWR cases, or roughly to the level of natural uranium on the average, by the DUPIC linkage.

#### 2.2 DUPIC Material Balance

Based on consideration of the two typical LWR burnup cases of 35 MWd/kgU and 50 MWd/kgU, it is possible to match the material flow between LWR and HWR and to determine the ratio of reactors (LWR/HWR) required, on the assumption of the same capacity for both LWR and HWR units. The ratio is 2.5:1 for the lower discharge burnup of 35 MWd/kgU and 4.8:1 for the higher burnup of 50 MWd/kgU, or 3.7:1 for the average burnup of 42.5 MWD/kgU.

In terms of nuclear material flow, the concept of the DUPIC linkage is fairly straightforward. The spent LWR fuel assembly is transformed into the HWR fuel bundle by way of the DUPIC fuel fabrication process. This removes the metallic components, including the cladding, from the spent LWR assembly. Almost all the spent LWR fuel materials would flow along with the bulk stream through the DUPIC fuel fabrication processes and scrap recovery, except for a small amount of irrecoverable discards. The waste stream from the DUPIC fuel fabrication processes would therefore consist of the metallic components from spent LWR fuel, and the gases and semi-volatile fission products released from the bulk fuel material treatment, in addition to the measurable discards and losses. There is no liquid waste arising from the DUPIC fuel fabrication processes which depend entirely on dry method, in contrast to wet processes from which liquid waste as effluent arise. Once fabricated, the DUPIC fuel pellets are loaded into metallic components of the (CANDU-type) HWR fuel bundle.

#### 2.3. Analysis of Synergistic Effects

The synergistic effects which can be expected from the DUPIC linkage between LWR and HWR, in comparison with the once-through cycle, can be summarized as follows:

• Removal of Spent LWR Fuel

Spent LWR fuel is transformed into DUPIC fuel for burning again in the HWR; hence there can be a reduction in the repository requirements for direct disposal of LWR spent fuel.

• Saving of Natural Uranium

Since the DUPIC fuel would replace natural uranium (or other enrichment) fuel for the HWR, significant resource saving can be expected in the corresponding LWR-DUPIC cycle. The environmental impact of mining the equivalent amount of uranium would also be avoided.

• Reduction of Spent Fuel Arising from HWR

Since the DUPIC fuel can increase the HWR burnup (as a result of the two-fold increase in fissile content, compared to natural uranium), a further reduction in spent fuel arising from HWR can be expected using the synergism of the DUPIC fuel cycle.

• Environmental Effects

Our ongoing DUPIC programme has recently revealed one interesting benefit pertaining to the geological disposal of spent DUPIC fuel. The DUPIC processing, and subsequent irradiation in a CANDU reactor, results in a decay heat curve with time for the spent DUPIC fuel which is very similar to that of the original spent PWR fuel, before recycling. Hence, significant additional energy can be derived from the spent PWR fuel, without incurring an additional burden in decay heat. Since the decay heat evolution

with time is a major determinant in the spent fuel emplacement density in a geological repository, the disposal costs of spent DUPIC fuel (per electricity generated) are considerably lower than for either spent CANDU natural uranium or spent PWR fuel [5].

It would be interesting to look at some other effects, be they positive or negative, that would require more complex and extensive analyses. Although some preliminary efforts have been made in this direction, future studies will need more detailed technical results from the R&D program which is making further progress each year.

#### 3. COMPLIANCE WITH SAFEGUARDS CRITERIA

One aspect of nuclear fuel cycle which calls for a special consideration, especially in the backend of the fuel cycle, is the proliferation risk of special nuclear materials (SNM). The international implications of this issue were recognized by the INFCE (International Nuclear Fuel Cycle Evaluation) in late seventies, decades after the Atoms for Peace Declaration. INFCE suggested some technical means to enhance deterrence to access to special nuclear materials in the conventional recycle stream, such as: partial decontamination, spiking with fission products, and denaturing. Recently, the US National Academy of Science suggested the concept of the "Spent Fuel Standard" in the context of excess plutonium disposition. The key idea behind the Spent Fuel Standard is to utilize the hostile conditions of spent nuclear fuel, as an inherent barrier to any clandestine access to the nuclear material contained therein [6]. In this sense, the Spent Fuel Standard can be regarded as an augmentation of the various past concepts, discussed at the time of INFCE.

As described previously, the DUPIC fuel cycle concept has technical characteristics that comply naturally with the Spent Fuel Standard, at all steps along the DUPIC linkage between LWR and HWR. In addition to the absence of any separation of SNM involved in the DUPIC fuel fabrication, the inherently hostile conditions of spent fuel material during refabrication require a heavily shielded enclosure all around the fuel fabrication processes, linked with safeguards surveillance systems, which constitute multiple layers of security. The DUPIC processing is self-contained, and there is no transport of intermediate materials outside of the facility: spent LWR fuel enters the facility, and fresh CANDU-DUPIC fuel leaves.

For further enhancement of the safeguardability, efforts are being made in the DUPIC program to develop non-destructive safeguards devices to measure SNM content in spent fuel for accountability [7]. The DUPIC program has already demonstrated some successful results from this work, which will hopefully contribute to other international safeguards applications.

#### 4. DUPIC CHALLENGES

Pursuing the beneficial effects of DUPIC synergism will be an interesting, but challenging enterprise. The unique feature of this exercise will be to develop suitable remote fabrication and handling of highly radioactive fuel. However, this is not a technically blind move, in the light of the current stateof-the-art for nuclear technology. There are, as a matter of fact, a number of technical references that can be consulted on this question from previous studies, not only from the DUPIC program, but also from others as well.

#### 4.1. Reactor Compatibility

Major studies of the technical feasibility of using the DUPIC fuel in CANDU-type HWR have been part of the activities, since the inception of the DUPIC program. These studies have included compatibility in the areas of nuclear, mechanical, control, etc. Although the results up to now have indicated no particular problem, further more-detailed studies will continue [8].

#### 4.2. Remote Fabrication of DUPIC Fuel

This aspect may be the toughest challenge that the DUPIC alternative will have to face. One can perceive, however, rather encouraging prospects in this context from careful examination of state-of-theart in nuclear technology. • Fuel Fabrication

While the technology of the fresh fuel fabrication process itself is quite classic and well-established on an industrial scale for a variety of HWR fuels, there are unique challenges with fabrication of DUPIC fuel. Ceramic fuel fabrication has an element of "black art" as well as science, and much trailand-error, and actual experience will be required to understand the factors affecting DUPIC fuel fabrication. Progress is being made [9], and a major milestone has recently been achieved by AECL and KAERI: the fabrication of 3 DUPIC fuel elements from spent PWR fuel, at AECL's hot-cell facilities. These elements will now undergo irradiation testing in the NRU research reactor [10].

#### • Remote Operation and Maintenance

This question is considered to be the most arduous and sometimes confusing factor in judging the DUPIC feasibility. Since the beginning of nuclear era, many efforts have already been made to develop and apply remote technology to such diverse work as post-irradiation examination, reprocessing and MOX fuel fabrication, radioactive waste handling, etc. The high-tech applications in this area of technology have significantly advanced the technical edge, as can be perceived in the current state-of-the-art. The challenge is how to make use of this continuing technological progress, with ingenuity to complement the DUPIC program. The simple design of the CANDU fuel bundle, and its small size and weight are expected to greatly facilitate remote fabrication.

#### 4.3. Economics

The initial thrust of work on the DUPIC program has been to assess whether it can be done on an experimental scale. Once technical feasibility has been established, the subsequent steps will include optimization of the DUPIC process for larger lab-scale testing, and more detailed assessments of the overall PWR/CANDU fuel cycle costs and economics. At this time, definitive conclusions cannot be made about DUPIC economics, since both technical and financial data for such an assessment are at an early stage. In spite of these difficulties, studies have been conducted at KAERI, using parametric analysis method [4]. These KAERI studies suggest that the DUPIC alternative may be able to compete with the once-through option, if cost-effective technology is developed for the DUPIC fuel fabrication method. A recent reference in support of this view can be found in literature on the Japanese program, Advanced Fuel Recycle System [11]. It can be interpreted from the reference that remote fabrication of radioactive fuel from partial decontamination would not add a significant cost burden, compared to the conventional method of separative processes.

In the future, further efforts will be needed to make more reliable assessments of the DUPIC economics, together with more careful analysis of system engineering.

#### 5. CONCLUSION

The DUPIC fuel cycle is an emerging alternative for the backend fuel cycle, which could provide a useful synergism between PWR and CANDU reactors in Korea (and between LWR and HWR, in general). A key feature of the DUPIC fuel cycle concept is its unique proliferation resistance. Other synergistic effects include natural uranium resource savings, and reduction in the amount of spent fuel that would have to be buried in the earth, thus helping to further minimize nuclear energy's environmental impact. In consideration of the huge amount of spent LWR fuel to be managed in the future, the DUPIC option can be a synergistic alternative for exhaustive burning of the fissile residuals, even without separating them, taking advantage of the high neutron efficiency of heavy water [12]. The effects of the DUPIC synergism in terms of overall fuel cycle economics are to be assessed in due course, as part of future development. The developmental efforts are now in full swing, under the framework of international cooperation, and in anticipation of multiple benefits on the national and international level.

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## A PROPOSAL FOR AN INTERNATIONAL PROGRAM TO DEVELOP DRY RECYCLE OF SPENT NUCLEAR FUEL

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#### Abstract

The dry oxidation-reduction process (called OREOX for Oxidation Reduction of Oxide Fuel) being developed by Korea and Canada, in cooperation with IAEA and the US State Department, is limited to recycle of spent LWR fuel into CANDU reactors (DUPIC). When first conceived and demonstrated via irradiation of test elements by Atomics International in 1965, (the process was called AIROX at that time) a wider range of applications was intended, including recycle of spent LWR fuel into LWR's. Studies sponsored by DOE's Idaho Office in 1992 confirmed the applicability of this technology to regions containing LWR's only, and described the potential advantages of such recycle from an environmental, waste management and economic point of view, as compared to the direct disposal option. Recent analyses conducted by the author indicates that such dry recycle may be one of the few acceptable paths remaining for resolution of the US spent fuel storage dilemma that remains consistent with US non-proliferation policy.

It is proposed that a new US program be established to develop AIROX dry recycle for use in the US, and this become part of an international cooperative program, including the current Canadian - Korean program, and possibly including participation of other countries wishing to pursue alternatives to the once through cycle, and wet reprocessing. With shared funding of major project elements, such international cooperation would accelerate the demonstration and commercial deployment of dry recycle technology, as compared to separate and independent programs in each country.

#### 1. INTRODUCTION

At the Global 93 Fuel Cycle Conference, a number of papers [1 - 3] were presented on the results of studies conducted on the subject of Dry Recycle of Spent Fuel. In addition to those presented by KAERI and AECL on the DUPIC program, which at that time was just beginning, a number of US participants described the results of work done in the US on dry recycle of US spent fuel into US LWR reactors. Both the DUPIC program, and these US based studies have as their origin the pioneering work done by Atomics International in 1965 to demonstrate the technical feasibility of dry thermal processes for decladding LWR spent nuclear fuel, and refabrication of the fuel pellet fragments into new fuel elements.[4] In that work, spent PWR fuel with a burnup of 21,000 MWD/T was declad, powderized via an oxidation reduction process, blended with partially enriched uranium oxide, repelletized and reinserted into the Engineering Test Reactor. The recycled fuel achieved an additional burnup of 10,000 MWD/T. Post irradiation examination revealed acceptable performance of the recycled fuel at PWR operating conditions. A second recycle of the AIROX fuel was being planned when the program was terminated.

Since 1965, the US has considered resumption of this dry recycle development program on at least two occasions, once as part of the NASAP/INFCE studies in the late 1970's, and more recently as a result of Idaho studies of AIROX in the early 1990's. [5] For various reasons the U.S. has chosen not to proceed with further development.

Circumstances have changed dramatically in the last few years. The current US program to store and dispose of LWR spent fuel has incurred major delays, and at the same time increased carbon emissions from fossil plants have focused attention on the importance to the environment of maintaining and growing our nuclear energy base. Furthermore, as a result of work by Korea, Canada, the IAEA and Los Alamos, the use of dry recycle has gained increased acceptability as a proliferation resistant fuel cycle alternative.

Based on these new circumstances, the time has come to reexamine the role of dry recycle in the United States, considering both its non-proliferation credentials, as well as its potential contribution to resolution of the spent fuel disposal problem. The author believes that such a reexamination will lead to renewed interest within the U.S. and a willingness to invest public and private funds into dry recycle technology. This paper describes the basis for this conclusion.

#### 2. UNIQUE ASPECTS OF AN LWR - LWR RECYCLE PROGRAM

Three areas of dry recycle technology which are unique to the LWR-LWR regime, as compared to LWR - CANDU regime are: (1) reactor performance issues, (2) process flow sheet additions, and (3) economics.

#### 2.1 Reactor Performance Issues

The Idaho studies conducted in 1992 [5] concluded that modern US PWR's when operating on full AIROX cores, would require the blending of enriched uranium or plutonium powders into the AIROX powders prior to pelletization. Several cases were examined. For example, to achieve 33,000 MWD/T in the recycled fuel, an increment amounting to  $2\% U^{235}$  enrichment is required. To achieve 55,000 MWD/T an increment of about 3.5%  $U^{235}$  enrichment is required. Economic analyses used the lower burnup case. Fuel management, reactivity and fuel performance analyses used the higher burnup case.

It was concluded that loading a PWR core with 1/3rd AIROX fuel, instead of a full AIROX core will lead to less of a departure from current accident analyses models, and therefore facilitate licensing. In fact a subsequent report of the INEL work [6] concluded that a 1/3rd AIROX core would perform somewhere in-between the performance of a 1/3rd MOX core and a virgin uranium core. Since 1/3rd MOX cores are in use already in Europe, and are to be licensed in the US as part of the weapons disposition program, there is high degree of assurance that AIROX cores, at least at the 1/3rd loading level, will be acceptable from a safety and licensing point of view. This is particularly relevant to nuclear units such as Calvert Cliffs, where license renewal applications are being considered to extend economic lifetime an additional 20 years.

Advanced ALWR's such as the System 80 plus, or the Advanced Boiling Water Reactor (ABWR) are more adaptable to full MOX cores, and therefore full AIROX cores, then are existing LWR's. Therefore, when considering use of recycled fuel in new US reactors, there are likely to be applications where a full AIROX core is practical (ALWR's).

#### 2.2. Process Flow Sheet

Figure 1 is a process flow sheet for AIROX when applied to LWR's. It has three significant differences from the DUPIC process flow sheet.

#### 2.2.1 Addition of Enriched Uranium or Plutonium

Recycle into LWR's requires the blending of significant quantities of medium enriched UO<sub>2</sub> or plutonium. The technology for blending in additional fissile powders is well understood, and no performance problems are foreseen. In the Idaho studies [5], a 500 MT/yr AIROX facility was examined, with feedstock consisting of about 435 MT/yr spent fuel, and 65 MT/yr fresh uranium oxide enriched to 17% U<sup>235</sup>.

#### 2.2.2 Loading of Pellets into Long Fuel Rods and Assembly of Rods

Recycled fuel pellets will be assembled into much longer fuel pins and more complex fuel assemblies, as compared with the DUPIC technology. The assembly of longer pins and full sized LWR assemblies will require additional robotic equipment, of substantially more sophistication than that required for CANDU assemblies. However, because of high americium levels, most of the assembly and inspection of full sized MOX fuel assemblies in Europe is already done by robotic, remotely operated equipment. Therefore, it is not a far stretch to adapt this equipment for canyon type assembly work.

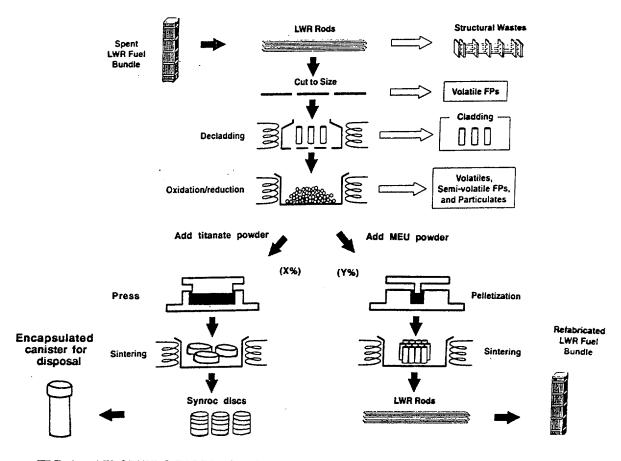


FIG. 1 - AIROX PROCESS FLOW SHEET - LWR TO LWR RECYCLE WITH SYNROC OPTION

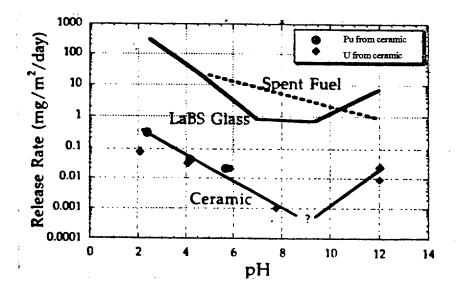


FIGURE 2 - RELEASE RATES; FLOW TESTS FOR CERAMIC (SYNROC), LaBS GLASS and SPENT FUEL (as reported by Lawrence Livermore Lab - March, 1998)

	AIROX Cost \$/KG	LEU PWR Cost \$/KG
Facility Operation	330	
Enriched UO2 Feed	400	
Waste Disposal and D&D	54	
Facility Capital Cost	42	
other	24	
Cost of AIROX or LEU Fuel	850	975
Correction for Cost of Money Correction for HLW Disposal	x	Y
TOTAL FUELING COST	850 + X	975 + Y

FIGURE 3 - COST COMPARISON (1992 IDAHO STUDY) AIROX VERSUS LEU FUEL

#### 2.2.3 Addition of Synroc Type Product for Treatment of Powders Not Destined for Recycle

It is proposed that a separate ceramic production line within the AIROX flow sheet, and process facility be provided for the treatment of AIROX processed powders that are not destined for recycle. This process addition allows for the blending of titanate and other mineral powders into the spent fuel powders, and pressing and sintering of hockey puck sized "Synroc" discs for encapsulation and disposal in a repository. This suggested addition to the AIROX system adds significant flexibility and potential benefits to the dry recycle technology. This could be important in the U.S., where performance improvements may be necessary to assure long term (beyond 10,000 years) safety and licensability of the proposed Yucca Mountain repository.

Some portion of the recycled fuel may have higher levels of actinides (perhaps due to multiple recycles) and therefore may not be appropriate for recycle. Pressing these powders into Synroc pucks creates a waste form for eventual disposal in a repository that is significantly more resistant to dissolution and leaching than either spent fuel or glass. Figure 2, which is taken from recent work performed by the Lawrence Livermore Laboratory (LLNL) for the DOE, compares the dissolution rate for Synroc containing up to 10% plutonium with that of spent fuel and glass. The Synroc is two to three orders of magnitude more resistant to leaching during long term storage than either spent fuel or glass. This greater stability of the waste form could be quite important in assuring the long term performance of a geologic repository, and gaining regulatory approval of repository operations. In the US this could be quite important in licensing of Yucca Mountain.

The US Department of Energy has selected this ceramic waste form for that portion of excess weapons plutonium disposition which is not suitable for MOX fuel. Presently, DOE estimates that about 17 MT of excess weapons plutonium will be converted to this Synroc form, encapsulated, enclosed in vitrified fission products and disposed of at Yucca Mountain. As a result, LLNL is developing industrial scale equipment and technology which could easily be adapted to the AIROX application, and is preparing a repository licensing case for this waste form.

#### 2.3. Economics

The overall cost of recycle, as compared with alternatives, is one of the most uncertain aspects of dry recycle. As part of its nuclear energy research initiative, we have suggested that the US engage in a brief evaluation of the economics of dry recycle technology. We are hopeful that such a study will begin this Fall.

Work done in the 1992 Idaho study partially addresses this question. Figure 3, based on the Idaho study, compares the cost of AIROX LWR fuel with the cost of a new LEU fuel.

The AIROX fuel was projected to cost about \$850/KG, compared to the cost of new LEU fuel of about \$975/KG. However, as the Idaho report noted, the AIROX costs were artificially low, because the cost of money to construct the 500 MT/yr AIROX facility was not considered. On the other side, account was not made of the savings in waste disposal costs that would accrue to the Government, and through the waste fund, to the utility, for avoiding the cost of disposal of the PWR spent fuel after its first cycle. Further study is required to determine the net effect of these omissions.

#### 3. ELIMINATING THE NEED FOR A SECOND REPOSITORY

The failure by the U.S. Government to take title to and store existing and newly generated spent fuel, as required by legislation, tends to reinforce utility reluctance to invest in nuclear power. Obviously, the current low cost of gas and coal fired power plants in the US also inhibits any investment in nuclear plants.

Sooner or later, U.S. fossil fuel prices will once again rise, due to events not presently foreseen. And when they do, it is important that a solution to the back-end of the fuel cycle be in place. Dry recycle of nuclear spent fuel, done in conformance with US non-proliferation policy, can go a long way toward providing that solution.

For example, dry recycle can help eliminate the need for a second repository, even with the continuation of nuclear generated electricity well into the next century. The current law limits the capacity of the first repository, to 70,000 metric tons (MT) of spent fuel. If all of our current 104 plants were to operate for their 40 year licensed lifetime, about 80,000 MT of spent fuel would accumulate. Because of the current economic deregulation of electricity in the US, it is likely that some of these plants will shutdown before their license expires, so current projections are that the 70,000 MT limit will be sufficient for current plants.

But what if the US becomes serious about reducing carbon emissions, and chooses to renew the operating licenses of some plants, (one utility has already announced his intention to do so) and other utilities choose to order new Advanced LWR's of the type recently licensed and certified by the NRC? Under present conditions, the US would have to find a site for a second repository. Considering the extraordinary difficulty in siting, designing and licensing the first repository, proposed for Yucca Mountain, the prospects for successfully siting a second repository appear rather bleak. This in turn would certainly inhibit utility decisions in favor of license renewal or new plant orders.

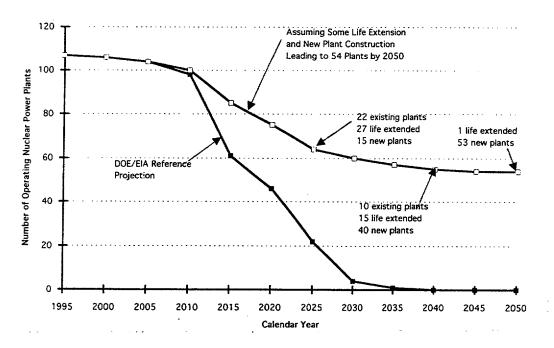


FIGURE 4 - TWO SCENARIOS OF NUCLEAR ENERGY IN THE 21ST CENTURY

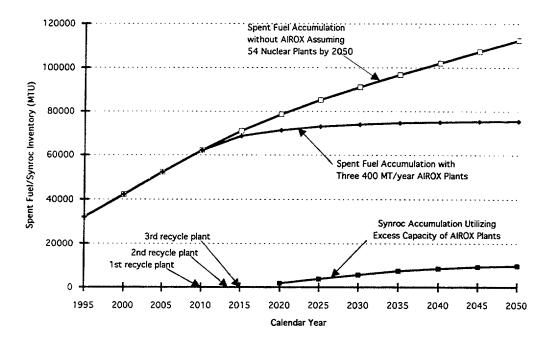


FIGURE 5 - US SPENT FUEL ACCUMULATION WITH MODEST EXTENSION OF NUCLEAR POWER - EFFECTS OF RECYCLE

To help evaluate how dry recycle could influence these utility decisions, we have examined a possible scenario in which about 54 nuclear plants continue to operate through the year 2050. This is an update of previous analyses reported last July to the INMM. [7] Figure 4 compares this scenario of modest nuclear usage in the 21st century with the current mid-range DOE projection [8] of phase out of all plants at their license termination (40 years). For the modest nuclear usage case we have assumed half of the 54 plants are existing plants with life extension (capable of burning 1/3rd AIROX cores), and half are new plants of the ALWR type (capable of burning full AIROX cores.) We have also assumed it will take a dozen years to introduce AIROX on a commercial basis, and an additional 5 years to install sufficient capacity to handle the 54 plants. Incidentally, such a scenario would lead to a reduction in U.S. domestic carbon emissions from the electric supply sector of about 50 million tons per year, about 10% of the total emissions from fossil plants in 1990,

Figure 5, which assumes this modest nuclear portfolio through 2050, illustrates the spent fuel accumulation in the US with and without AIROX. The first scenario assumes no recycle, and quickly leads to the situation where the limits of the existing waste repository are exceeded. Under such circumstances, a utility, even with a good economic case, would be reluctant to order a new plant. In the second scenario, which includes partial recycle, the spent fuel backlog levels off, as old fuel gets recycled into the new, or extended life plants. In this case, the need for a second repository is either eliminated, or at the least, postponed well into the second half of the next century. Whether or not a second repository is required, and when, will depend on whether more new nuclear plants get built to use the recycled fuel.

# 4. AN INTERNATIONAL DRY RECYCLE DEMONSTRATION PROGRAM

The President's Committee of Advisors on Science and Technology (PCAST), and the Clinton Administration have recommended, and the Congress is considering, a new Nuclear Energy Research Initiative in Fiscal Year 1999. The purpose is to enhance the prospects of sustained nuclear energy usage in the 21st century and thereby contribute to reductions in Carbon Emissions as committed by the U.S. at the Kyoto conference. Topics proposed for this new research initiative include "proliferation resistant fuel cycles", extending the burnup of LWR fuel, and improving the nuclear waste disposal regime. AIROX, if developed and used in the US would fall under all three of these topics. It is therefore proposed that the DOE include a modest effort on AIROX beginning in Fiscal Year 99, leading in subsequent years to an international based program to demonstrate the technology at an industrial scale. The program would have three phases.

## 4.1 Phase 1 - Initial U.S. Based R&D Effort (1999 - 2003)

In the first phase, the US under NERI would utilize DOE research facilities, such as those at Idaho, to repeat and augment the original AIROX experiments conducted back in the 1960's. We would try to learn from what has recently been done in Korea and Canada, but would focus our own efforts on the unique aspects of recycle into LWR's including the impact of re-enrichment on the process and on the economics, the fabrication of LWR specification pellets and testing in the Advanced Test Reactor, and the implications of adding the Synroc process to the process flow sheet. This would be a U.S. funded program, conducted in parallel with the ongoing programs in Korea and Canada.

# 4.2 Phase II Joint Demonstration Program, LWR/CANDU Lead Test Assemblies (02 - 07)

The second phase of the program would extend the programs in all three countries to a demonstration phase. Scale up would be a factor of 5 to 10 compared to the lab scale phase 1. Full sized fuel pins would be fabricated for demonstration in commercial reactors, and to test the licensing process. This would be akin to the LTA program in the US MOX Weapons Disposition program. It could also produce AIROX CANDU bundles for irradiation in Canadian reactors, and thereby test the regulatory process on CANDU reactors. This would be a shared program, with each party contributing funds and expertise to the selection of facilities, design and procurement of process equipment, and to the operation of the demonstration facility.

## 4.3 Phase III - Cooperative Prototype Scale Program (2005 - ?)

The third phase would scale this up one step further to a prototype operation, perhaps at the 50 MT/yr level. The US already has several partially built facilities which could be converted to this purpose such as the FPF (Fuel Processing Facility) at Idaho or the FMEF at Hanford. These would be considered, in addition to greenfield facilities in the host country. Here again, participating countries would contribute funds and expertise, and equipment, would share in the operation and learning experience, and would benefit from having domestic plants operate on AIROX or DUPIC fuel.

Successful implementation of this three phase program would create the conditions needed to attract private investment in commercial scale AIROX facilities in the U.S., Korea, or other countries, by the second decade of the 21st century.

This is a vision of what can be achieved in the long run. The benefits to the US and other countries seeking to improve their environment and take advantage of clean nuclear energy would be large indeed. As a first step, I ask the support of the IAEA, AECL and KAERI representatives at this conference, and other interested countries, for this new initiative.

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## FABRICATION OF CANDU DUPIC FUEL

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## Abstract

An important new fuel cycle that exploits the synergism between CANDU® and pressurizedwater reactors (PWRs) is the Direct Use of spent PWR Fuel in CANDU (DUPIC). In this fuel cycle, spent PWR fuel is reconfigured, using only dry processing techniques, to make it compatible with a CANDU reactor. The dry processing technique is inherently simpler than wet chemical processing techniques used for recycling spent fuel. Actinides and fission products are retained in the fuel, so that DUPIC fuel is highly radioactive, affording the fuel cycle a high degree of proliferation resistance. AECL's project to develop the fuel cycle has now progressed to the stage of fabricating DUPIC fuel elements for irradiation testing in a research reactor. The goal of this phase of the project is to demonstrate that the DUPIC fuel cycle is technically feasible. A major part of the technical feasibility study is demonstration of the irradiation performance of DUPIC fuel under CANDU conditions. Spent PWR fuel has been subjected to the oxidation and reduction of oxide fuels (OREOX) process, and the resulting powder has been fabricated into CANDU-quality pellets. The DUPIC pellets have been loaded into fuel elements for irradiation testing in the NRU research reactor at the Chalk River Laboratories. The fabrication stages included spent fuel decladding, powder production using the OREOX process, powder milling (to improve sinterability), pellet pressing, sintering, centreless grinding, element loading and element welding. This paper details the fabrication of the DUPIC pellets and elements and initial results of their characterization. The equipment used for fabrication of the DUPIC fuel elements is described, and the irradiation plan for these elements is also outlined.

## 1. INTRODUCTION

Atomic Energy of Canada Limited (AECL), the Korea Atomic Energy Research Institute (KAERI) and the United States Departments of State and Energy (USDOS, USDOE) are involved in a joint program to develop a process for the Direct Use of spent PWR fuel in CANDU reactors (DUPIC). A phased approach has been followed in developing this process. Phase I was a review of 7 different methods of reconfiguring spent pressurized-water reactor (PWR) fuel to make it compatible with CANDU reactors. The final recommendation from Phase I was to pursue the oxidation and reduction of oxide fuels (OREOX) option [1], a process in which powder is produced from irradiated fuel pellets and is then fabricated into CANDU pellets, elements and bundles.

Phase II is the experimental verification of the feasibility of using the OREOX process to fabricate DUPIC fuel and irradiation testing of this fuel in a research reactor. In this phase, AECL is focusing on powder and pellet technology development, and KAERI is setting up for fabrication of both pellets and, eventually, full bundles.

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## 2. BACKGROUND

Small-scale hot-cell experiments were completed at AECL's Chalk River Laboratories (CRL) using spent PWR fuel. Pellets meeting CANDU specifications for density (expressed as a percent of the theoretical density (TD) of the fuel, currently specified to fall in the range between 95% and 98% TD) were fabricated successfully [2]. Fission-product releases during the process were quantified, and technology for trapping volatile cesium was demonstrated. These experiments were conducted on individual pellets.

In laboratory scale experiments using SIMFUEL (i.e., simulated extended burnup fuel), kilogram quantities of SIMFUEL pellets were processed into pellets meeting CANDU specifications, using the OREOX process. Some decladding techniques were also demonstrated.

## 3. PROGRESS

After 3 small-scale hot-cell experiments were conducted at CRL to demonstrate fabrication of CANDU-quality pellets using spent PWR fuel, a campaign was initiated in the hot cells in AECL's Whiteshell Laboratories Shielded Facilities (WLSF), to fabricate DUPIC fuel elements for irradiation testing in a research reactor. Equipment for in-cell fabrication was purchased or constructed, commissioned out-of-cell, and transferred into the hot cell, as required [3].

In this campaign, pellets were mechanically extracted from the 15-cm-long-cut segments of PWR cladding. There was no need to split open the cladding or to oxidize the pellets to remove them. The fuel did not appear to adhere to the inside of the cladding.

The spent PWR fuel used in this campaign was from the same source as the fuel that had been used in experiments at CRL. The fuel had a discharge burnup of 28 MW·d/kg U and had cooled for 24 years prior to DUPIC processing [4]. Three DUPIC elements were fabricated during the in-cell campaign. Approximately 3 kg of spent PWR fuel was processed using the OREOX process. The process used was as follows:

- declad and extract the spent fuel pellets from the sheath;
- use the OREOX process to produce a fine powder;
- mill the powder using a vibratory mill to increase sinterability;
- prepress, granulate, and press the powder into pellets;
- sinter the pellets to high density;
- grind the pellets to final diameter and surface finish, using a centreless grinder;
- load the pellets into fuel sheaths (one end cap is resistance-welded before assembly);
- weld the second end cap, using tungsten-inert-gas (TIG) welding technique; and
- leak-test the completed element.

Figure 1 shows a view of the fabrication campaign taken through a hot-cell window.

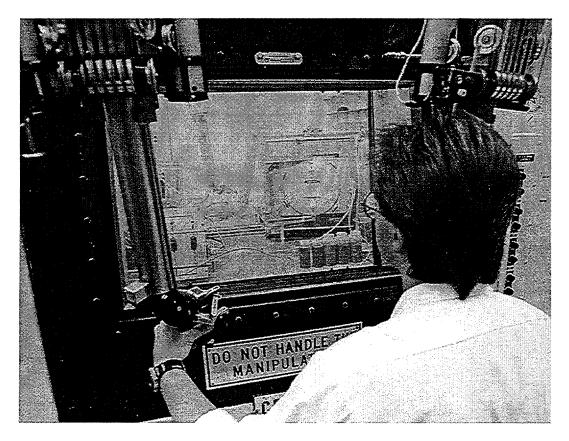


Figure 1. Fabrication of DUPIC fuel in a hot cell. The OREOX furnace is visible in the back of the cell, off to the right. The polyethylene bottles in front of the furnace contain OREOX powder.

The DUPIC elements have the designations BB02, BB03 and BB04. All elements contain a plenum (BB04 has two) and natural-uranium pellets at the ends of the stack. The plena are composed of Zircaloy-4. Plena are used to provide additional void space to accommodate released fission gas. Natural-uranium pellets at the ends of the stacks are used to suppress end flux peaking, and thus reduce hot regions at the ends of the stacks. The elements are designed to be mounted on a NRU demountable element (DME) bundle core. Figure 2 shows a photograph of the fuel stack in element BB02, and Figure 3 shows a schematic drawing of a DUPIC fuel stack. The completed DUPIC elements are shown in the photograph in Figure 4. Details of the compositions (lengths and masses) of the pellet stacks in the 3 elements are given Table 1.

Of the 3 kg of original PWR fuel that was processed in this campaign, 1.3 kg was fabricated into fuel pellets and then loaded into fuel elements; approximately 1 kg was subjected to the OREOX process but was not processed further (because of time constraints and availability of the hot cells), and the balance was either contained in metallurgical samples or lost as waste material through processing techniques.

The processing histories of the various fuel pellets varied considerably. Pellets in BB04 were made from material that was subjected to the OREOX process; after the material was processed into pellets, the pellets were rejected because of excessive cracking, and were resubjected to the OREOX process and refabricated into pellets. This set of pellets had a lower green density, and thus a smaller sintered diameter (11.99 mm rather than 12.19 mm). Overall, this powder processed and sintered very well.

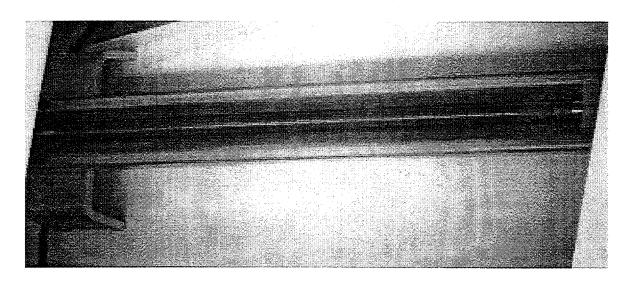


Figure 2. Photograph of fuel stack in element BB02

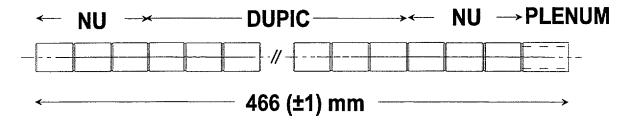


Figure 3. Schematic drawing of DUPIC fuel stack.

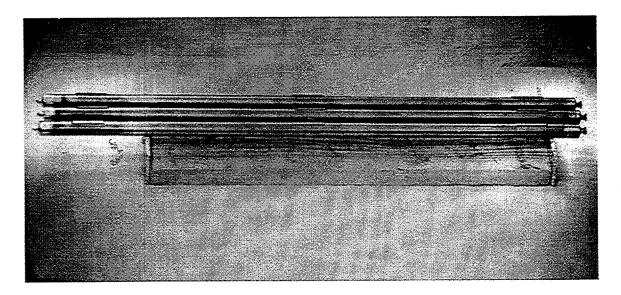


Figure 4. Completed DUPIC elements.

## TABLE 1. DUPIC FUEL ELEMENT STACK COMPOSITIONS

Element Identity	BB02	BB03	BB04
Plenum Length (mm)	24	24	17 (x 2)
Natural U Mass (g)	87.0	82.0	64.0
Natural U Stack Length (mm)	74.5	70.3	45.0
DUPIC Pellet Mass (g)	438.0	442.7	433.5
DUPIC Stack Length (mm)	367.5	371.7	387.0

Pellets in the other elements were subjected to the OREOX process only once. It was found that as-fabricated OREOX powder oxidized rapidly in spite of being passivated (slightly oxidized to provide a protective oxide layer). Excessive oxidation can cause cracking in sintered pellets. As a result, powders that had been subjected to the OREOX process were fabricated into pellets as quickly as possible after production. Even so, there were considerable difficulties with pellet pressing and sintering. The target minimum density was 10.3 g/cm<sup>3</sup> (estimate to be 95.3% of the theoretical density of this fuel, 10.81 g/cm<sup>3</sup>). To achieve this density, some pellets had to be sintered for as long as 20 h. Pellets in BB04 (made from powder that had been subjected to the OREOX process twice) only required 2 h for sintering. All pellets were sintered in a horizontal tube furnace at 1650°C in flowing argon and 4% hydrogen. The furnace load was limited to 250 g (15 pellets) by hot-cell safety constraints (i.e., the potential release of fission products into the cell).

All pellets were dry-centreless-ground to a surface finish of approximately 0.8  $\mu$ m Ra (the CANDU specified surface finish).

The sintered pellets met the CANDU specification for density (expressed as percent of theoretical density) with a minimum density of 10.3 g/cm<sup>3</sup> (95% of the theoretical density of spent PWR fuel at a burnup of 28 MW•d/kg U).

Measurements of chemical content of the fuel before and after the DUPIC fuel fabrication process indicated that volatile cesium was released during the process. It is expected that krypton, iodine and xenon were also released. All other fission products and transuranic elements remained in the fuel.

#### 4. FUTURE PLANS

Irradiation-testing of the DUPIC elements in the NRU reactor will commence during 1998. The elements will be irradiated to a burnup of approximately 20 MW•d/kg HE at power levels typical of those of a CANDU reactor, starting at approximately 60 kW/m and declining over the history of the irradiation to 30 kW/m. The performance of the fuel will be assessed after the irradiation.

Given the experimental nature of this fuel and the desire to obtain information on performance aspects (including especially gas release performance under normal operating conditions), it is considered prudent at this stage to perform a conservative irradiation to ensure survival of the elements throughout the irradiation. The DUPIC elements will be loaded onto a DME bundle and inserted into an intermediate flux position in the NRU reactor test loop for 570 full-power days, achieving a burnup of 20 MW·d/kg HE. A plot of the expected DUPIC element linear power (kW/m) versus irradiation time (full-power days) is shown in Figure 5. The powers achieved in this NRU irradiation test will approximate the calculated envelope of peak element linear powers for DUPIC fuel bundles in a CANDU reactor, fuelled using a 4-bundle shift (also shown in Figure 5).

After irradiation, the DUPIC elements will be removed from the bundle and sent to the CRL hot cells for post-irradiation examination (PIE), including element profilometry, fission-gas volume, element burnup, and pellet and sheath microstructures.

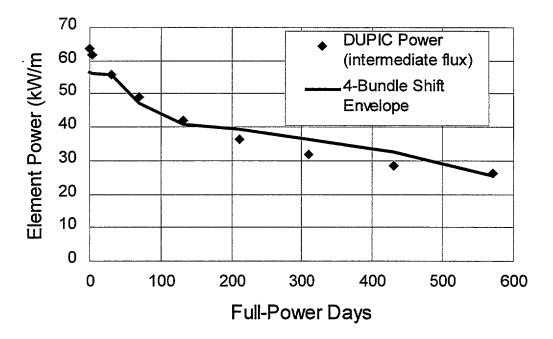


Figure 5. Mid-plane element linear power as a function of time for DUPIC elements on a naturaluranium DME core in an intermediate flux site in the NRU reactor. Also shown is the DUPIC 4bundle shift power envelope at the corresponding burnup.

The goal of this irradiation is to assess the performance characteristics of DUPIC fuel under CANDU conditions. Assessment will be based on a comparison of the PIE data from this irradiation with data from natural uranium and slightly enriched fuel irradiations done under similar conditions.

## 5. CONCLUSIONS

Three DUPIC elements were successfully fabricated in the Whiteshell Laboratories Shielded Facilities hot cells. The fuel pellets meet CANDU requirements for density and surface finish. The pellets were welded into elements that can be mounted on a NRU demountable element bundle core. Irradiation-testing of these elements is planned to commence in 1998 and will be followed by PIE and performance modelling.

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