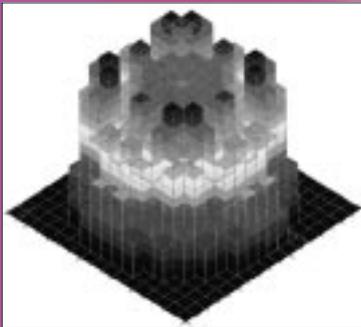


Advanced Reactors with Innovative Fuels



Workshop Proceedings
Villigen, Switzerland
21-23 October 1998



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Proceedings of the Workshop on

Advanced Reactors with Innovative Fuels

hosted by

PAUL SCHERRER INSTITUT



*Villigen, Switzerland
21-23 October 1998*

NUCLEAR ENERGY AGENCY
ORGANISATION FOR ECONOMIC CO-OPERATION AND DEVELOPMENT

ORGANISATION FOR ECONOMIC CO-OPERATION AND DEVELOPMENT

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- *assessing the contribution of nuclear power to the overall energy supply by keeping under review the technical and economic aspects of nuclear power growth and forecasting demand and supply for the different phases of the nuclear fuel cycle;*
- *developing exchanges of scientific and technical information particularly through participation in common services;*
- *setting up international research and development programmes and joint undertakings.*

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FOREWORD

Nuclear power plants operating today utilise about 1% of the energy potential of the uranium they use as fuel, namely the part that is easily and economically extractable with standard technology. What is left is called in present terminology “spent fuel”. There has been an ongoing debate – already lasting several decades – as to what to do with this spent fuel. Past projections based on the resources and forecasts of energy requirements world-wide predicted a rapid depletion of uranium resources. Fast reactor programmes were then developed as a means of extracting a larger proportion of the energy potential from uranium and its derivatives present in the spent fuel. Some countries consequently embarked into spent fuel reprocessing and recycling. Additionally, the oil crisis of 1973 led to strengthening nuclear power programmes.

Since then, several events have changed the picture. First the Three Mile Island accident, then the catastrophe of Chernobyl and the concerns as presented in the media disenchanted public opinion. A period of both hidden and open hostility to nuclear energy emerged in the general public. Since then, a great effort has been invested in research and development for improving safety in nuclear power plants; all of these plants have consequently been modernised. The opening of energy markets world-wide added a strong short-term economical competition component; nuclear power had to adapt, improve technology and reduce costs. The development of advanced nuclear power systems was accordingly slowed down.

Nuclear power represents a substantial share of electricity production (about one quarter in the OECD area), but is only about 5% of the total energy used world-wide. The largest part of this energy comes from fossil fuels. Fossil fuel resources are quite large, but their extensive use feeds the concern that they could contribute to a possible global warming of the atmosphere through greenhouse emissions and to climate change. Satellites have observed intercontinental movements of pollution clouds stemming mostly from fossil fuel combustion. One such pollution cloud was recently discovered hovering over the Indian Ocean, its size being roughly the area of the continental United States. Far reaching environmental impacts could result.

Within the OECD and elsewhere the issues of “sustainable development” are addressed covering environmental, economic and social aspects. Such issues also encompass availability of resources. In fact, shortages of or difficulty in accessing resources have often been at the origin of conflicts. Can nuclear energy play a role and contribute to sustainable development? In the nuclear field the question has been raised as to whether it is ethically acceptable to skim off 1% of the energy potential from uranium and declare the remaining 99% as waste. Do we have the right to deplete energy resources in this way without considering the requirements of future generations? Are there other ways of utilising the huge potential of uranium and other nuclear fuels such as thorium in accordance with the criteria of sustainable development? In order to be able to answer such questions in an objective way it is necessary to identify, based on the most recent technological evolutions, what possibilities do exist, what their characteristics are, and to what extent they meet established safety, economical and environmental criteria. A sound scientific-technical knowledge base first needs to be established, which can be called upon by policy and decision makers.

The Nuclear Science Committee of the OECD/NEA intends to contribute to building such a sound knowledge base. One step in this direction is the Workshop on Advanced Reactors with Innovative Fuel Cycles. This workshop was hosted by the Paul Scherrer Institute, Villigen, Switzerland, from 21-23 October 1998. The proceedings of the workshop, including panel discussions on the role international organisations should play in this context, are presented here.

This text is published under the responsibility of the Secretary-General of the OECD. The views expressed do not necessarily correspond to those of the national authorities concerned.

In memory of...

∞ ***Pedro Landeyro*** ∞



Pedro Landeyro had wished to participate and contribute to the ARWIF'98 workshop. His illness did not permit him to do so. Though he was unable to be present at the workshop, he had hoped to submit a completed article to be published in the proceedings. He was unfortunately unable to finish this last project. He died on 9 December 1998.

Pedro had submitted an extended abstract for ARWIF'98 containing some of his results. Though incomplete, we are including this work in the proceedings. In doing so, we wish to pay tribute to Pedro for the outstanding work he accomplished in the field of criticality safety calculations and more recently in accelerator driven transmutation systems. He will be greatly missed by his friends and colleagues.

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EXECUTIVE SUMMARY

Motivation, scope and goals

Plutonium and minor actinide recycling in thermal and fast reactors is being studied in many countries with the aim of maintaining and developing fuel cycle options which can be adjusted to changing demands and constraints. The challenge is to move towards an economically and socially sustainable nuclear energy system based on advanced reactors – advanced water-cooled reactors, fast reactors and perhaps accelerator based, hybrid reactors – and new types of fuel cycles which help to minimise the waste arising.

In this context, topics of interest are: the multiple recycling of plutonium in thermal reactors, enhanced consumption of surplus plutonium in thermal and fast reactors, reduction of the uranium demand of thermal reactors to stretch the fissile material resources, transmutation of minor actinides to extract fission energy and reduce the quantity and toxicity of the actinide waste, etc.

The scope of the workshop is comprised of the reactor physics, fuel material technology, thermal-hydraulics and core behaviour of advanced reactors with different types of fuels and fuel lattices. Reactor types considered are water-cooled and fast reactors as well as hybrid reactors with fast and thermal neutron spectra. Emphasis is on innovative concepts and issues related to the reactor.

The purpose of the workshop is to enhance information on R&D activities and to identify areas and research tasks where international co-operation can be strengthened. In this context, particular goals are to identify the roles which can be played by existing experimental facilities as well as possible needs for new experimental facilities. The conclusions of the technical sessions are synthesised and have been discussed by a round table on international co-operation.

Workshop organisation

<i>General chairman</i>	<i>Scientific advisory committee</i>
W. Kröger, PSI	Y.I. Chang, ANL, USA
<i>Local organising committee</i>	M. Delpech, CEA, France
P. Wydler	P. D'hondt, SCK-CEN, Belgium
C.A. Degueudre	D. Haas, ITU-CEC
G. Ledergerber	K. Hesketh, BNFL, UK
J.M. Paratte	H. Mouney, EdF, France
A. Stanculescu	M. Nakagawa, JAERI, Japan
<i>Workshop secretariat</i>	M. Ochiai, JAERI, Japan
R. Van Doesburg	S. Pillon, CEA, France
R. Ringele	E. Sartori, OECD/NEA

Participation

The workshop was attended by 110 participants from 18 countries and four international organisations.

The participation by establishment type was as follows:

Research laboratories and university institutions	70%
Industry	20%
Utilities and safety/regulatory organisations	10%

The full list of participants is provided in Annex 1.

Technical programme

The workshop was organised in four plenary sessions during which 22 presentations were made, followed by three parallel sessions during which 23 papers were presented. The final plenary session was devoted to the session summaries and to a panel on international co-operation. The complete technical programme can be found in Annex 2.

Session summaries and panel discussions on international co-operation

Introduction

Papers on the different technical topics were presented partly in the plenary sessions and partly in parallel sessions. In order to provide a general overview of all presentations to participants and with the objective of facilitating the discussion during the panel, the session chairmen presented their summaries and conclusions first.

For the purpose of guiding and focusing the presentations and discussions, Peter Wydler had prepared in consultation with the panel members a questionnaire containing the key issues to be addressed. This questionnaire, together with summarised answers given during the discussions is enclosed as Annex 3.

The structure of this summary consists of:

- The summary of the sessions.
- The summary of the panel discussion.
- The questionnaire with summarised answers and conclusions.

Session summaries

Chairmen: Alain Zaetta (CEA Cadarache)

Rudy Konings (NRG Petten)

Masayuki Nakagawa, Toshitaka Osugi (JAERI, Tokai-mura)

Kevin Hesketh (BNFL Springfields)

Summary of sessions on advanced U/Pu oxide based reactors – Chair: A. Zaetta

A summary of papers presented during the first session of the workshop on advanced U/Pu oxide based reactors, during which presenters outlined the main issues in plutonium management, renewable resources and the closed fuel cycle, is given below.

A variety of light water and fast reactors using different fuels are capable of fully incorporating plutonium into their fuel cycles; options range from “concentration” (plutonium without uranium) to “dilution” which requires the presence of slightly enriched uranium. A number of PWR concepts that use uranium-free plutonium fuel include APA, while the CAPRA programme established a concept necessitating a high plutonium concentration in fast reactors to counterbalance minor actinides absorption. Plutonium utilisation in LWRs can also be enhanced with highly moderated 100% MOX cores with moderation ratios ranging from 4 to 6 for PWRs, and from 6 to 7 for BWRs. The recycling of reprocessed uranium in LWRs, an essential element in closing the fuel cycle, was considered a relevant issue because it contains minor actinides, and needs treatment like other transuranic elements. Fuel fabrication that makes use of recycled uranium must also be competitive with conventional fuel cycles, and utilities must act to facilitate this process by careful consideration of technological solutions. Uncertainties regarding production of ^{232}U (as well as the daughter product ^{208}Tl) should be addressed within an international framework such as the NEA/OECD Working Groups.

MOX fuel breeding for PWRs and BWRs aims at effective resource utilisation, and is based on current PWR and BWR technologies. Innovative PWR concepts, such as tight hexagonal lattices, were presented in a breeding PWR case study in Japan whereby the breeding ratio is 1.1 with a D_2O moderated heterogeneous core design with fertile zones. Two innovative BWR concepts using tight lattice configurations were also discussed. The first concerned a RBWR (Resource-Renewable BWR) which operates with mixed oxide fuel within an epithermal spectrum and displays a breeding ratio of 1.0, while the other case involved a BBWR (Breeding BWR) with a fast neutron spectrum and a breeding ratio >1 . However, the presence of rare earth elements in the fuel cycle may deteriorate core characteristics such as the breeding ratio. A negative void reactivity effect can be achieved with the presence of the cavity-can in the streaming channel structure; furthermore, the reactor has a potential to transmute minor actinides.

KAERI is currently developing a concept to increase uranium resource utilisation in CANDU reactors. DUPIC (Direct Use of Spent PWR Fuel in CANDU reactors) fuel technology uses spent PWR fuel (35 GWd/t) that can be directly fabricated into a reusable fuel for CANDU reactors (additional 15 GWd/t). Fuel characteristics are markedly different from standard CANDU fuel types. International co-operation has already been established through the IAEA and an experimental programme is underway. This research aims at achieving competitive costs.

JAERI is involved in the design of several innovative MOX fuelled PWRs and BWRs, with an ambitious goal to achieve 100 GWd/t burn-up and three-year cycle length with a water-cooled full

MOX core. Proposed designs feature moderation ratios and plutonium contents of 2.6 with 12% for PWRs, and Pu content, and 2.0 with a 16% for BWRs, respectively; they are feasible from the perspective of reactor physics parameters.

A more fundamental approach is being investigated at IRI, in establishing fuel temperature coefficients (FTC) for several fuel types from MOX fuel to uranium-free inert matrix fuel, two of which included reactor-grade and weapons-grade plutonium. All fuels, including those containing weapons-grade plutonium, have negative FTCs.

Experimental research facilities

Zero power facilities for reactor physics analyses are needed for the validation of data and codes. The LR-0 facility, for example, is suited for a simulation of a VVER 1000 and 440 with a hexagonal lattice configuration, and for conducting basic neutron physics experiments. A weapons-grade MOX programme was proposed for VVER 1000.

The PROTEUS facility in Switzerland has developed a three-year LWR experimental programme using real fuel assemblies with some generic research and development aspects. The programme is supported by Swiss utilities and is being conducted in two experimental phases for BWRs and PWRs.

Conclusion

Three conclusions can be drawn regarding the various fuel technologies and reactor design issues discussed above:

- *First*, fuel performance improvements can occur by enhancing MOX fuel burn-up and re-using spent fuel, as exemplified in the DUPIC and URT concepts. Such improvements in the fuel cycle will also act to reduce costs.
- *Second*, there is a wide range of options in terms of consumption and breeding using plutonium in LWRs and FRs. The implementation of such options are strongly linked to the *time scale* considered. Research and development concerning various plutonium utilisation routes are thus justified.
- *Third*, there are several specific issues concerning present and future research and development. Current facilities – which include irradiation facilities, zero power reactors, hot laboratories and nuclear data management centres – operate under strong restrictions. Thus, research and development needs often arise that cannot be adequately addressed. It is therefore important to maintain and develop R&D “tools”, and to define the role of international organisations within this context as that of co-ordinating, fostering and providing guidance. In order to achieve this objective in the short-term, it is necessary to create a comprehensive survey report that acts to clarify the needs, develop strategies and gain the support of all partners.

Summary of sessions on uranium-free reactors – Chair: R. Konings

Numerous fuel cycle systems for uranium-free reactors were discussed in this session, a summary of which is given in the table below (Table 1). For the purpose of this summary, four types of fuels

Table 1. Fuel cycle systems for uranium-free reactors

Materials	Additives	Assembly	Core loading	Reactor	Strategy
Y-SZR	Er ₂ O ₃	Homogeneous	Partial	BWR	Recycling
MgAl ₂ O ₄	Gd ₂ O ₃	Heterogeneous	Full	CANDU	Once-through
SiC	ThO ₂			LWR	
CeO ₂	UO ₂			FNR	
ThO ₂	¹¹ B				
Metals					

Table 2. Uranium-free fuel types and materials

Uranium-free reactor fuel type	Materials
SS-IMF	(Zr, Y, Pu, X) O ₂
CERCER	(Zr, Y, Pu, X) O ₂ in spinel or MgO
CERMET	(Zr, Y, Pu, X) O ₂ in metal
Thoria	(Th, Pu) O ₂

considered in current uranium-free reactor programmes are outlined (Table 2). These fuels are being developed with four principal objectives in mind: capacity for plutonium burning, reduction of radiotoxicity and actinide transmutation, optimal use of natural resources, and improved safety.

Promising reactor physics characteristics have been achieved for ZrO₂-based solid solution *inert matrix fuel (IMF)* assemblies. Although fabrication technology is only available on a laboratory scale, 100% (homogeneous) IMF cores have been shown to reduce plutonium as effectively as for MOX fuels without requiring any further reprocessing treatment (“once-through”). The (low) thermal conductivity of this fuel type is one of the major uncertainties for its application.

Although not really inert, *thoria-based fuels (TD)* show good Pu burning capability for some LWRs and Na-cooled FRs. The technology for thoria-based fuels is partly available from experience from the past. Thoria-based fuel is also of great relevance from the point of view of resources.

Two other types of inert-matrix fuels are being studied. Research concerning the combination of CERamic fuel and a METallic matrix (*CERMET*) for PWRs, based on zirconia and a conducting metal, is mainly theoretical and little experimental work has been carried out. Materials composing the fuel have not yet been adequately defined, and burn-up studies and accident research, which offer good potential for CERMETs, are to be initiated. Much experimental work is being undertaken on CERamic matrix and CERamic fuel (*CERCER*), mainly for “once-through” burning of plutonium in LWRs. The fuel concepts for CERCER fuel are converging in the sense that the MgAl₂O₄ (spinel) is foreseen as matrix and plutonium (or americium) dissolved in stabilised zirconia as fuel. Issues to be addressed in R&D for CERCER include resistance to irradiation, reactor-initiated accidents (RIA) and increases in thermal activity.

Silicon carbide (SiC), which is proposed as an IMF for CANDU reactors, is currently not considered in fuel programmes in Europe and Japan.

Three main conclusions can be drawn from this session's presentations. *First*, no common strategy for uranium-free fuels is emerging. *Second*, materials issues are poorly addressed. *Third*, licensing procedures for new fuels may take from 15 to 20 years to complete.

Summary of sessions on reactors with non-oxide fuels – Chairs: M. Nakagawa and T. Osugi

The advantages and rationale behind the use of new non-oxide fuels and the properties and experimental work undertaken by institutes concerning these fuels was addressed in these sessions. Papers presented at the plenary and parallel session on reactors with non-oxide fuels covered the topics listed in Table 3.

Table 3. Matrix with number of papers per topic discussed

	Metal	Nitride	Molten salt
Reactor	1	3	
ADS			2
Fuel	1	1	1
Cycle		2	1

The advantages of the use of and research and development into non-oxide fuels lies in their:

- Passively safe response to anticipated transients without scram (ATWS).
- High linear pin power and high thermal conductivity.
- Excellent neutron economy.
- Effective use of neutron for plutonium and/or minor actinide burning (high heavy metal density).

The large proliferation resistance of these fuels is illustrated in remote reprocessing and re-fabrication and the fact that non-oxide fuels are associated with MA and FP by the pyrochemical process.

The following findings with regard to current programmes incorporating non-oxide fuels into reactor fuel cycles were presented:

- Tests on metallic fuel in fast reactors have yielded a very high burn-up capability in excess of 20 at%.
- Examination of core performance of the nitride fuel fast reactor (LMFBR) were presented together with the pyrochemical process which has the feasibility of recovering the radiotoxic ^{14}C and the expensive ^{15}N .
- Studies on large plutonium consumption in fast reactors within the context of the CAPRA programme revealed that pure PuN fuel dissolves properly during PUREX reprocessing operations, with a high Pu burning rate and dynamic performance of PuN core.

- The KALIMER liquid metal reactor design contains a uranium metallic fuelled core with a power of 390 MWt.
- In a comparative study of dynamic behaviour of a nitride fuelled LMR core and an oxide core, the ULOF (“unprotected loss of flow”) event displayed a larger Doppler feedback effect when it was necessary and smaller effect for nitride cores in unfavourable conditions.
- In a study of thermal decomposition behaviour of UN and (U_{0.8} Pu_{0.2}) pellets, the initial thermal decomposition temperatures of UN and MN pellets was found to be at least 1 800°C, in various atmospheres. In the higher temperature range, thermal decomposition occurred with vaporisation of metal.
- Molten salt reactor technology in Russia was reviewed and evaluated including aspects of fuel technology, container materials, components and fuel clean-up.
- Experimental work is being carried out in the Czech Republic on liquid fuel concepts, which have several advantages over traditional solid nuclear fuels. New reactor systems using liquid fuels provide for nuclear incineration of spent fuel from conventional reactors and a clean energy source.

Summary of session on international activities – Chair: K. Hesketh

The chairman began the session with a brief summary of activities of the OECD/NEA Nuclear Science Committee of relevance to this conference. There then followed three invited papers by representatives of the IAEA, the EC and EFTTRA (a research collaboration comprising several European research organisations and the utility EDF). Major themes of these activities included thorium fuels, new reactors/new fuel cycles, evolutionary/advanced LWRs, P&T, ADS, materials and nuclear data acquisition and evaluation.

In the areas of advanced fuel cycles and partitioning and transmutation, each of these organisations are very active and there are extremely diverse research programmes in progress. The presentations highlight a number of important points:

- The level of activity clearly demonstrates an extraordinary degree of interest both at the scientific level and at the level of the politicians/fund providers.
- There are many diverse driving factors underlying the various activities. These are determined by individual countries unique perspectives and perceived needs. There is clearly no one perspective that will apply to all countries and the multiplicity of perspectives must be respected.
- International activities are stimulating work in the area of advanced/innovative fuel cycles and reactors, both theoretical and experimental.
- There is a clear need for continuing co-ordination between the various international bodies involved to exploit synergies and avoid duplication.

Role of international organisations

At a time of stagnation in the industry outside Asia, there is a need to maintain continuity of competence until another period of plant construction begins. In the intervening period the industry needs to develop its knowledge of the boundaries of the technologies that are feasible, through research into advanced reactors and innovative fuel cycles. These two needs are complementary in that cultivating the R&D effort maintains competencies. The role of international bodies might therefore be:

- To continue to co-ordinate and stimulate R&D activities in the field.
- To continue to act as a central repository of knowledge and expertise.
- To continue to provide the infrastructure for such activities as nuclear data evaluation, where individual countries' resources are no longer sufficient.
- To continue to provide effective gearing of financial and human resources through collaborative projects.
- To continue to monitor the availability of experimental facilities and to facilitate the construction of new facilities, where needed, through co-operative projects.
- To continue to provide a respected source of authoritative advice both within and outside the industry.

Panel summary

Chair: *Wolfgang Kröger, Peter Wydler*

Moderator: *Hans Fuchs*

Members: *Poong Eil Juhn, Henri Mouney, Masayuki Nakagawa, Massimo Salvatores, Philippe Savelli, Leon Walters*

The panel members were chosen to represent different types of organisation:

- *Utilities:* H. Fuchs (ATEL), H. Mouney (EDF)
- *Research organisations:* M. Nakagawa (JAERI), L. Walters (ANL)
- *International organisations:* P.E. Juhn (IAEA), Ph. Savelli (OECD/NEA)
- *Nuclear Science Committee:* M. Salvatores (CEA)

Peter Wydler opened the panel and explained its organisation. A discussion and the conclusions followed a first round of presentations by the panellists. (The summary gathers for each panellist the topics he discussed as well as answers to queries from the participants). Peter Wydler then gave the floor to Hans Fuchs, whose role was to moderate the panel discussion on international co-operation.

Masayuki Nakagawa (JAERI)

For the next 10-20 years energy resources seem to be assured. Starting around 2020-2030 *energy security problems* are likely to arise due to a shortage of natural resources; then nuclear energy will play an increased role. There is not much time to prepare for the developments and realisations needed. The issue of near term versus long term resources needs to be addressed, as high quality scientists and engineers need to be trained and the necessary scientific budgets for following up on the different ideas need to be allocated.

The position of Japan is as follows: currently the nuclear share in electricity production is 40%, and this trend will continue particularly in view of the *reduction of CO₂ emissions*. This will determine the choice of energy resources, specifically in about 20 years, when the energy demand in Asia will be increasing and consequently energy security problems will rise. Asia is then expected to bypass the current energy consumption of all OECD Member countries combined. Japan's energy programme depends on this target time. It is important for Japan to implement the international agreement on reduction of CO₂ emissions. The government of Japan has a plan to construct 15-20 new nuclear power plants for complying with the agreed reduction of 6%. One difficulty is finding new sites for NPPs. The option being studied is to introduce more innovative reactors that could be built nearer to urban areas.

Japan has no particular policy concerning the burning of plutonium. Originally MOX was meant for use in fast breeder reactors, but their building and exploitation has been delayed, in addition FUGEN will be closed down in 2005. Therefore, *new types of high-conversion or breeding reactors* are being studied. Intermediate storage of fuels would increase the flexibility of using plutonium.

Japan's proposal for co-operation concerns "*R&D on Inert Matrix Fuel (IMF)/Target System for Transmutation of Plutonium and Minor Actinides*". The ongoing joint research concerns:

- *IMF system for effective Pu annihilation*. A workshop on IMF was held this same week involving the following organisations: PSI, JAERI, CEA, ITU, ECN, ENEA, POLIMI, etc. Information was exchanged on IMF materials (fabrication, thermo-physical properties, irradiation tests), core design (annihilation rate, safety analysis) and benchmark tests (burn-up of IMF pin/assembly)
- *IM for MA transmutation*. Joint research is being carried out between ECN, CEA, ITU, etc. This involves irradiation experiments at the HFReactor.

Some of these activities are carried out through a more informal co-operation and information exchange. The role for international organisations should be that of assisting in giving these activities a more stable framework. Proposal for future work is as follows:

- *Co-operation in the framework of the NSC*:
 - Co-operation items: R&D of IM (fabrication, thermo-physical properties, irradiation tests), core performance (burn-up of Pu/MA-IM, evaluation of integral nuclear data of MA by irradiation experiments), environmental safety analysis (direct disposal of spent IM).
 - Co-operation style: Information exchange (e.g. workshop), joint experiment.
 - Available facilities at JAERI: FCA (Doppler reactivity measurement), NSRR (fuel behaviour experiment under RIA conditions – U fissile).

- *Workshop on Very High Conversion Water-Cooled Reactors Research*: JAERI proposes to hold a workshop on this topic based on the following background:
 - Feasibility study on very high conversion water-cooled reactors with the objective of providing one possible option for long-term energy supply with experienced water reactor technology.
 - In the 1980s it was recognised that this is not easy to accomplish, but many studies led to designs that are more easily attained.
 - It is now worth pursuing the feasibility with increased emphasis, because conventional FBRs have encountered some difficulties.
 - Recent investigations give some promising results for further study.
 - The main purpose is to exchange information on such research and to promote it further. Useful results from previous studies carried out in the 1980s should be appropriately included.
 - Research areas to be included are: reactor physics, thermal-hydraulics, reactor safety, materials, etc.
 - Through this activity, further co-operation is intended to be considered in more detail.

Henri Mouney (EDF)

It is not obvious for utilities to give advice on long term R&D devoted to advanced reactors and innovative fuels. Utilities' short- and medium-term concern is that NPPs be *operated in an economical and safe manner*.

- A first priority is a continuous improvement of existing systems, their components and fuels. For that objective no new large tests are required.
- The improvements made and suggested at the workshop are promising and sometimes surprising, but they represent in essence old, good ideas.

Management of plutonium is a major concern and the following options can be envisaged:

- Burn it or waste it in conventional reactors.
- Save natural resources through improved use of ^{238}U and ^{232}Th because fissile materials will be needed in a few decades. The Japanese concept of breeding PWRs seem to be a promising option and should be studied further.

The U-free fuel concepts are a very long-term option for the utilities but of interest for minor actinides transmutation only (French law requests that this option be studied).

The non-oxide fuels are interesting. This option should be investigated for the long-term for licensing purposes.

Research and development in the nuclear field is an insurance for keeping the nuclear option alive, especially now when a decreasing proportion of nuclear power is observed in the world.

In summary, *maintaining competitiveness* of power plants in the deregulated market is the keyword for utilities. In order to stay competitive with existing plants, high burn-up is the key issue. Competing with gas fired plants using co-generation will be difficult. This can be achieved in the new plants such as the EPR by increasing the power to 1 400-1 800 MWe not on the load follow but on the base load basis. Such reactors would be operated for 40-50 years. Because staying competitive is not easy, the time between now and when these new types of plant will be built should be used to get prepared to stay competitive. In order to achieve this:

- The existing R&D work plan should be kept alive.
- Both realistic and dream objectives should be explored/investigated with the appropriate level of research.
- This can be done through international co-operation and sharing.
- The regulatory framework needs to be stable and political willingness to continue is required.

Poong Eil Juhn (IAEA)

There is a need to *maintain the scientific knowledge base* for a long-term perspective of nuclear power. Recently the World Energy Congress has started to recognise the role of nuclear power in supplying a stable energy resource. In addition there is a need *to curb the release of CO₂* and no new major energy resource is forthcoming to replace fossil fuels, besides nuclear. Renewable energies represent less than 1% of the current energy supply and could grow at most to a range between 5-20% over the next 50 years. CO₂ reduction can be achieved through nuclear power. Advanced reactors are important in this respect. In order to fit next generation's needs such reactors have to be inherently safe and understandable to the public. Nitride and metal high-density fuels could be a solution, as they seem to meet such criteria.

International organisations can co-ordinate this development and *bring about a consensus of the directions to be taken*. For the long-term there is a need for saving uranium resources and starting in 20-30 years thorium should be utilised as its ore deposits are larger than those of uranium. In any case it should become part of future strategies. The possible benefits of using *thorium together with uranium* should be identified; preliminary studies show a potential benefit for radiotoxicity reduction compared to exclusive uranium fuel cycles.

In developed countries there are at present no further plans for constructing new large size reactors. Only Asian countries have such plans. However for the implementation of nuclear technology *expert professionals need to be available*. This is possible only if R&D continues. The nuclear field attracts fewer students today, and a sharing among Western and Oriental countries might be one way to preserve this technology. A single developing country cannot afford to construct large size reactors higher than 1 000 MWe due to small grid capacity, but is limited to small or medium size reactors in the 100-500 MWe range. Sharing electricity between neighbouring countries, regional co-operation and issues of common waste repositories should be developed with help from the different international organisations.

Nuclear energy will play a role, according to a discussion that has taken place at the last IAEA General Conference, if it stays economically competitive, if it will be proliferation resistant, if it has relatively simple waste problems and if it is inherently safe. One example mentioned is a Russian design lead-bismuth cooled fast reactor fuelled with uranium/plutonium nitrides and with stainless steel cladding having a power of 150-300 MW. A critical experiment is envisaged and a demonstration plant should be built over the next 20 years. There are also reinvigorated nuclear energy research initiatives (NERI) of the US DOE for the next seven years.

Massimo Salvatores (CEA)

At present we do not see a growing nuclear energy scenario, even though there is a clear role for nuclear energy. The issue of lifetime extension of NPPs gives the context for the near term.

Concerning fuel cycle questions, there are particular difficulties with regard to launching new large-scale projects. Which type of R&D, if any, is required?

Concerning *innovative reactors* there are three different *objectives* depending on the different perspectives and four major challenges for R&D:

- Extending current fuel burn-up, full U utilisation in an evolving technology with emphasis on economy and use of energy resources.
- Providing means for managing plutonium and burning actinides if needed in a safe and economical manner.
- For a visionary long-term: concepts with enhanced burn-up – simplified fuel cycles giving rise to less waste, conformance with deterministic safety requirements (Weinberg and Orlov proposals).

For all three, high burn-up fuels and concepts are the central issues. We have to find the best way for achieving these objectives, following criteria such as *safety, economy and simplified concepts*.

The challenges for R&D are:

- At the *study level*: International co-operation on *harmonising views* (taking into account the diverse perspectives) is required and co-ordination between laboratories where financing is available though in a limited way.
- *Fuel and materials*: There are many new candidates for R&D, the basic properties need to be assessed; results of today's studies on paper need to be confirmed in the laboratory. Databases need to be set up for the purpose. Moreover, it should be established where behaviour under irradiation for validation of new fuels is required. Validation of fuels takes a long time and many different fuels have been discussed and proposed during the workshop. Analytical experiments should complement laboratory experiments; modelling of materials from a fundamental point of view is essential, thus exploiting knowledge from experimental databases.
- *Simplified fuel cycles*: The IFR is still an excellent example, meeting the requested criteria. Assessment of what other experiments are needed for validation should be made.

- *Experimental facilities:* Zero power facilities have a high relevance for neutronics studies and are essential, but at present funding is difficult; new lattice experiments are needed, new materials data are required. Hardly any facilities exist that could satisfy all needs in reactor physics for future long-term programmes. Irradiation facilities will be required covering thermal and fast neutron spectra. By the year 2010 all fast spectrum irradiation facilities might be phased out. International co-ordination and co-operation will have to play a role and help sharing needed facilities with new advanced loops required for testing e.g. new coolants. JAERI has proposed joint experiments for high temperature reactors. A new attitude towards opening the facilities is also emerging in Europe. There are promising co-operative initiatives in the EU projects as exemplified by the EFFTRA meeting held this week.

We should keep the door into the future open through concrete actions on:

- Databases on basic material properties.
- Benchmarks for intercomparing performances and scenarios for innovative concepts.
- Joint experiments, in particular in the field of irradiation and neutronics.

There is a clear *role for the Data Bank* to accomplish several of these actions or to help their promotion.

This should be complemented by a *review of needed facilities*. A general review is being carried out to this purpose by the CSNI within the SESAR group in which one chapter is devoted to reactor physics needs. This requires to be expanded for the specific needs discussed at the workshop. Discussion between participants in the workshop and members of the SESAR group should assure that consistent conclusions are reached and recommendations made. (Pierre D'hondt assures this link as member of both groups.)

Philippe Savelli (OECD/NEA)

Nuclear energy is seen in the OECD within the context of such global issues as *sustainable development* for which favourable conditions need to be created. There are three dimensions to this:

- The economic aspects.
- The human and social aspects.
- The environmental aspect.

Energy is one of the key areas in this and careful analysis is under way. It is a pillar for sustainable development, but could also become an obstacle to it.

The global aspects considered by NEA for nuclear energy are:

- Competitiveness (recent study shows an erosion of nuclear compared to gas fired plants).
- Maintaining a high level of safety: new reactors and fuel design development.
- Safe waste management/minimisation of nuclear waste, deep geological repository, reduction of radiotoxicity.

An additional issue concerns the availability of resources for the long-term future.

Most issues addressed at the workshop are relevant in the more general context of OECD views. The role of international organisations is that of stimulating the work (see summary of K. Hesketh) as today nuclear energy is facing difficulties. The role of the NEA and of other international organisations is to help Member countries keep their options open, to keep governments informed, to anticipate potential issues, e.g. risk of losing knowledge and data, and launch initiatives in line with Member countries' requirements. The discussion taking place at this workshop is timely in the context of the NEA's *redefinition of strategic objectives* in particular as concerns nuclear science. Some of the main areas of possible contributions by the NSC and the Data Bank are maintenance and development of databases and the addressing of more basic issues related to materials and their behaviour.

In conclusion all the new ideas discussed at the workshop should allow to gather information that would serve to show the extent to which nuclear energy is a sustainable source of energy: what approaches need to be continued, *organisation of necessary infrastructure* to achieve competitiveness and maintenance of resources, managing waste and maintaining high level safety records. There is a need to establish the right costs associated with the different energy systems and to "internalise externalities". Currently used methodologies, which do not include the full cost of energy for the society, may lead to distortion of the energy markets.

Leon Walters (ANL)

The situation in the USA is unique compared to other countries with large nuclear programmes; through administration policy there is no possibility of reprocessing spent fuel, so there is no excess of separated plutonium. However, an excess of weapons-grade plutonium for strategic needs has been declared, which will be to a large extent irradiated as MOX in existing reactors and thus transformed to the "spent fuel standard." The large energy potential of the plutonium in spent fuel and the enormous energy in our uranium resources is not taken into account by the current administration in the USA. The fast reactor option is the only sensible way to realise this benefit to humankind.

The idea of burning plutonium and transmuting fission products has re-emerged in the USA, but still there is little incentive for research as politically perceived. However, involvement of the USA on the issues discussed at this workshop should take place in a co-operative way. The USA has a great deal to offer in terms of fuel performance experience with all types of fuel including dispersion fuels.

The issues of particular importance in this workshop that are associated with reducing the separated plutonium inventory have provided many new research opportunities and challenges for your young engineers and scientists. As well, this research will keep your facilities viable. It is important that new young people keep moving into the nuclear business such that we *establish a continuity of expertise across generations*. This new research activity helps meet this objective.

A question was raised concerning the need to develop *proliferation resistant fuel cycles* that address the issues raised by opponents to nuclear energy. The IFR along with other similar technologies such as the Russian pyroprocess offer solutions to the proliferation issue. For some opponents, it is possible that we can convince them we have solutions. For others, the proliferation concern is yet another barrier to erect against nuclear energy development. These opponents could never be swayed otherwise. Thus, to develop yet better proliferation resistant technologies is probably fruitless. Our advocates must continue to send the message that we are penalising humankind by depriving them of the promise of unlimited energy by responding to these issues where perfectly good solutions exist and have existed for some time.

Hans Fuchs (ATEL)

M. Fuchs directed the discussion on international co-operation and shared the concern of the lack of research resources for meeting the new challenges ahead.

He reported on the recent *decisions by the Swiss government* concerning nuclear energy. These include the extension by 10 years of the operating licence for NPP Mühleberg, an authorisation for a 15% power increase for NPP Leibstadt and an intent to discuss both a go-ahead for a low/medium level waste repository and the question of limiting the lifetime of existing NPPs (above 40 years). These discussions do not mean phasing out nuclear power, as a proposal for a new federal law would foresee the possibility of a referendum for new NPPs. The problem of reducing air pollution will remain; it is far from the target emissions envisaged which cannot be achieved without the use of nuclear energy. This is also true for CO₂ emissions.

If we take a vision towards the period 2025-2050 we note that in the years 2020-2025 we will have about 8 billion people and in 2050 about 10 billion. It is not clear whether by then the population will have stabilised. The coming decades will thus bring an *energy challenge*; the envisaged consumption of fossil fuels will be a challenge to the environment. Important local problems will first emerge such as air pollution, which will become more and more regional and global, limiting our choices of energy sources. Concerning resources, available capital is also not so abundant; the time is ripe for strategic thinking for R&D. The situation concerning fuels, reactors and schemes is similar to the 60s, when we had many proposals by researchers, occasionally called “paper moderated and ink cooled reactors”. *Strategic decisions are required concerning scenarios* on fuel types and reactor types, as to which developments are robust for the different scenarios, because we do not know exactly what the future will bring in the next decades. During this period in which we have to adapt for changes we will face turbulent scenarios; knowing turbulence is ahead, let’s fasten our seat belts, turn on our grey cells and try to solve the problems together.

Wolfgang Kröger (PSI)

Switzerland, at the heart of Europe, although not a member of the EU, hosted the seminar because PSI is able to contribute to the *development of innovative ideas* in nuclear energy through its unique facilities, e.g. the hot laboratory where research on plutonium and actinides is carried out, the PROTEUS zero power facility for research in reactor physics and the SINQ spallation neutron source as well as the SLS synchrotron light source in the future. To widen the support for their continued operation, PSI is interested in making the facilities available for international projects.

A recent seminar of the EU addressing issues of nuclear energy in changing times has arrived at interesting conclusions: as already pointed out by Ph. Savelli, sustainable development and the role of nuclear power can be expected to receive increased attention. Competitiveness is one of the goals, but at fair prices where external costs are internalised. In this context, PSI is involved in developing a set of *indicators for assessing current and future energy systems and technologies* which allows all systems to be put into perspective and could provide a basis for judgement and comparison. In addition, we have to understand the *relation between safety and cost* and approach it in a new way (it must not be a contradiction *per se*).

Recognising that the future starts now, we have to take appropriate actions which may be summarised as follows:

- Find new forms of bilateral, trilateral and international co-operation taking advantage of the open research market and of support which can be given by the EU, the NEA and the IAEA.
- Develop strategies for nuclear energy to regain its role and support in our society (answering the many remaining technical questions alone is not sufficient) and get the young generation involved in new ways.

Specific actions

The NSC has reviewed the proposals made in general terms during the workshop at its Bureau meeting on 15 December 1998. The NSC Working Party on Physics of Plutonium Recycling and Innovative Fuel Cycles (WPPR) was charged to investigate at its next meeting (4-5 February 1999) how the specific proposals made can be implemented within its work programme and to provide a summary and conclusions for the NSC meeting of 2-4 June 1999.

It was suggested that *in two years time a new workshop* should be held reviewing the use of irradiation facilities, sharing of work and projects and, more generally, preparing a report on the status and development of research infrastructure.

OPENING SESSION

Chair: W. Kröger

THE VIEW POINT OF SWISS NUCLEAR UTILITIES

Hans Fuchs

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President of UAK (Association of Swiss NPP Owners)

Abstract

Nuclear power comprises 40% of total electricity production in Switzerland, and Switzerland currently has five reactors operating at four sites. Nuclear energy has the capacity to provide an economically viable and clean alternative energy source for the future. Despite a ten-year moratorium on the construction of new power plants implemented in 1990, the Swiss government remains basically in favour of nuclear power, although no consensus has been reached regarding the future of nuclear power in current energy policy. Public support for nuclear technology has eroded significantly to the point that expansion of nuclear technologies, including the construction of two nuclear waste storage facilities, has been delayed. Within this context, the role of Swiss utilities in providing financial and technological stimulus to research and development at a time of decreasing government and public support and increasing financial restrictions is paramount.

Swiss nuclear power plants

Nuclear power comprises 40% of total electricity production in Switzerland, while hydroelectric power and oil and gas represent 58% and 2% of the country's total electricity generation, respectively. The 3 079 MWe net electric capacity of Swiss nuclear reactors (see Table 1) makes up slightly less than 1% of total world nuclear electric capacity, while electricity from nuclear power amounts to 24 TWh/year or slightly more than 1% of total world nuclear energy production. At present the Gösgen nuclear power plant is number nine on the world list for total nuclear generation.

Table 1. Electricity production and installed capacity of Swiss nuclear reactors [1]

Name	Type	Capacity MWe net	Start-up	TWh 1997
Beznau I	PWR	365	1969	2.7
Beznau II	PWR	357	1972	3.1
Mühleberg	BWR	357	1972	2.6
Gösgen	PWR	970	1979	7.9
Leibstadt	BWR	1 030	1984	7.8
		3 079		24.0

Several Swiss reactor development programmes were launched in the 1940s and 1950s, commencing in 1946 with Switzerland's first reactor research and development programme initiated by Brown Boveri & Cie. (BBC). By 1955 more than 150 private companies had come together to form "Reactor Ltd.", which was to build and operate a new privately-owned research centre in Würenlingen with two reactors on-site: SAPHIR and DIORIT. In 1960, the Swiss government took over the company, then known as EIR (Eidgenössisches Institut für Reaktorforschung). EIR and SIN (Schweizerisches Institut für Nuklearphysik) then merged in 1998 to form the Paul Scherrer Institute (PSI) [2].

The fuel cycle

There are four principal power plant owners/operators in Switzerland (Table 2), among which there is a growing collaboration on technical and political aspects of nuclear technology and development, including use of nuclear fuel reprocessing strategies and intermediate storage of nuclear waste products.

Table 2. Owners/operators of nuclear power plants in Switzerland

Name of Owner/Operator	Name of Reactor
Bernische Kraftwerke AG (BKW)	Mühleberg
Nordostschweizerische Kraftwerke AG (NOK)	Beznau I and II
Kernkraftwerk Gösgen-Daeniken AG (KKG)	Gösgen (lead: Atel)
Kernkraftwerk Leibstadt AG (KKL)	Leibstadt (lead: EGL)

Reprocessing was seen as the only practicable means by which to manage spent fuel, and even with the foreseeable availability of intermediate storage facilities, spent fuel reprocessing remains an established recycling process deemed essential for the longevity of nuclear power. Approximately 1 100 tonnes of spent fuel – or about 30% of the fuel arisings, assuming 50 years lifetime – has been

or will be reprocessed in France and the UK. Plutonium recycling, in the form of MOX fuel utilisation, has been taking place at Beznau since 1978 and at Gösgen since 1997. The Gösgen reactor also uses plutonium from the Mühleberg reactor. In the last few years MOX fuel has been perceived on the one hand as less economically attractive as compared with UO_2 fuels due to low uranium prices and low energy/enrichment prices, and on the other hand more environmentally sustainable as reprocessing allows recycling, produces a smaller quantity of waste than direct disposal, and is clearly repository-proof. Reprocessed uranium (Rep U) is also being employed (partially) at Beznau and from 1999 onwards at Gösgen.

Switzerland possesses two disposal facilities for low and medium level radioactive waste: ZWIBEZ, located on the site of BEZNAU, and BZL, which is found on the site of PSI. Two further facilities are expected to become operational in the near future: the ZWIBEZ facility for highly active waste and the ZWILIAG facility. ZWILAG, a company founded by Swiss nuclear utilities, is in the process of erecting an intermediate storage and waste treatment facility for vitrified highly active and other waste and fuel elements in Würenlingen. The facility is expected to be operative at the end of 1999. Concerning a repository for low and medium active waste, Nagra (a Swiss national co-operative for the disposal of radioactive wastes comprising plant operators/owners and Swiss Confederation members) finished its 20-year-long site feasibility studies in 1993 when the Wellenberg was selected as the “preferred” site for final storage of low and medium level radioactive wastes. While the residents of the local community of Wolfenschiessen accepted the project, it was halted in 1995 by a 52% to 48% vote of the canton of Nidwalden [3]. Following further safety studies, the Swiss safety authorities and working groups comprising members from the Swiss energy department and the canton of Nidwalden confirmed the suitability of the Wellenberg site. GNW (a co-operative of NPP owners and the host community) is starting a “second approach” which, if fostered by the Swiss government, would foresee first a vote on an exploration shaft and later on the repository itself [4].

As regards disposal of highly active waste, Nagra will complete the basic proof of the safety/feasibility of repositories by 2001.

Utility support for long term research and development

The implementation of a 10-year moratorium in September 1990 has significantly affected short and long term plans for and investment in new reactor development in Switzerland. However, the advancement of reactor technology should be a valid priority for several reasons. First, such advances would allow current nuclear suppliers to solve problems and to maintain safety levels of existing reactors. Second, research and development programmes act to increase awareness of current and future issues and problems in the nuclear field, including raising public consciousness and providing vocational incentives for the next generation. Third, developments in reactor technology would act to support industrialising nations that are relying increasingly on nuclear power as an electricity source.

Swiss nuclear utilities have maintained a high level of support for reactor development [5]. Medium to long term research programmes have been set up at PSI in the fields of waste management, LWR safety, reactor physics and systems engineering, material technology and nuclear processes and thermo-hydraulics. PSI has also undertaken specific work for advanced reactors at the PANDA facility concerning the validation of passive systems for SBWR, ESBWR and SWR-1000. PSI also joined the European Utility Requirements Group (EUR) to foster standardised advanced designs. Development of the European Passive Plant (EPP), an upgraded version of AP-600, has received similar support from Swiss utilities.

Currently, the Swiss Federal Institute of Technology is offering (both in Zürich and Lausanne) education and training programmes in the field of nuclear engineering/physics. A diminishing number of technical colleges (HTLs) offer some basics of reactor technology. An operator training programme is also in place at PSI (Reaktorschule PSI), financed by the Swiss NPP owners. The Federal Office of Energy's Safety Board (HSK), a supervising authority of the federal government which assesses the safety of Swiss power plants, also carries out regulatory safety research in conjunction with PSI, universities and engineering companies [6].

The public, Swiss energy policy and nuclear power

Democracy in Switzerland, as governed by a Confederation of States under the 1874 Constitution, allows the active participation of the public in the political system by means of elections, petition, initiatives (requiring 100 000 signatures in less than 18 months) and referenda (requiring 50 000 signatures in three months). This system has proven highly efficient, although difficulties have arisen involving issues such as membership to the European Community and the EEA (European Economic Area), and concerning nuclear power, which is vulnerable to media campaigns and big headlines. While from a policy standpoint the federal government has supported the need for Switzerland to use nuclear technology to fulfil its energy requirements, local opposition and resulting media criticism has delayed its expansion. Following opposition to a nuclear plant project at Kaiseraugst from the late 1960s to the aftermath of Chernobyl, several anti-nuclear initiatives were instigated in Switzerland in 1979, 1984 and 1990. Voters narrowly rejected all initiatives, including a phase-out of nuclear energy (September 1990). In September 1990 a 10-year moratorium for the erection of new nuclear power plants was accepted by a majority of 54.6% of Swiss voters [7].

Despite weak support for continued nuclear energy use in Switzerland, it must be said that Switzerland relies heavily on nuclear power to achieve its clean air targets. In fact, limits for nitrogen oxides and ozone are violated in many regions of Switzerland, and greenhouse gas emissions would increase by at least 25% without nuclear power. The target for 2010 as defined in the recent "Federal Law on the Reduction of CO₂ Emissions" (1996) is a 10% decrease in greenhouse emissions compared to the base year 1990 [8]. The "Energy 2000" action programme, Switzerland's primary energy policy initiated in 1990 following the Montreal Protocol (1987), aimed at increasing efforts over a ten-year period to stabilise consumption of fossil fuels and electricity, to increase the contribution of renewable energy sources, and to at least stabilise CO₂ emissions by the year 2000 [9]. The role of nuclear power within this framework was outlined as expanding the capacity of existing nuclear plants by 10% by the year 2000. Commencing with the 10% increase of power in Mühleberg and smaller power upgrades in Gösgen and Beznau, followed by the recent permit for a 15% increase in Leibstadt, the total capacity increase will be less than 10%. However, thanks to higher availability, total nuclear generation is expected to be 10% higher in 2000 compared to 1990.

As in most other European countries, the opening of the electricity markets is detrimental to the prospects of building new nuclear power plants in the short and medium term, given the high initial capital requirements and the long time for its amortisation. Understandably, this situation is also not very helpful for long term research and development in such fields as innovative fuels.

Government support for research and development

In 1995, the Swiss government energy R&D budget allocated 28% of funding or \$US 51.02 million to nuclear fission and fusion. This figure has been decreasing gradually by between \$US 2.00

million and \$US 6.00 million per year since 1985, when funding allocation was \$US 75.46 million. More specifically, this decline can be seen in LWR R&D, which has fallen from \$US 22.57 million in 1988 to \$US 5.92 million in 1995, and R&D concerning other converter reactors and the nuclear fuel cycle. At present there is a general decline in world-wide government investment and public support for research and development in nuclear technologies [10]. Given the current environmental and economic climate – targets for reductions of greenhouse gases, free market squeezes on utilities, moves to abandon nuclear power and tight government budgets – it is little wonder that Switzerland is experiencing difficulties obtaining support for research and development in nuclear power. It is clear that global and national strategies, “shared goals” and solutions need to be found which can incorporate and promote nuclear technology.

From an environmental perspective, nuclear and hydroelectric power represent two “environmentally friendly” or emission-free energy sources [11]. Given the incipient effects of climate change caused by increased burning of fossil fuels, coupled with an increasing world population and rising pollution levels, nuclear energy may provide a more economically and ecologically sustainable alternative energy source. However at present, nuclear energy represents only 17% of electricity generation world-wide with only slightly increasing output. Although an increase in uranium prices may be likely during the next 10 years or so and considering that more than 40% of uranium consumption comes from secondary sources (recycling, civil and military stocks), a sustainable electricity supply will have to rely on a much more complete use of uranium resources than with present reactors.

The role Swiss utilities can play in this increasingly difficult socio-political context is of course rather limited. Of paramount importance is the striving for excellence in the operation of existing nuclear power plants, combined with efforts to enable and promote an enhanced role of nuclear power for the benefit of future generations.

In view of the imminent threats of climatic change, mostly to the poorest (over 30 000 people were killed in 1998 by partly man-induced storms and inundations), atavistic moves by left/green parties to abandon CO₂-free nuclear power appear clearly as highly irresponsible. No need to say what this means for the nuclear R&D community: it is futile to work silently on long term developments of reactors and innovative fuels when nuclear power becomes an endangered species in some countries. The time to act is now: defend the prospects for our children, raise your expert voice – loud and clear!

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A UTILITY VIEW POINT

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Abstract

In less than 30 years, nuclear power in France and Europe has demonstrated its capacity to fulfil electricity needs at economical costs. With more than 75% of electricity generated in France in 1997, nuclear power is seen by a majority of French people as a contribution to energy independence and as producing electricity at a competitive cost.

In its Indicative Nuclear Programme, issued in 1997, the European Commission points out the important share of nuclear energy in the EU (30% of total electricity generation) and its favourable consequences for high technology know-how, reduction of the greenhouse effect, and improved security of energy supply.

In France, we have used feedback to standardise installation and gradually improve operating techniques. As a result French nuclear plants meet high standards of performance and reliability.

Introduction

In less than 30 years, nuclear power in France and Europe has demonstrated its capacity to fulfil electricity needs at economical costs. With more than 75% of electricity generated in France in 1997, nuclear power is seen by a majority of French people as a contribution to energy independence and as producing electricity at a competitive cost.

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In France, we have used feedback to standardise installation and gradually improve operating techniques. As a result French nuclear plants meet high standards of performance and reliability.

Nevertheless, we can now observe that the institutional debate focused in the past on the means of generation is moving towards fuel cycle matters where the weakest links in the nuclear power chain appear:

- Firstly, public opinion sees the lack of a proven long-term solution to the question of waste disposal as a failure on the part of the nuclear industry to address the problem in a serious and effective manner.
- Secondly, in spite of the technical and industrial progress made throughout the chain, the fuel cycle is still the area with the greatest potential for cost reduction. But the pressure created by the deregulation of electricity markets and the low cost of fossil fuels makes it important to address this issue right now, if we are to make progress.

Nuclear power is able to adapt its capacity thanks to very effective R&D. But it also presents some constraints due to very high investment costs and very long time scales: 10 years to build a plant with a 30-50 year lifetime, 15 years at least to run the complete fuel cycle.

The nuclear operator's constraints and objectives

For the nuclear operator, four overriding requirements dominate fuel cycle considerations:

- 1) To gain the maximum from investments. This means a good level of performance for all uranium and MOX fuels, with a burn-up as high as possible.
- 2) To reduce the cost of fuel, which currently represents 25% of the cost of each kWh. This is the major factor as said previously that can still be linked to total cost reduction once the initial investment has been made.
- 3) To minimise the risks of environmental impact of byproducts (plutonium, minor actinides, fission products).
- 4) To prepare for the economical and political uncertainties of the future. This means adopting strategies and solutions that are sufficiently flexible to adapt to future political decisions and economic trends.

Finally, though, we should not forget that when a utility operates a nuclear power plant, the first objective is to generate a low cost electrical kWh in a reliable and safe plant in order to remain competitive. The fuel cycle as a whole must help to reach this objective and particularly when we are dealing with advanced reactors and innovative fuels, without forgetting this management of the back end of the fuel cycle.

A French law of 30 December 1991 is of crucial importance in this respect, particularly the first issue of the 15-year research programme related to separation and transmutation of long-lived nuclear wastes. It is clear that this law has been successful in making the French nuclear sector more receptive to ideas and new solutions. This was largely due to the extended time frame allocated to the research programme. This time frame also made it possible to separate political decisions from scientific and technical advances and the selection of industrial strategies.

Possible future strategies

In France, work has been carried out at a national level by a task force bringing together the various operators from the nuclear industry (utility, manufacturers, reprocessors, etc.) and the associated research bodies. The group reviewed all the issues related to the fuel cycle in France and assessed 10 possible future strategies of increasing complexity from direct disposal of spent fuel to P and T of minor actinides and long-lived fission products, passing by plutonium recycling in various types of reactor.

This review was aimed to verify the pertinence of the current strategy in the light of preliminary research results and trends in industry. The first conclusions, partly specific to the French situation, establish that the strategy followed up to now is not leading down a one-way street, but will leave the door open to a wide range of options.

In fact, the present EDF strategy, that is recycling of separated plutonium in the 900 MW PWR, with adaptation of reprocessed quantities of UO₂ fuel to the recycling possibilities, is flexible enough to manage the different scenarios examined. An important point is also that no major industrial decisions need to be taken before the deadline of the above mentioned law in 2006.

Thus, what we can foresee over the next 30 years is that the context of nuclear energy will not be deeply modified in areas such as specifications concerning LWR nuclear plants and treatment of used fuels (reprocessing, storage and repository, etc.).

Moreover, these specifications will be valid world-wide and they will condition the R&D programmes. For a utility, nuclear energy must be reliable, less expensive and offer a longer service life. This involves placing the emphasis on materials and components improvement rather than on global innovation during this period.

The situation would change in the second half of twenty-first century, only if a shortage of uranium began to emerge, as well as a shortage of fuel and gas.

At this time, the world would need sustainable sources of energy, not only a transitional solution associated with ²³⁵U. As far as we can see today, this source can only be found with ²³⁸U and/or thorium. Any reflection about innovation and R&D problems in the field of nuclear energy must be placed within that framework. The problems to be solved are quite serious: What fuel systems? U or Th?; What kind of reactors? Liquid metals, gases, molten salts, hybrid systems, etc.

We may think that we are entering a long-lasting period of innovation of re-building concepts, and also of choices between major alternatives. However, if we compare the years ahead with the 1950-1970 period, when the concepts underlying our present nuclear industry were created, we must realise that the period we are entering is far less favourable to technical innovation.

From these considerations, the EDF draws two conclusions:

- 1) We have time to implement new technologies, in fact:
 - The future of present or advanced light water reactors is well defined at least until 2050, with N4 and EPR reactors in France.
 - The most “hard” strategies for the back-end of fuel cycle such as P&T or geological disposal, need not to be implemented before 2050.

The facts give time to decide what strategy to choose, in order to manage plutonium and other long lived radioactive products.

- 2) As said previously, at this time we will probably apply specifications and rules for reactor safety and fuel cycle management which will become European or international.

Requirements are far more stringent today, as well as more widely accepted and understood. But these requirements finally prohibit sudden changes and make increasingly difficult to transfer research to industrial development. The difficulty and the cost of safety approach, the hazards associated with public opinion have developed among industrial investors a resistance to risk and innovation which will impose severe constraints on the evolution.

What place for innovative concepts?

The technologies needed for implementation of the examined strategies are not immediately available and present various degrees of maturity; some of them still require decades of development. The role of R&D is essential in order to make available new technological solutions if needed. For the nuclear industry short and mid-terms are the priority, focused on the understanding of ageing and life extension of old reactors, the design and the operation of the next standardised plants. Long term research on innovative technologies must be supported by nuclear industry but is primarily dependant of public and political authorities.

We can then consider this R&D as a factor of flexibility and adaptation of coming strategies in terms of feasibility, performance, reliability, safety and public acceptability.

For the French utility, the plutonium issue should be solved first. For the innovative technologies in the long term, we should develop new fuels able not only to withstand severe accidents but also to reduce waste. This is why we support consistent fuel R&D programmes in the frame of a tripartite agreement between the EDF, the CEA and FRAMATOME; the programme for Pu recycling is called CAPRA, and that for minor actinides burning is called SPIN.

Fuels for plutonium recycling

LWR

Pu recycling in current PWRs is an industrial reality in France, Germany, Switzerland and Belgium. Today 20 units are licensed in France for 30% of MOX fuel loading and 16 loaded. Requests for eight additional reactors are being submitted by the EDF to the safety authorities, and they are scheduled to be licensed by the turn of the century. During the last 30 years around 40 tonnes of Pu have been recycled with success in the French reactors, with performance always progressing and significant feedback on fuel fabrication and behaviour emerging.

Examination of irradiated rods has shown that MOX fuel has a behaviour similar to that of UO₂ (water corrosion, deformation), but is less favourable for gas releases. At the opposite end of the spectrum, MOX fuel is less sensitive than UO₂ to PCI (pellet-cladding interaction). Finally, experimental analytical and global irradiations on a monitoring programme are so far performed to qualify MOX fuel for higher burn-up.

New Pu fuels

The progressive degradation of the Pu isotopic vector due to higher discharge burn-up of UO₂ fuel will require in the future to increase the Pu content of the next MOX fuel generation, with consequences to be anticipated on fabrication plants (MELOX is licensed up to 12% Pu content) and on reactor core safety parameters (void effect, ...). The EDF is presently running the "PARITY MOX" project in order to qualify MOX fuel for higher burn-up and Pu content.

Innovative fuels

CAPRA fuel concept

The MOX fuel CAPRA is designed for an increased Pu consumption and the possible use of variable Pu isotopic composition. It must be qualified because the 35-45% Pu content range is out of the scope of the current fast reactor fuel experience.

For a higher level of Pu burning, a further increase of the Pu content in the range 60-80% is needed: reprocessing constraints will then lead to the use of nitride fuel, with the problem of ¹⁴C production to avoid.

At the last stage, for a maximum level of Pu or MA burning, U-free fuels could be considered. Such fuels are currently studied or investigated by the CEA for Pu burning in PWRs (APA fuel concept) for Am transmutation in dedicated targets. In both cases, the choice of compounds to be used remains open (oxide, nitride, metallic) and would be supported by strategy and cycle technology considerations.

Long term considerations

Throughout the next few decades, LWRs are almost certain to predominate, with evolutionary changes to permit more extensive utilisation of MOX fuel and probably some synergy with other types. In the long term, other systems may be gradually adopted such as:

- Fast neutron reactors that realise the full energy potential of uranium and can sustain a balance between production and consumption of plutonium.
- Accelerator driven subcritical systems, if their advantages are assessed to outweigh their complexity.
- Possibly molten-salts systems (critical or subcritical) as a mean of amplifying the whole cycle.

As regards fuel, oxide is now standard. Using nitride with better performances would favour higher Pu incorporation, but ^{14}C formation has to be avoided by utilisation of nitrogen enriched in isotope 15. The possible formation of ammonia on contact with water, through a failed clad or on dissolution would present some difficulties. Manufacturing technology has never been industrialised.

Carbide – with rather similar advantages and more drawbacks – is apparently less favoured.

Metallic alloys with zirconium have been proposed for IFR to be pyroelectrochemically reprocessed in a tightly closed cycle.

In molten systems continuous reprocessing would be limited to the removal of fission products and would be integrated with the reactor. Suitable chemical means is certainly one of the main development requirements. But the potential advantages of this concept are its applicability to both thermal or fast neutron spectrums, uranium or thorium based (or even inert support), both breeder or burner, with promising characteristics in terms of safety and efficiency. Nevertheless, since the fuel and coolant are the same fluid, dispensing with cladding, a barrier is missing in the principle of containment in depth for fission products that are particularly volatile.

Finally, thorium can be envisaged and investigated as a substitute for uranium as fertile material in a nuclear system in order to reduce the formation of transuranics. Problems would not come from manufacturing fuel for any reactor type so far considered, but would arise in reprocessing for a sustained thorium ^{233}U cycle.

Concluding remarks

Development of nuclear systems and fuels will certainly be evolutionary for some decades, with more strong innovations expected later. The main demand at present is to reduce cycle costs with an optimised plutonium management. If the incineration of minor actinides and perhaps long-lived fission products has to be implemented, then specific targets may be needed to overcome various difficulties.

Much of the technology required will require a long gestation period for full development and implementation to occur.

The benefits to be gained from a structured research programme conducted at the national level over a reasonably long period are promising. However this framework can achieve little without a robust and flexible strategy. The EDF has therefore adopted a position on the middle ground between two extreme solutions: extensive reprocessing combined with multirecycling on the one hand and direct disposal of non-reprocessed elements on the other.

We must also remember that while the choice of options and strategies may remain open in the long term, this does not mean that they are equivalent in terms of their value or cost. The electricity market is opening up throughout the world, and all the players in the market are going to face keen competition. This being so, the EDF must ensure that the solutions selected minimise the impact on the cost of each kWh generated. We may consider that, as the energy requirements of the world grow, the price of energy will grow too, so that the amount of resources devoted to develop innovative solutions will increase accordingly. Nevertheless, the road from R&D to industrial realisation will remain long, full of difficulties and even shipwrecks. In this respect, the nuclear industry's problems are international and it is essential to reach a satisfactory future position. That purpose will require extensive and fruitful co-operation.

ADVANCED U/Pu OXIDE-BASED REACTORS

Chair: A. Zaetta

PLUTONIUM UTILISATION IN PWR AND FR

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Abstract

A large number of studies have been devoted in France and in Europe to plutonium utilisation in thermal and fast reactors. This paper aims at giving a summary of the different strategies of plutonium management in PWR and FR, firstly by defining the physical factors which limit plutonium content and plutonium consumption in conventional reactors, and secondly by introducing the design changes necessary to make multiple recycling feasible. Finally, a comparative examination of the different Pu burner concepts will be proposed, not only at the reactor scale, on the basis of core performance or plutonium consumption efficiency, but also at a nuclear park scale on the basis of technological impact on MOX and UOX fuel cycle plants and on the waste radiotoxicity level in a final repository.

Introduction

The only route for producing plutonium in a reactor is neutron capture by ^{238}U so that the main guideline for consuming plutonium is to reduce the uranium content in the core. This could be achieved in an FBR by withdrawing the fertile blankets, but also can be accomplished – regardless of the reactor – by increasing the Pu enrichment in the fuel. The higher the plutonium content of the fuel, the higher the Pu consumption. However, the high initial reactivity, due to the high Pu content, must be offset by reducing the fuel inventory in the core or increasing the neutron leakage. All these core modifications have serious consequences on safety parameters (like coolant void effect, Doppler coefficient, fraction of delayed neutrons, ...) and on reactor performance (like reactivity loss and cycle duration). This paper aims at giving different solutions to be used to overcome these difficulties, and analyses the consequences of plutonium management on the whole fuel cycle (raw materials savings, fraction of nuclear electric power involved in the Pu management, consequences on the radiotoxicity level in final repository, ...). It is focused on two types of scenario: one involving a low fraction of the nuclear park dedicated to plutonium management, the other involving a dilution of the plutonium in all the nuclear park (scenario with 100% of PWR or 100% of FR).

Physical limitations of reactors

PWR case

The main limitations due to the enhancement of the plutonium content are related to the coolant void effect and to the core control in nominal or transient conditions. In case of voiding, the spectrum becomes faster, the neutron flux in the thermal region tends towards zero and is concentrated in the region from 10 keV to 1 MeV. Thus, all the captures by ^{240}Pu and ^{242}Pu in the thermal and epithermal resonance disappear and the ^{240}Pu and ^{242}Pu contributions to the void effect become positive. Finally, the higher the Pu content and the poorer the Pu quality, the larger the void effect.

Concerning core control, Pu enrichment leads to a decrease in β_{eff} , the efficiency of soluble boron and control rods. In addition, the Doppler effect tends to decrease when Pu replaces U, so that in case of transients the core could diverge again if the control is not effective enough. As for the voiding effect, the plutonium degradation and the ^{240}Pu and ^{242}Pu accumulation after multiple recycling lead to spectrum hardening and to a decrease in control. One solution would be to use enriched boron in soluble boron and shutdown rods. Calculations performed with a 100% MOX-fuelled core indeed show acceptable characteristics and cooling transient accident behaviour. But the reactivity margins are not large enough to allow multiple recycling and large burn-up. Thus, the amount of plutonium recycling is limited in a standard 100% MOX PWR.

It is clear that to go further in terms of plutonium consumption and safety guarantees, more advanced PWR concepts are needed. Four different concepts are presented here:

- The high moderation reactor (HMR) concept, involving a 100% MOX core of the EPR type, and a high moderation ratio of 4 to improve control reactivity efficiency.
- The PLUTON concept with a plutonium-free uranium fuel. Uranium is replaced by other materials like inert matrix or thorium.
- The APA concept, in which some standard UOX fuel pins are replaced by annular uranium-free fuel pins.

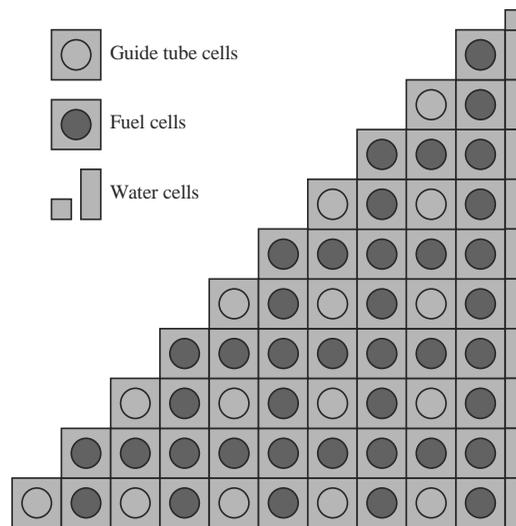
HMR concept [1-4]

To extend the moderation ratio (V_m/V_f), different solutions exist:

- To reduce the pin diameter while keeping constant the pitch or vice-versa.
- To reduce both the pitch and the pin diameter.

The last solution allows to reach the higher moderation ratio ($V_m/V_f = 4$) by changing the geometry of assemblies: 19×19 instead of 17×17 , reducing the pin diameter by about 2 mm compared to standard one (Figure 1).

Figure 1. HMR concept: 19×19 assembly, 81 guide tubes



PLUTON concept [2,4]

The PLUTON concept is based on a plutonium without uranium oxide fuel ($\text{PuO}_2 + \text{inert matrix}$). To offset the very strong and negative impact on the void and Doppler effects due to the suppression of ^{238}U , permanent moderator (ZrH_2) pins are introduced in the subassemblies (Figure 2).

Furthermore, to offset the reactivity gain due to Pu enrichment, burnable poisons such as erbium or gadolinium could be used. When thorium replaced uranium, the Doppler and the void effects are improved compared to the use of inert matrix. But this assumption implies a thorium fuel cycle, which is not of concern in this paper.

APA concept [2,4]

The APA assembly is an alternative approach for which pins with plutonium without uranium fuel, $(\text{Pu, Ce})\text{O}_2$, replace some standard UOX pins in the assemblies. The annular shape of the plutonium pins allows to reach locally large moderation ratio. Thus, the assembly contains 36 annular plutonium pins, 120 standard UOX fuel pins and 20 guide tubes (Figure 3). The initial plutonium content and uranium enrichment are optimised in order to obtain a flat power distribution in the assembly.

Figure 2. PLUTON concept

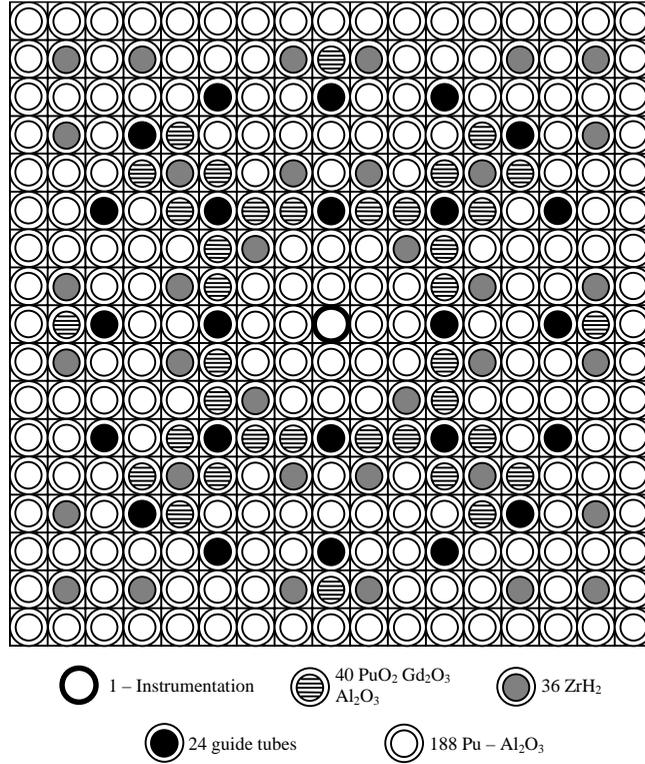
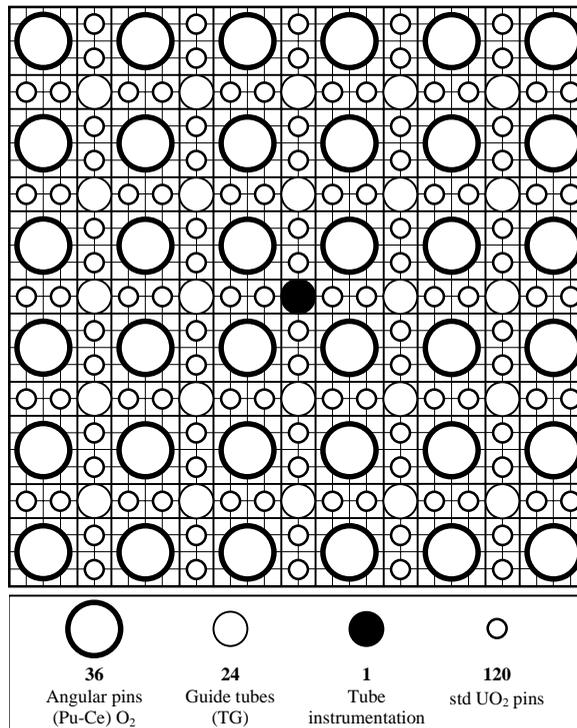


Figure 3. APA concept



When the plutonium is diluted in all the PWR park, the plutonium content of the fuel could be kept sufficiently low (< 2%) so as not to overly worsen the core performance of the standard PWR by using some additional ²³⁵U. This is the concept of the MOX fuel with an enriched uranium support (EUS-MOX fuel).

Table 1 gives the performance of each type of reactor by comparison to the standard PWR fuelled with UOX.

Table 1. Characteristics of different PWR used as Pu burner compared to a standard PWR

	Standard PWR 100% UOX fuel (Vm/Vf = 2)	HMR 100% MOX fuel (Vm/Vf = 4)	APA Pu w/o U fuel	PLUTON Pu w/o U fuel	EUS-MOX 100% MOX fuel
Fuel composition					
Initial ²³⁵ U content (%)	4.5	0.25	–	–	3.6
Initial Pu content per S/A (%)	–	6.6	10-15	11.5	1.3
Mass balance					
Pu consumption (kg/eTWh)	+29.3	-72	-76.5	-134	+6
MA production (kg/eTWh)	3.8	9.5	9	11	6
Reactivity coefficients (BOC/EOC)					
Boron efficiency (10 ⁻⁵ /ppm)	-7.3/-8.7	-9/-12	-4.9	-8/-10	-6/-7
Total void effect (%)	-65/-46	-48/-42	-14	+5/-30	-50/-43
Doppler effect (10 ⁻⁵ /°C)	-2.96	-2	-2	-1	-3
MTC *	-52/-61	-37/-43	-17.3/-36.1	-31/-33	-57/-65

* MTC: moderator temperature coefficient

The HMR and the EUS-MOX PWR present the best performance. The safety coefficients are globally acceptable and very close to that of a standard PWR fuelled with UOX. On the contrary, the PLUTON concept does not present sufficient margins as regards the coolant void effect. The APA concept presents acceptable features. Some improvement should be obtained through optimisation of the design, but both these concepts should not allow to recycle plutonium more than once or twice. Indeed, degradation of the plutonium quality by multiple recycling greatly worsens core performance.

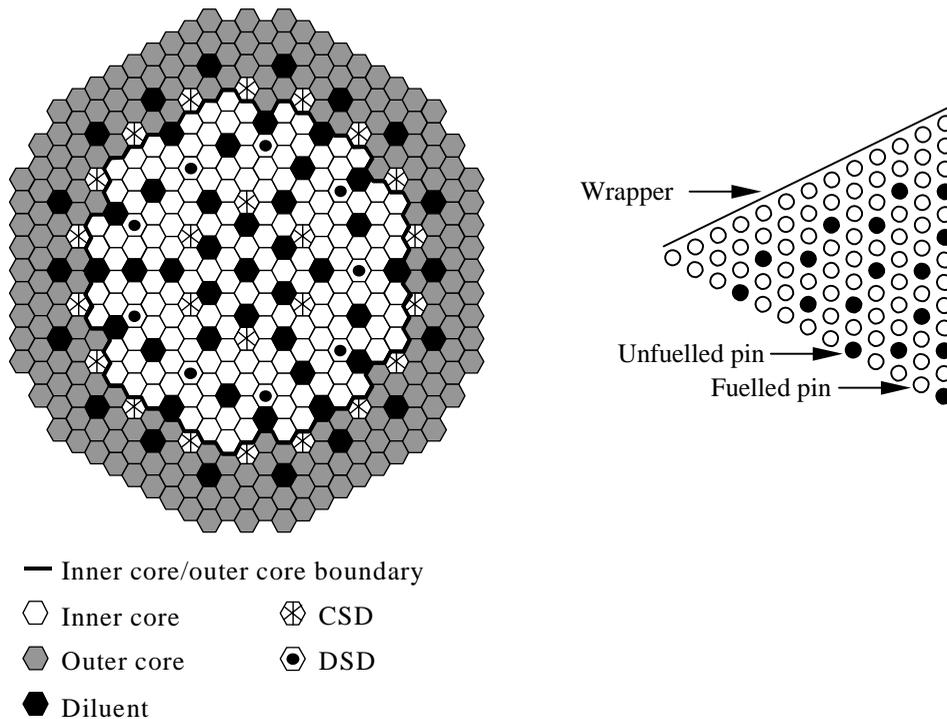
FR case

Parametric studies, carried out in the frame of the CAPRA programme to establish the design orientations of Pu burner cores, showed that a large reduction of the fuel inventory (or dilution) is necessary to be able to operate with a large core and a high plutonium content [5]. To illustrate that point, a conventional 1500 MWe breeder contains about 40 tonnes of mixed oxide, a burner based on the use of 45% Pu oxide fuel (which is the limit content if the MOX reprocessing technology must be kept) about 20-25 tonnes whereas the Pu inventory remains constant (9-10 tonnes). In the reference CAPRA 4/94 design, the fuel dilution is done firstly by pins of small diameter containing fuel pellets with a large central hole. Secondly, the heterogeneous subassemblies (SAs) contain a large number of pins (469) about one-third of which would be empty of fuel but filled with an inert or, if necessary,

a moderator material ($^{11}\text{B}_4\text{C}$ is the preferred candidate). Thirdly, the core would comprise about 50 diluent subassemblies containing no fuel. Those diluent SAs are an essential feature of the CAPRA design which allow minimisation of the sodium void worth, flattening the power map and management of the strong reactivity losses by temporary poisoning some of them. This design offers a large flexibility and makes it possible to operate the core in a wide range of breeding ratios merely by changing the content of the fuel SAs. On the other hand the main drawbacks of this conceptual option compared to a conventional FBR is the lower residence time and the management with intermediate mid-cycle shutdowns for heavy diluent removing to control the excess of reactivity.

Fortunately, the low damage dose on the clad materials of the CAPRA 4/94 core suggests the potential for an increase in burn-up and in fuel residence time. An optimisation study has been performed, including thermalmechanics studies to confirm the acceptability of fuel pin behaviour under high local irradiation and to optimise the pin-bundle configuration (Figure 4) [6]. The neutronic characteristics and performance are given in Table 2, by comparison to the reference design.

Figure 4. CAPRA concept: heterogeneous core and heterogeneous assemblies



The high burn-up core shows promising features. The fuel residence time is increased by about 45% compared to the reference 4/94 core, even if the cycle length is reduced of about 20%. The Doppler constant is improved by the decrease of the initial Pu content, but the sodium void reactivity is worse. However the change of the sodium void is sufficiently low to be balanced by simple design modifications, such as additional boron in the blankets to increase the core leakage. Furthermore, for all the design base transients (USTOP, USLOF, UFLOF) studied there is a large margin to material phase changes and the strip system should be effective. Moreover, the shutdown criteria are met for the basic CSD array indicating a possibility of operating for a whole cycle without an intermediate shutdown for reactivity management via diluent management.

Table 2. Characteristics of CAPRA 4/94 Pu burners compared to a standard FBR

	Standard FBR (EFR)	CAPRA 4/94		CAPRA 4/94 high burn-up core
Content of diluent pins	–	MgAl ₂ O ₄	¹¹ B ₄ C pins	¹¹ B ₄ C pins
Mass balance				
Pu initial content (vol %)	21	IC: 43.0 OC: 44.7	IC: 43.6 OC: 45.3	IC: 38.1 OC: 41.0
Pu consumption (kg/eTWh)	-8 (+16 core; -24 blankets)	74.2	73.4	61.5
MA production (kg/eTWh)	2.6 **	9.7 *	10.3	8.1
Core performance				
Fuel residence time (efpd)	5 × 340	3 × 285	3 × 285	6 × 220
Peak damage dose (dpa NRT Fe)	190	124	120	178
EOC void effect (pcm)	2100	1564	1395	1943
EOC Doppler effect (10 ⁻⁵ /°C)	-650 (core)	-455	-627	-569
β _{eff} (pcm)	362	324	320	317

IC: inner core, OC: outer core

* Core loaded with Pu degraded isotopic quality (recycled twice in a MOX/PWR)

** Standard PWR Pu

Multiple recycling of plutonium: scenarios studies

Scenarios studies allow us to analyse the consequences of the multiple plutonium recycling at the nuclear park scale, that is not only on the reactor performance but also on the whole fuel cycle (fuel inventory), from the MOX and UOX fabrication plants (raw materials savings) to the reprocessing plants and the final repositories (mass flow, reprocessing losses and waste radiotoxicity). As it has been explained previously, The PLUTON and the APA concepts are not of concern for this analysis. Their safety margins are not sufficient to sustain the multiple recycling of the plutonium.

Images of nuclear parks with Pu recycling

To compare different images of nuclear park involving different plutonium burners, it is necessary firstly to define a reference case (installed power: 60 GWe, annual production: 400 TWhe) and secondly to recycle plutonium so far as to reach the equilibrium state. The equilibrium state is reached when all the plutonium coming from one cycle is used in the following one (after dilution with Pu ex-UOX or Pu ex-MOX) and when the Pu isotopic vector is kept constant from one cycle to the following one. The number of burner reactors needed is determined by the balance of Pu production and consumption. Results are presented in Table 3 for nuclear parks involving a fraction of HMR with 100% MOX or CAPRA HBU reactors.

In all cases, the number of Pu burners needed to reach an equilibrium between plutonium production and consumption is quite stable during the multiple recycling. Furthermore, it does not depend significantly on the type of burner and is nearly 20%.

Table 3. Images of nuclear park of 60 GWe-400 TWhe with plutonium management

Scenario involving →	HMR	CAPRA HBU
Installed power in the nuclear park		
UOX-PWR (%)	78	72
MOX-PWR (%)	22	11
CAPRA (%)	–	17

Thanks to these comparable images, it should be possible now to analyse the feasibility and the interest of each type of plutonium burner in a real context of multiple recycling, not only in terms of safety parameters, but also of raw materials savings and mass flow in each plant of the fuel cycle. To complete the analysis, the park with 100% EUS-MOX PWR is also compared to a park containing 100% of self-sufficient fast reactors (SSFR) of the EFR-type.

Safety parameters changes

The analysis of the safety parameters changes between the first cycle (Table 1) and the equilibrium cycle (Table 4) allows us to comment on the efficiency of the different concepts for recycling plutonium indefinitely.

When plutonium is recycled indefinitely in a HMR-type PWR, the void effect is drastically worsened by about 70 to 80%. However the coefficient is kept negative and the other safety parameters remain acceptable. The plutonium consumption reaches as high a value as 100 kg/TWhe but is accompanied by a rather high minor actinides production (more than 27 kg/TWhe) and an important degradation of the plutonium.

Pu recycling in fast reactors is certainly the most suitable route. Of course, multiple Pu recycling, with dilution with uranium and plutonium of constant quality, also degrades the Pu isotopic composition and requires the increase of the fuel Pu content to offset the reactivity swing. This leads to a worsening of the Doppler effect, because of the decrease of ^{238}U fraction, and the sodium void effect, because of the hardening of the neutron spectrum due to the poorer Pu quality. However, the CAPRA cores have some interesting features which allow to improve the safety parameters such as a large unfuelled volume (near 40%). All this empty place could be used either by moderators (preferably $^{11}\text{B}_2\text{C}$) which soften the spectrum, increase the Doppler effect and reduce the void effect, or by the fuel itself in order to decrease the initial Pu content of the fuel and to reach the limit content of 45% at the equilibrium stage. Thus, the Pu quality should be less degraded because of the better production of ^{239}Pu from ^{238}U .

The high burn-up core inherently offers the following two features: to enhance the fuel residence time the Pu inventory has to be increased and to accommodate the decrease of the safety coefficients due to the high burn-up, the ratio moderator-to-fuel has to be maximised. This is partially reached by increasing the number of fuelled pins and by reducing the fuel pellet hole from 2.16 to 1.5 mm. Empty pins and SAs are filled by moderator material. An acceptable set of safety parameters have been performed, as is shown in Table 4. If need be, additional improvements, especially for the sodium void worth, should be obtained by minor design changes like an addition of boron in blankets, central plenum or more efficient moderators like hydride materials.

Table 4. Safety parameters changes of the burners when plutonium is recycled indefinitely (equilibrium cycle). Values in brackets refer to the changes between the first and the equilibrium cycle.

Scenario involving →	22% HMR	17% CAPRA HBU	100% EUS-MOX PWR	100% SSFR
Initial Pu content (%)	18	46	2	16.5
²³⁵ U content (%)	0.25	–	3.8	0.25
Core performance (BOC/EOC)				
Soluble bore efficiency (pcm/ppm)	-6 (+33%)/ -7 (+11%)	–	-5.6 (+0.4%)/ -6.5 (+7%)	–
Void effect (%)	-9 (-81 %)/ -13 (-69%)	1.7 (+35%)/ 2.7 (+11%)	-43 (+14%)/ -40 (+7%)	-15%
Doppler effect (pcm/°C)	-2.3 (-15%)	-680 (+14)/ -736 (+15%)	-3.2 (-7%)	-15%
TCM (pcm/°C)	-43 (-16%)/ -52 (-21%)	–	-59 (-3%)/ -67 (-3%)	–
Reactor mass balance sheet *				
Pu consumption (kg/TWhe)	-104	-71	0	0
Np prod (kg/TWhe)	0.2	0.3	1.6	0.5
Am prod (kg/TWhe)	22.8	12.3	4.4	3.2
Cm prod (kg/TWhe)	4.4	2.4	2.2	0.3
AM prod (kg/TWhe)	27.4	15.0	8.2	4.0
Plutonium grade: ²³⁸Pu/²³⁹Pu/²⁴⁰Pu/²⁴¹Pu/²⁴²Pu				
BOC	5/25/26/10/34	4/31/38/8/19	5/42/23/12/18	1/56/36/15/3
EOC *	5/14/27/9/45	3/27/42/7/21	“	“

* After 5 years of cooling time

For reactors filled with a low plutonium content fuel (EUS-MOX PWR and SSFR), the impact of the successive plutonium recycling is certainly the lightest. All the safety parameters remain relatively constant from one cycle to the other so that it may be assumed that the multiple recycling of plutonium is feasible in such reactors. However, once again, the thermal spectrum leads to a greater depletion of the plutonium isotopic vector and a larger production of minor actinides. Furthermore, the curium quantity is particularly enhanced: about 8 times greater than for a 100% SSFR park.

Mass flowsheet

Analysis of the mass flowsheet allows us to characterise and to compare the potential of each concept in terms of fuel savings, fuel inventory in the fuel cycle and reduction of waste radiotoxicity. Results are presented in Table 5.

When plutonium is recycled in PWR as MOX fuel with depleted uranium, the MOX fabrication capacity should be of about 200 tonnes per year. That corresponds to an evolution of 25% of the fabrication plants from the UOX technology to the MOX one, only 15% for CAPRA scenarios. Finally the plutonium use allows to save 20-30% of natural uranium and UTS. The most advantageous case is once again the CAPRA scenario.

Table 5. Annual mass flow in each cycle unit for a nuclear park of 60 GWe/400 TWhe

Scenario involving →	22% of HMR	17% of CAPRA HBU	100% EUS-MOX PWR	100% SSFR
Enrichment stage				
Natural uranium (t U)	6 360	6 080	6 640	45
Uranium enrichment (MUTS)	4.7	4.4	4.7	0
Fabrication/reprocessing stages				
UOX (t of HM)	690	645	–	–
MOX (t of HM)	190	93	880	381
CAPRA (t of HM)	–	18	–	–
Waste				
Plutonium (t)	0.03	0.03	0.02	0.06
Minor actinides (t)	3.6 (Cm = 0.5)	2.8 (Cm = 0.4)	3.3 (Cm = 0.9)	1.6 (Cm = 0.1)

* CAPRA reference scenario: Pu is recycled twice in MOX reactors before recycling in CAPRA reactors. Pu-ex MOX and ex-CAPRA are diluted with Pu ex-UOX.

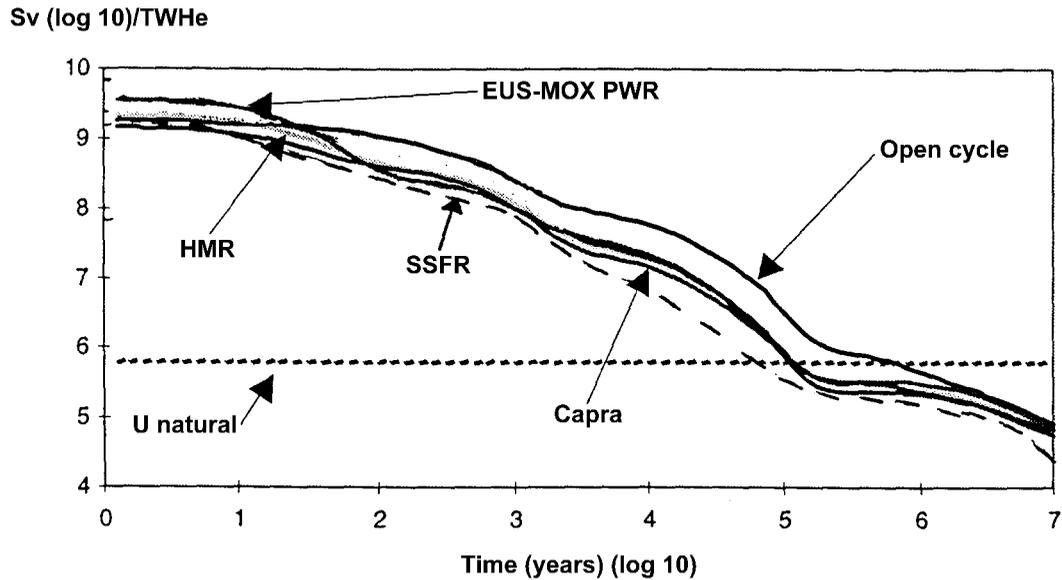
Concerning the park with 100% EUS-MOX PWR or 100% SSFR, the fuel savings and in particular the uranium savings is in favour of the FR. Only 380 tonnes of MOX fuel are needed whereas 880 tonnes have to be fabricated for the PWR park.

But the most interesting comparison is certainly in terms of minor actinides production and in particular as regards curium production. Curium management will indeed be one of the major problems to be solved in a further minor actinide partitioning and transmutation programme. Of course, plutonium consumption necessarily increases minor actinides production, whatever the burner concept, but this production must be kept as low as possible. Among the burner reactors the CAPRA ones certainly have the best potential: they are able to burn plutonium, whatever its isotopic composition, with the lowest minor actinide and curium production.

Radiotoxicity level of the final repository

To conclude the comparative analysis between the different ways of burning plutonium, an interesting point of view is to study the consequences of plutonium recycling on the level of radiotoxicity of the final repository and the evolution over years, taking into account the number of recycling and the reprocessing plutonium losses which are assumed to be 0.1% of the plutonium discharged. Figure 5 presents this evolution all along the time scale, including MOX and UOX fuel management. The park equipped by CAPRA reactors produces the lowest quantity of minor actinides (15 kg/TWhe). Consequently the best reduction of waste radiotoxicity (factor of 5) is obtained. A special concern should be devoted to the 100% SSFR park which allows the best gain factor. On the contrary, the EUS-MOX PWR park gives the highest radiotoxicity level at the beginning of storage because of the large curium production. After about 200 years, when all the ^{234,244}Cu is decayed in ^{239,240}Pu, the evolution of the radiotoxicity level becomes quite similar to the other cases.

Figure 5. Radiotoxicity level of the final repository taking into account Pu management



Conclusion

Plutonium recycling, which is of primary interest in a strategy of waste radiotoxicity reduction, can be achieved in both thermal and fast reactors. In all cases the radiotoxicity reduction factor which can be reasonably reached is 3 to 5. But the consequences on the nuclear park are not fully identical according to the type of reactor and depend essentially on the plutonium content of the fuel.

The feasibility of an indefinite plutonium recycling in a few PWR (about 20% of the electrical nuclear park) is not demonstrated yet. The best concept (HMR), designed to enhance the moderation ratio up to 4, does not allow to reach suitable coolant void parameters. Compared to a standard UOX-PWR case, the higher plutonium content leads to increase the minor production by a factor of 7. For a nuclear park of 60 GWe and 400 TWh, 200 tonnes of MOX fuel have to be fabricated. That corresponds to about 25% of the capacity of the fabrication plants turned out to the MOX technology. It is noticed that with a moderation ratio kept to 2, the Pu cannot be managed indefinitely. Only two turns could be accommodated by a standard PWR.

The concept of plutonium without uranium fuel (PLUTON) is not really feasible in a strategy of multiple recycling. But it remains interesting in the case of once-through strategy.

Finally, the superiority of the fast reactors, as burner or self-sufficient breeder, for managing plutonium, whatever the isotopic grade, is clearly demonstrated here. First of all, it is possible to increase the plutonium content of the fuel theoretically up to 100%, but more realistically up to 45% without depressing too much the safety parameters of the reactor. Secondly, the plutonium consumption, even if the Pu grade is poor, is accompanied by the smallest minor actinide production and in particular by the smallest curium one. Finally, MOX fabrication is limited to a small scale (less than 110 tonnes). Obviously, the validation phase of the CAPRA concept is not as advanced as in the case of PWR (the MOX fuel is now an industrial reality). Furthermore it has been questioned by the suspension of SUPERPHENIX. However it must be recalled that an important programme of fuel validation is still in progress in the Phenix reactor (2 pins of pelleted $(U, Pu_{0.45})O_2$ fuel under

irradiation) and BOR60 (8 pins of pelleted and vipac ($U_{0.55}, Pu_{0.45}$) O_2 fuel ready to be irradiated), but also in Siloe (1 pin of ($U_{0.55}, Pu_{0.45}$) O_2 fully examined) and HFR (2 pins of pelleted ($U_{0.55}, Pu_{0.45}$) O_2 and ($U_{0.55}, Pu_{0.40}, Np_{0.05}$) O_2 under post-irradiation examinations).

Special attention should be devoted to the 100% SSFR park. It allows to combine fuel savings, low minor actinides and especially curium production and consequently the best reduction of radiotoxicity in final repository.

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PLUTONIUM MULTIRECYCLING IN A 100% MOX CORE WITH A HIGH MODERATION RATIO

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Abstract

Recycling plutonium in standard PWRs using 100% MOX loadings brings about a degradation of the control mean efficiency. Different solutions are studied at the CEA, in order to mitigate this effect. One possibility could be to increase the moderation ratio (highly moderated reactor, or HMR, concept). Another way of improvement, while using a standard PWR, is to limit the plutonium content in the reactor, which means that to reach the target burn-ups, enriched uranium must be used (MIX concept). Other possibilities, like the advanced plutonium assembly (APA) concept are also under study at the CEA. This paper focuses on the highly moderated reactor; more details on the MIX and APA concepts can be found in Refs. [1] and [2]

Introduction

Recycling plutonium in standard PWRs using 100% MOX loadings brings about a degradation of the control mean efficiency. Different solutions are studied at the CEA, in order to mitigate this effect. One possibility could be to increase the moderation ratio (highly moderated reactor, or HMR, concept). Another way of improvement, while using a standard PWR, is to limit the plutonium content in the reactor, which means that to reach the target burn-ups, enriched uranium must be used (MIX concept). Other possibilities, like the advanced plutonium assembly (APA) concept are also under study at the CEA. This paper focuses on the highly moderated reactor; more details on the MIX and APA concepts can be found in Refs. [1] and [2]

Assembly characteristics

A parametric study carried out to assess the effect of the moderation ratio demonstrates the strong impact on the reactivity coefficients. For example, in a 100% MOX core, the increase of the moderation ratio from 2 (standard PWR) to 4, brings about an increase of the boron efficiency by a factor 2.5, and a 20% decrease on the Doppler coefficient. The moderator temperature coefficient becomes less negative as well. The main cause is the difference in the Pu inventories: increasing the moderation ratio provokes a 60% decrease of the Pu mass in the assembly (for a similar fuel management). Furthermore, the moderation ratio must be as high as possible in order to improve plutonium consumption and reduce minor actinide production.

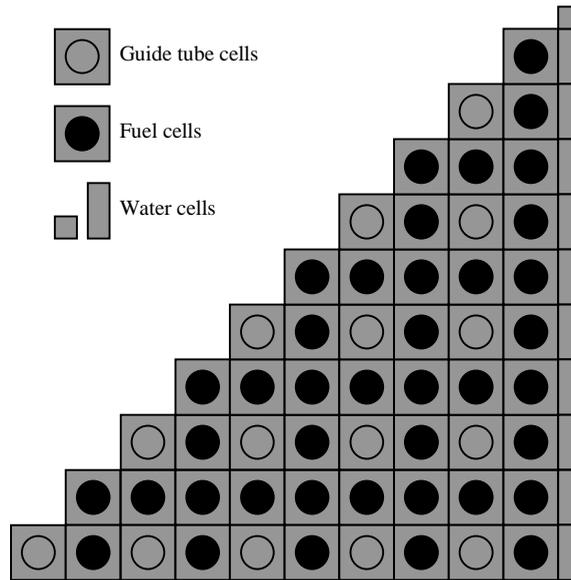
However the safety criterion concerning the nucleated boiling crisis sets limits on the surface heat flux released by the fuel rods. Thus at constant power, a geometry of the 17×17 type cannot be retained for moderation ratios higher than 3.1. But in turning to geometries of the 19×19 type (while retaining the external dimensions of the fuel assemblies), a moderation ratio of 4 can be reached. The latter configuration was chosen; it is characterised by a fuel pellet diameter of 6.12 mm and a 1.13 cm pitch. In this situation the neutron spectrum is thermalised but it remains characteristic of a MOX fuel.

Compared with a 17×17 assembly with a moderation ratio of 2, the 19×19 assembly chosen contains 40% less heavy metal, i.e. 312 kg per assembly, and the specific power is 55% higher, i.e. 56.6 W/g. Among the various solutions studied, the lattice containing 81 guide tubes (Figure 1) presents the best power flattening within the assembly owing to the regularity of the lattice. More information concerning the studies carried out at the core level, i.e. core fuel loading pattern for different fuel managements, control rod ejection, core cooling transient, etc., can be found in Ref. [3].

The advantages of the HMR with regard to 100% MOX reactors with conventional moderating ratio can be summarised as follows:

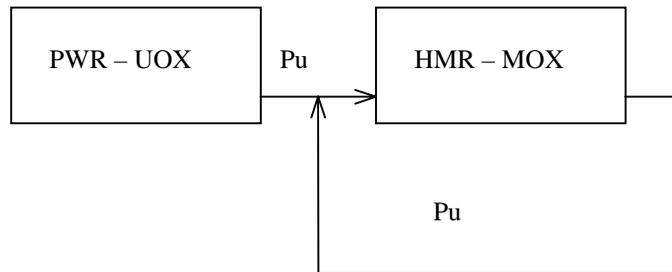
- Better efficiency of the control means (soluble boron and control rods).
- Improvement of the void coefficient.
- Increased Pu consumption per TWhe.
- Decreased minor actinides production per TWhe.

Figure 1. 19×19 assembly, 81 guide tubes, $V_{\text{mod}}/V_{\text{fuel}} = 4$



Multirecycling capabilities

The multirecycling strategy using UOX and HMR is represented on the following figure.



Three fuel managements are studied: 4×12 , 2×24 and 3×18 months corresponding respectively to average burn-up at discharge of 55.2, 60.4 and 66.3 GWd/t. Furthermore, to evaluate the impact of ^{241}Pu decay, two different cooling times (3 and 7 years) before reprocessing are taken into account for each fuel management.

The HMR is initially loaded with Pu coming from UOX reactors, i.e. containing about 65% fissile Pu, and the corresponding initial Pu enrichments are between 8 and 10%. Because the neutron spectrum is well thermalised, the Pu quality degrades very rapidly and at the end of the first cycle it contains already only about 40% fissile Pu. As a consequence, the Pu content must be increased; it goes from 8÷10% for the first cycle to 12÷14% and 14÷16% for the second and third cycles. Furthermore, the proportion of the Pu transmuted into minor actinides, instead of fissioning, increases: 15% for the first cycle, 20% and 25% for the second and third cycles.

After about 10 cycles an equilibrium is reached and the enrichments are between 25 and 30%; the fissile Pu at BOC and EOC are about 40% and 30%. The impact of the cooling time on the Pu content at equilibrium is not very large; it leads consistently to an increase of the necessary Pu

enrichment of ~5% (relative). The Pu destruction capabilities are between 90 and 100 kg/TWhe corresponding to about 18% of the Pu loaded in the reactor; unfortunately about 30÷40% of the destroyed Pu is transformed into higher actinides compared to only ~15% for the first cycle.

Even though the core is made up of 100% MOX assemblies, the reactivity coefficients for the first and second cycles are very similar to those of standard UOX PWR core because of the positive effects brought about by the increased moderation ratio. However, to make sure that a voidage of the core always leads to a decrease of the reactivity, the Pu content should be limited to about 12%; hence the number of recyclings might be limited to 2.

Conclusion

Different PWR core concepts allowing important Pu loadings are studied at the CEA. One of them is characterised by a high moderation ratio. It has a 19×19 assembly with a moderation ratio of 4. Increasing the moderation ratio of a 100% MOX core improves of the control means efficiency, the Pu consumption and decreases the minor actinides production. The Pu isotopic quality deteriorates very rapidly and the Pu content must be increased to reach the same burn-ups. However because of the void coefficient problem, the Pu content should be limited to about 12%, which means that the number of recyclings might be limited to 2.

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ASPECTS OF URANIUM RECYCLE IN LIGHT WATER REACTORS

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Abstract

The recycle of uranium might seem out of place in this conference, since it is already established in some commercial power plants and does not obviously fit with either advanced reactors or innovative fuels. However, the recycle of uranium does involve many detailed issues, many of them similar to those which will need to be addressed if minor actinides are to be transmuted in reactors or accelerator driven systems. Since uranium is an actinide, and some of the minor uranium isotopes generated during irradiation have important radiological implications, there is an obvious parallel. Therefore a review of the technical issues of uranium recycle might well raise some points relevant to minor actinides.

Introduction

Recycling the uranium recovered from reprocessing in thermal reactors is an important step in closing the nuclear fuel cycle. It provides the reactor utility a saving on uranium requirements and reduces the back-end storage costs. The main reason why utilities might consider loading reprocessed uranium fuel, however, is the economic savings that are possible. With reprocessed uranium fuel the fuel feed stock is already owned by the utility and therefore is available at zero cost. Though the costs of converting reprocessed uranium to UF₆, enrichment and fuel fabrication are higher, they are offset by the savings on natural uranium ore. The economic balance breaks even at an ore price of around 10 \$/lbU₃O₈. As it happens, this is not far from the actual price of uranium ore on the spot market at present, but this is not considered sustainable in the long term, so that significant economic savings would be expected when viewed as a long term investment.

The recycle of reprocessed uranium is already established in some European pressurised water reactors (PWRs) and has long been used in the UK's advanced gas reactors (AGRs). Many complex technical issues need to be addressed in order to get reprocessed uranium recycle established, and this paper provides a brief overview of them.

Reprocessed uranium isotopics

When thermal reactor fuel is reprocessed in current commercial reprocessing plants, the uranium is separated off from the plutonium, the minor actinides and the fission products. The reprocessed uranium (Rep U) constitutes at least 95% of the mass of material recovered and must either be stored or recycled. Compared with its initial isotopic composition before irradiation, Rep U is depleted in ²³⁵U, but it has enhanced concentrations of ²³²U, ²³⁴U and ²³⁶U and traces of fission products and minor actinides. Table 1 gives the concentrations of these isotopes in uranium ore and the range of values expected from Rep U derived from the Thorp reprocessing plant.

Table 1. Uranium isotopic characterisation of Rep U

Isotope	Concentration in natural uranium	Concentration in unenriched Rep U
U ²³²	None	0.2-5 ppb
U ²³⁴	0.006 w/o	0.015-0.025 w/o
U ²³⁵	0.711 w/o	0.6-1.2 w/o
U ²³⁶	None	0.2-0.7 w/o
U ²³⁸	99.288 w/o	98-99 w/o

²³²U is not found in natural sourced uranium (which will be referred to here as non-irradiated uranium – NIU), as it is short-lived (70 year half-life). It is generated at part per billion (1 in 10⁹) levels or lower by some very low probability transmutation chains, from ²³⁴U and ²³⁸U. Despite its low abundance, it is very important radiologically, because its decay products, particularly ²⁰⁸Tl, emit very penetrating gammas. Fabrication of fuel assemblies from Rep U requires careful attention to protect against this external gamma source.

²³⁴U is a naturally occurring nuclide which is present in NIU. It has radiological implications in all uranium fuel fabrication plants, because it is an α-emitter and therefore is an internal irradiation hazard particularly from inhalation of airborne particles. In Rep U, enhanced levels of ²³⁴U occur due to (n,2n) reactions on ²³⁵U, making Rep U a somewhat higher internal ingestion hazard than NIU.

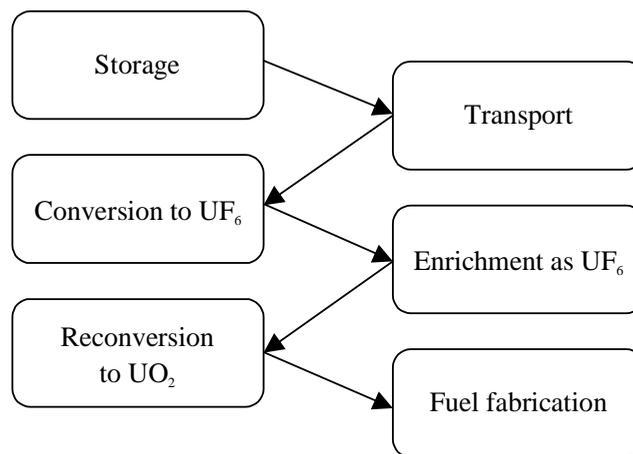
^{236}U does not occur naturally and is present in Rep U as a result of (n, γ) reactions on ^{235}U . It is not a radiological hazard, but it nevertheless important as a neutron absorber: It has a significant resonance absorption cross-section and therefore is a neutron poison. Because of this, the ^{235}U enrichment level must either be increased if the reactivity lifetime of the fuel is to be kept constant, or a shorter reactivity lifetime must be accepted.

There are also trace quantities of fission products and minor actinides in Rep U, due to imperfect separation in the reprocessing plant. These are a minor radiological issue and have no other effect on its performance.

The presence of these minor uranium isotopes has implications for all stages involved in recycling Rep U. The next section considers what these stages are.

Recycle steps

The steps involved in uranium recycle are:



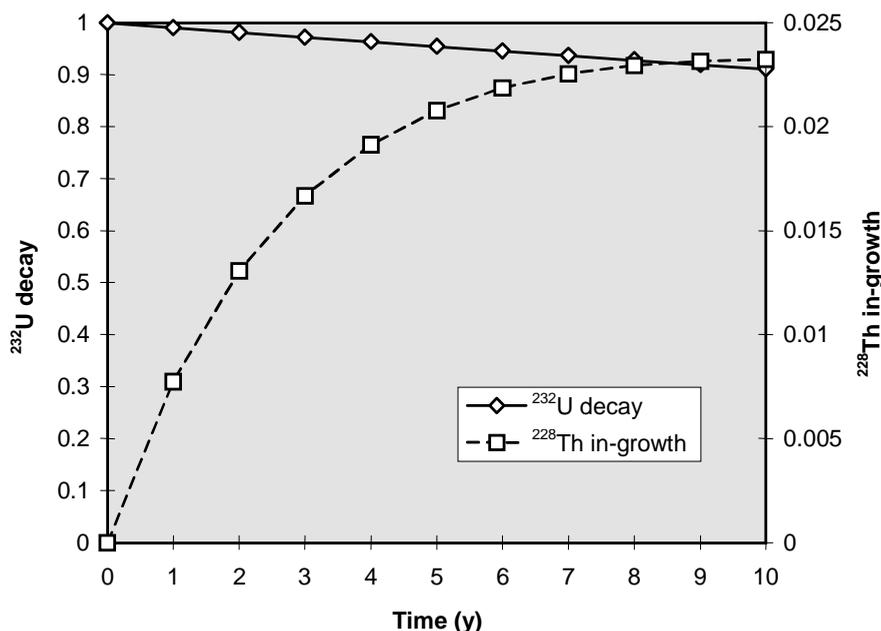
Storage

Rep U can be stored as UO_3 , U_3O_8 or uranyl nitrate. The main consideration for storage is the accumulation of ^{232}U daughter products, the gamma dose from which builds up to equilibrium within a period of 10 years, and which reaches half its equilibrium level within approximately two years. The activity of the daughter products is governed by the first isotope in the chain ^{228}Th , which has a two-year half-life. All the subsequent nuclides in the chain are very short lived and are in secular equilibrium with the ^{228}Th , meaning that each nuclide decays at exactly the rate of ^{228}Th . Figure 1 illustrates the ^{232}U decay and the ^{228}Th in-growth. Radiological control measures are therefore necessary to limit personnel doses.

Transport

Transport of Rep U as UO_3 or U_3O_8 for conversion to UF_6 requires special approvals to be in place from the appropriate authorities. An overpack arrangement may be necessary for material at the high end of the range of residual ^{235}U enrichments for criticality control.

Figure 1. ^{232}U decay and ^{228}Th in-growth



Conversion

Conversion to UF_6 is necessary if the Rep U is to be re-enriched. It is essential to convert Rep U in a dedicated conversion plant, for two reasons: first, conversion plants for natural uranium are not cleared to handle greater than 0.711 w/o material. Secondly, customers would not wish to see cross contamination between NIU and Rep U sources. For these reasons, BNFL has started construction of a dedicated conversion plant for Rep U. This is based at Springfields near Preston and has a nominal capacity of 1 200 tU per year. It uses fluorination flame reactor process. The requirement to handle greater than 0.711 w/o material necessitates safe-by-shape vessels as a criticality limiting measure. Moreover, the presence of ^{232}U daughters and traces of plutonium, minor actinides and fission products results in active residues that need to be retained in an engineered store prior to ultimate disposal as intermediate level waste.

Enrichment

Centrifuge enrichment of Rep U is already available commercially via Urenco. Centrifuge enrichment is well suited for Rep U, since the inventory of UF_6 in the cascades is very low and the residence time very short. The principal areas where Rep U has special radiological implications is in the emptying of feed cylinders in autoclaves and in filling the product cylinders.

In the centrifuge cascades the minor isotopes are enriched along with the ^{235}U . ^{236}U is actually enriched by a slightly lower factor than ^{235}U , since it is nearer in mass to ^{238}U , while ^{234}U is enriched slightly more than ^{235}U . Being so much lighter, ^{232}U is concentrated almost entirely in the product, with almost none finding its way into the tails stream. Therefore Rep U tails are not gamma active to any significant extent from ^{232}U daughters. Table 2 shows the range of isotopic concentrations in Rep U after enrichment, and the corresponding values for enriched uranium from natural ore.

Table 2. Comparison of enriched NIU and ERU Hex

Species	Concentration in enriched natural uranium	Concentration in enriched reprocessed uranium
^{232}U	None	2-25 ppb
^{234}U	0.0015-0.04 w/o	0.03-0.15 w/o
^{235}U	2-5 w/o	2.5-5 w/o
^{236}U	None	0.5-2.5 w/o
^{238}U	96-98w/o	92-97 w/o

Rep U may potentially be recycled in Candu reactors, in which case the residual enrichment is sufficient without an enrichment step. Rep U recycle in Candu may have economic advantages over LWRs, partly because of avoiding the enrichment costs, and also has the advantage that all the material is utilised in reactor and there are no tails.

Re-conversion

Re-conversion refers to the chemical reduction of UF_6 gas to UO_2 powder suitable for fuel fabrication. The external gamma dose from Rep U in this stage is very small because the evaporation of UF_6 in the autoclaves leaves behind all the ^{232}U daughters (which form non-volatile fluorides) in the UF_6 cylinder. Prompt processing of UF_6 in the reduction kilns allows no time for fresh build-up of the daughter nuclides and therefore gives a low gamma source. Strict containment is still necessary, however, because of the ^{234}U internal dose hazard.

Fuel fabrication

Fuel fabrication of Rep U requires careful attention to containment, because of the ^{234}U internal dose hazard, and also to external dose management due to the ^{232}U daughters.

For ^{232}U external dose control, appropriate levels of shielding are introduced where necessary. A limiting specification is defined for ^{232}U content, based on dose-uptake assessments for the actual plant (using the $\mu\text{Sv}/\text{hour}$ rates and actual occupancy times). Finally, careful attention is paid to minimising product throughput times to limit the build-up of ^{232}U daughters.

A particularly important issue for ^{232}U doses is that of residues recovery. UO_2 residues arise in fuel fabrication operations from pellet grinding, reject pellets and other sources. These are routinely recycled as U_3O_8 by carefully planned blending in following fabrication campaigns. This ensures that fissile material losses are minimal and that there is only a small loss of separative work. For NIU fuel fabrication there are no time limits for use of the recycled material and no restrictions on blending except the 10-20% limit which typically applies to U_3O_8 addback. However, for Rep U residues there is an inherent time limit due to build-up of the ^{232}U daughters, which means that all such material must be recycled within a short time frame. Secondly, in a fabrication plant where NIU and Rep U materials are co-processed, it is important to ensure that residues recovered from a Rep U campaign do not contaminate NIU fuel for a following customer.

These constraints require careful planning of fabrication operations. Practical schedules can be drawn up which will ensure that all the residues from a fabrication campaign for a Rep U customer are entirely incorporated in deliveries for that customer.

UF₆ cylinder washing

The cylinders used for transporting UF₆ to and from the enrichment plant are normally washed and re-used when used to transport NIU. For Rep U, however, cylinder washing needs to be delayed to allow time for the ²³²U daughters to decay. These form non-volatile fluorides that remain in the cylinder on evaporating the UF₆. After removal of the uranium, they are no longer in equilibrium and will decay according to the two-year half-life of the longest lived precursor ²²⁸Th. The external dose rate from the cylinder is increased once the UF₆ is evaporated because there is no longer any bulk shielding from the uranium and this might require remote handling procedures for prompt washing of the cylinders. The most cost effective approach is to store the cylinders in an area where access is prevented until the ²³²U heels have decayed sufficiently.

In-reactor performance

For a utility which intends to use Rep U assemblies, there may be minor fuel assembly handling issues for fresh fuel to address, because of the external dose from ²³²U. This may require modifications to fresh fuel assembly handling procedures, which might include extra shielding requirements.

Rep U is distinguished from NIU through the presence of the minor isotopes ²³²U and ²³⁶U. Once loaded in-reactor, the ²³²U has no effect on the performance of the fuel and core, there being no significant neutron absorption at ppb levels. ²³⁶U, however, is present in enriched Rep U fuel at concentrations of typically 1.5 w/o. At this concentration it has a significant neutron absorption effect, in large part through its resonance cross-section. If the reactivity lifetime of the fuel is to be maintained the same as for NIU, it is therefore necessary to compensate with a higher ²³⁵U enrichment.

Because of the resonance absorption, the precise effect of ²³⁶U depends on the reactor system. For light water reactors (LWRs) there is a rough equivalence formulation that for every four ²³⁶U atoms, the ²³⁵U content should be increased by one atom. This is only an approximate guide, and a detailed lattice code calculation is required to verify it for the particular combination of reactor and fuel management scheme in use. With ²³⁶U at the typical 1.5 w/o level, an increase in ²³⁵U enrichment of around 0.4 w/o is typically required. This will affect the core characteristics, since the Rep U assembly will have a higher thermal neutron absorption cross-section on account of the combined effects of the ²³⁶U and the enhanced ²³⁵U enrichment. The effect on the core performance is modest in degree, but nevertheless should be accounted for in the nuclear design and safety assessment analyses for the core. Although many of the important design and safety parameters such as power peaking factors and reactivity coefficients are modified to a small extent by Rep U, the impact on the safety assessments are usually minimal. This is especially the case for a reactor where the safety case is based on a probabilistic risk assessment, as changes to safety parameters tend to have as many beneficial as detrimental impacts and overall there is a cancellation over the spectrum of fault sequences analysed.

²³⁴U has a comparable resonance absorption cross-section to ²³⁶U, but it is much less of an issue due to the fact that it is present at lower concentrations (typically 0.1 w/o). Moreover, ²³⁴U is in time transformed by neutron capture to ²³⁵U and so eventually contributes to the fissile content. This makes the reactivity equivalence coefficient for ²³⁴U very dependent on discharge burn-up; it decreases with increasing discharge burn-up as more of it is converted to ²³⁵U.

Another consideration that arises is the 5.0 w/o criticality limit that typically applies to present fuel fabrication plants. There are already some PWR utilities using enrichments as high as 4.95 w/o

and some BWR utilities using bundles with rods at the same level. For such rods the criticality ceiling would prevent over-enrichment of the ^{235}U being applied and there will necessarily be an adverse impact of reactivity lifetimes for the assemblies.

There are no fuel rod mechanical/thermal performance issues for Rep U.

Overall, the main impact to a utility of accepting Rep U assemblies for the first time will be the need to re-examine the safety case and demonstrate that the presence of Rep U will not invalidate it. This may be a major barrier to a utility, not least because of the implications in terms of man-effort, but also because utilities are often reluctant to invite such re-examinations by their licensing authority. For most plants, licensing Rep U loading should not prove problematic and would be a one-off cost that should not significantly diminish the economic incentive in favour of Rep U.

Once irradiated, spent Rep U fuel will have a different isotopic characterisation to spent NIU fuel. The ^{235}U content at discharge will be somewhat higher, due to the higher initial ^{235}U enrichment for the same reactivity lifetime. The presence of ^{236}U from the start of irradiation leads to a higher inventory of ^{238}Pu , which is important as a heat source. In addition, the ^{232}U remains in the fuel at the several ppb level present on fuel loading. These latter two considerations will influence the back-end treatment of Rep U assemblies. As regards spent fuel handling and storage in the ponds, the extra heat output from ^{238}Pu is a small effect and Rep U assemblies are not significantly different to conventional NIU assemblies.

Concluding remarks

This paper has given a brief review of the technicalities of uranium recycle. Are there any learning points for partitioning and transmutation of minor actinides, given that uranium recycle is very similar in principle to recycling minor actinides homogeneously in fuel?

The first point is that recycling uranium, simple though it is in principle, nevertheless raises many detailed operational and licensing issues at all stages. Some of these issues, although straightforward, can nevertheless take considerable time and effort to address and clear with the relevant licensing authorities. Transport issues in particular can take a long time to resolve, particularly if there is a need to gain approval in more than one country.

The core performance implications of reprocessed uranium highlight how a relatively minor change to the fuel isotopic characterisation can have a noticeable impact in thermal reactors. The irradiation of minor actinides in thermal reactors will likely have a much larger impact that has the potential to have a significant effect on core operations. It will be important for the utility to have a complete understanding of all these implications and this aspect should not be neglected in the early stages of research and development.

The isotopic characterisation of low abundance isotopes such as ^{232}U can be very sensitive to the specific irradiation history and are therefore difficult to predict. Good knowledge of the isotopic characterisation is important for Rep U and may be even more so for minor actinides.

A difficult issue to address with minor actinides will be the dose budgets in fuel/target fabrication. These are difficult to construct without detailed knowledge of the fabrication plant design and occupancy times. This aspect has proven time consuming to establish for Rep U plants, and the difficulties will be much more acute for minor actinides.

The area of residues arisings and recovery/disposal in fuel fabrication operations requires careful attention with uranium recycle. By their nature, minor actinides are potentially more of a concern in this respect and it will be necessary to establish a clear understanding of the quantities and forms of residues arising and their implications. It is even conceivable that the production of intractable residues/waste forms arising from minor actinide recycle might actually undermine the rationale for recycling them in the first place, so considerable attention is merited to this particular question.

Finally, uranium recycle clearly shows how important it is to have a clear economic incentive for utilities to consider using a novel fuel cycle. Although there are strategic and other benefits of recycling uranium, the deciding factor for all potential users is that the economics must be at least competitive with a conventional NIU fuel cycle and preferably demonstrate an advantage. If minor actinide burning is to be realised in any future reactors, the utilities will need to be able to realise economic benefits by so doing. Careful attention needs to be directed to devising mechanisms whereby utilities will be able to see a direct benefit by co-operating in the minor actinide burning programmes.

THE CONCEPT OF A BREEDING PWR

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Abstract

A breeding PWR which is based on the present PWR technologies was designed using heavy water coolant and a very tight lattice core. Feasibility studies for the breeding PWR were performed in the nuclear design, thermal hydraulic design, fuel assembly design, safety evaluation, core internal design and plant equipment design. There are many problems to be studied, but no essential problems as regards the feasibility of this reactor.

Introduction

A summary of research and development for the Japanese PWR is shown in Figure 1. A first generation PWR started as from 1970, then step by step development resulted in the present advanced PWR (APWR) [1]. The APWR can be used for full core MOX utilisation purposes [2,3,4]. Advanced control rod design has also been developed for APWR [5].

After the APWR, the next generation PWR is planned to be developed as the standard future PWR [6]. One design option of the next generation PWR is a high conversion PWR. Many studies were performed for the tight lattice core, high conversion PWRs [7-10].

Heavy water coolant is another selection for high conversion PWR [11-14].

Recently, some delay in sodium coolant FBR development has been estimated in Japan. Therefore, there is a need to study the high conversion or breeding LWRs as an alternative option.

Recent studies about the breeding PWR are summarised as follows.

- A light water coolant tight hexagonal lattice with about a 0.5 mm fuel rod to rod gap will give a breeding ratio of around 1.0.
- A heavy water coolant tight hexagonal lattice with about a 1 mm rod to rod gap will give a breeding ratio of around 1.1.

This paper describes the design status of the heavy water breeding PWR.

Objectives

The purposes of this study for the breeding PWR are as follows:

- Multi-recycle Pu utilisation in PWR.
- Natural uranium savings.
- Simplified reprocessing with U, Pu, actinides, long-lived fission products (LLFPs).

If this type of breeding PWR is selected, Pu fissile ratio after burn-up is almost the same as that as before burn-up, which is illustrated in Figure 2. Therefore, multi-recycle Pu utilisation is possible without any reactivity demerit.

If the breeding PWR with the breeding ratio of around 1.1 is introduced after 2030, natural uranium consumption in Japan does not increase over one million tonnes until 2100. Therefore, natural uranium savings are attained using these breeding PWRs, which can be seen in Figure 3. Simplified reprocessing is the next stage target of commercial reprocessing plants; this is closely related to low decontamination factor fuel contents with actinides and LLFPs.

The breeding PWR enables to use such low decontamination factor fuels because of a very fast neutron spectrum.

Design objectives of this breeding PWR are as follows:

- The breeding ratio is around 1.1.
- Coolant void coefficient is negative during reactor operation.
- Reactor power is up to 1 000 MWe.
- Low development cost and low construction cost.

Figure 4 shows the breeding ratio vs. hydrogen (or deuterium) to the heavy metal number density ratio, which shows that the heavy water coolant and tight lattice core maintain the breeding ratio above 1.1.

A selection of heavy water coolant and a tight lattice core design can fulfil the above objectives using the improvements of present PWR technologies.

Feasibility of a breeding PWR

The design areas listed in Table 1 are studied in order to examine the feasibility of a breeding PWR.

Table 1. Design areas studied in order to examine the feasibility of a breeding PWR

Nuclear design	Breeding ratio, reactivity, void coefficient, etc.
Fuel assembly design	Very tight lattice fuel assembly, fuel integrity, etc.
Thermal hydraulic design	Thermal margin, core pressure drop, etc.
Safety evaluation	Non-LOCA, LOCA safety evaluations, etc.
Core internal design	Very tight lattice core internals, etc.
Plant equipment	Heavy water system equipment, etc.

Nuclear design

- *Breeding ratio*

The breeding ratio is around 1.1, which is shown in Figure 4. The core is composed of seed fuel region and blanket fuel region which is necessary from negative void coefficient design.

- *Reactivity*

Reactivity decrease vs. burn-up is about 3% $\Delta\rho$, which is very small compared to the present PWR. Figure 5 shows the typical burn-up characteristics of this reactor.

The Pu fissile ratio is about 12 wt% for seed fuel.

Reactivity control in steady operation is maintained by enriched $B^{10}B^4C$ control rods. For accident reactivity control purposes, however, enriched B^{10} chemical shim is used.

- *Void coefficient*

Negative void coefficient is designed by the following design combination:

- Seed and blanket fuel heterogeneous core design.
- Skewed power shape design to peripheral core area.
- Neutron absorber positioning in core and reflector area.
- Hydrogenous material positioning in core such as ZrH₂ rod.

There is much uncertainty in the void coefficient calculations for complete void condition. It is necessary to perform extended studies for verification of this void coefficient analysis [15].

Fuel assembly design

- A very tight hexagonal lattice fuel assembly is manufactured in test assembly size.
 - A fuel assembly of rod to rod gap with about 1 mm using spacer grid design is manufactured in test scale size.
 - A fuel assembly of rod to rod gap with about 0.5 mm using spiral fin clad design is manufactured in test scale size.
- Fuel integrity is evaluated as follows:
 - Stainless steel clad is used because of low neutron capture in fast energy spectrum.
 - Steady state fuel integrity is evaluated with normal PWR fuel integrity evaluation study tools.
 - Irradiation test studies will be necessary in the future.
 - Fuel assembly flow tests will be necessary in the future.
 - Ultra high burn-up fuel design is preferable for this fast breeder PWR because of long core life.

Thermal hydraulic design

- *Thermal margin*

Thermal margin is estimated as follows:

- Estimation from DNB tests for HCPWR with very tight lattice.
- Estimation using KfK DNB correlation for very tight lattice.

It is possible to design the very tight lattice core with nearly the same thermal margin to the present PWR design.

DNB tests for very tight lattice core will be performed to verify the right thermal margin.

- *Core pressure loss*

A rough evaluation of core pressure loss and its effect on core internal design, RCP design, etc. were performed. Core pressure loss will be examined by flow tests.

Safety evaluation

Preliminary accident analyses for the breeding PWR were performed (LOCA and non-LOCA accidents).

- Evaluation of effects from core parameters change.
- Evaluation of effects from very tight hexagonal lattice core.
- Safety equipment improvements will be applicable to the severe events.

LOCA tests for a very tight lattice core will be performed.

Core internal design

The core internal design for a very tight hexagonal lattice core were evaluated.

- APWR large reactor vessel is used.
- Core internals for hexagonal lattice fuel assembly and large diameter control rod are being designed.

Flow tests for the newly designed core internals will be performed.

Plant equipment

Plant equipments are nearly the same as those of APWR except for the following:

- Heavy water system equipments are added (heavy water concentration control system, heavy water separation equipment, etc.).
- RCP development with higher head.

Problems to be studied

The problems that should be studied in order to develop the above breeding PWR are listed in Table 2.

Table 2. Problems that should be studied in order to develop the breeding PWR

Nuclear design	Verification of design methods by critical experiments, verification of void coefficient, etc.
Thermal hydraulic design	Verification of thermal margin by DNB tests, etc.
Fuel assembly design	Manufacturability tests, flow tests, irradiation tests, etc.
Safety evaluation	Heat removal experiments in LOCA tests, etc.
Core internal design	Core internal flow tests, control rod insertion tests, etc.

Summary

A feasibility study for a breeding PWR with heavy water coolant and nearly the same PWR hardwares allows for the possibility of the following designs:

- Very tight hexagonal lattice fuel assembly with rod to rod gap around 1 mm.
- Seed and blanket fuel heterogeneous core.
- Nearly the same linear heat rate to that of the present PWR.
- Nearly the same MOX fuel design.

The breeding PWR will be one of our selections for twenty-first century reactors if the above problems continue to be studied steadily.

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Figure 1. Japanese PWR improvements

Present
↓

	1970s	1980s	1990s	2000s	after 2010s	Future options
Japanese PWR improvements	First generation PWR 9 units <ul style="list-style-type: none"> • US technology • Japanese made 	Second generation PWR 7 units <ul style="list-style-type: none"> • Reliability improvement • Safety improvement 	Third generation PWR 7 units <ul style="list-style-type: none"> • Original technology • Economy improvement 	Fourth generation PWR <ul style="list-style-type: none"> • Large unit power • Economy improvement 	Next generation PWR <ul style="list-style-type: none"> • Operation flexibility • Maintenance improvement 	Breeding PWR
Core and fuel improvements	Fuel fabrication in Japan	Reliability improvement	High burn-up (48 Gwd/t)	High burn-up Phase 2 (55 Gwd/t) MOX fuel utilisation	High burn-up Phase 3 (~70 Gwd/t) Full MOX utilisation	Tight lattice fuel assembly Hexagonal fuel assembly

Figure 2. Pu fissile ratio exchange

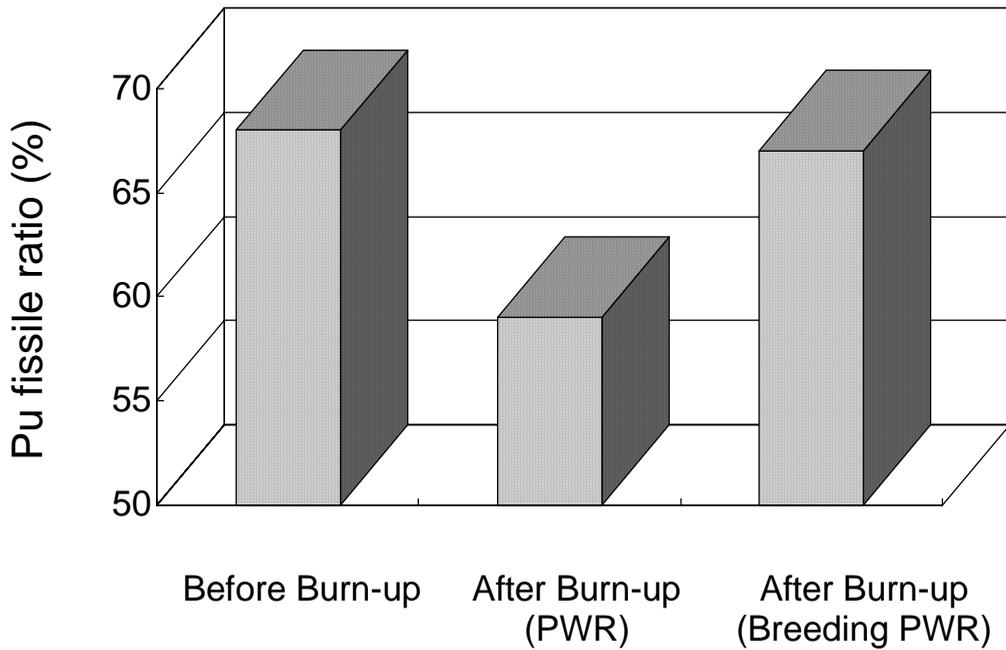


Figure 3. Natural uranium utilisation in Japan

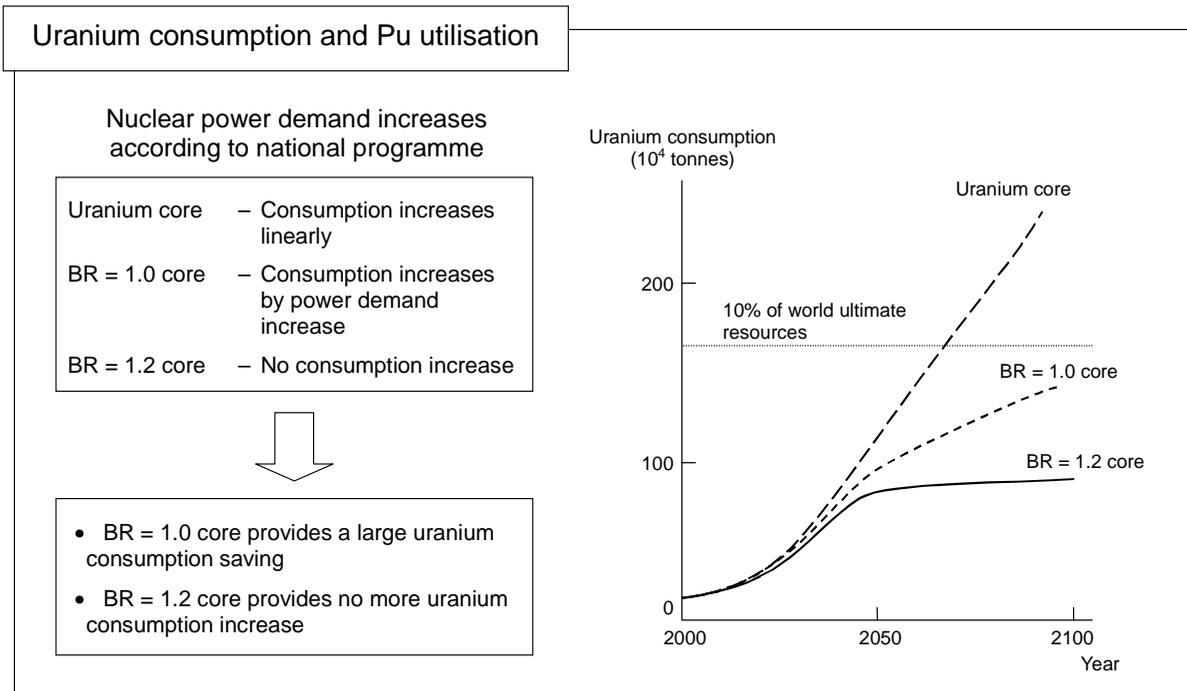
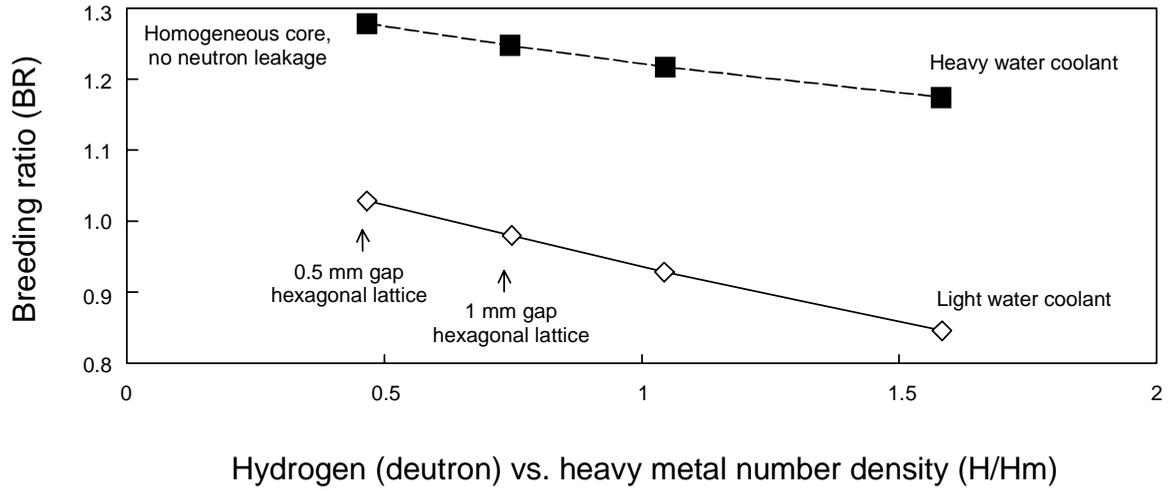
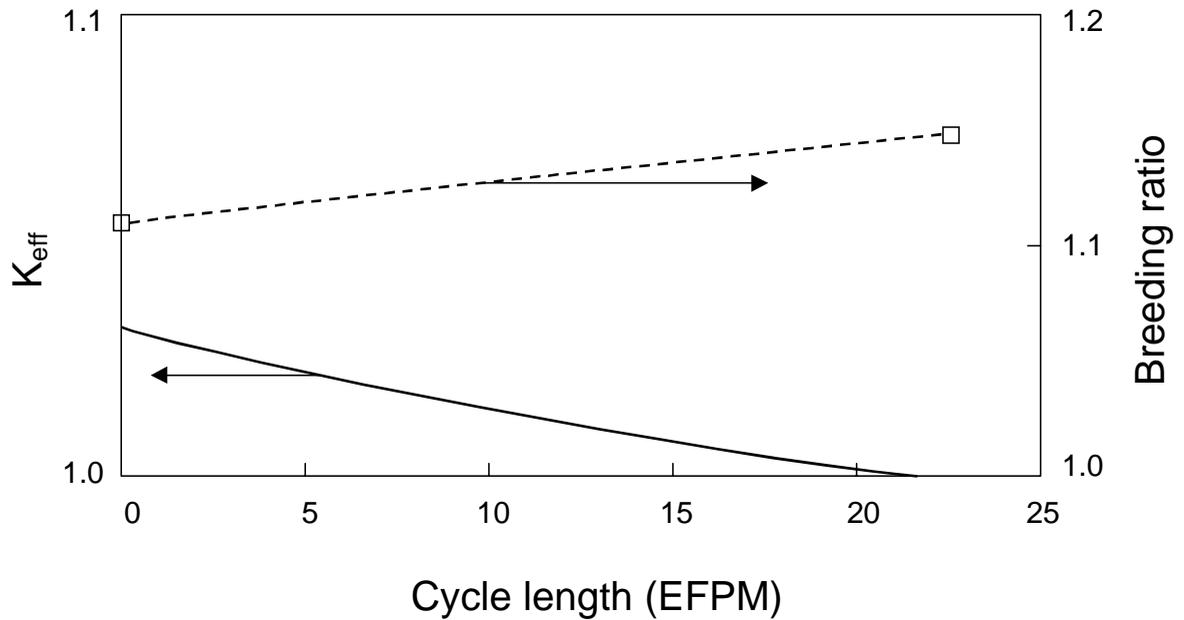


Figure 4. Breeding ratio vs. H/Hm



Flexible change in BR is possible using heavy water contents change

Figure 5. Burn-up characteristics



STUDY ON FAST SPECTRUM BWR CORE WITH BREEDING CHARACTERISTICS

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Abstract

In BWR cores, tight lattice configurations result in a very hard neutron spectrum. Although this neutron spectrum could allow breeding characteristics when MOX fuels are applied, it becomes difficult to maintain a negative void coefficient because the increase of η value for ^{239}Pu and the fast-fission effect in ^{238}U make the reactivity of the core increase when the void fraction of the core is increased. The major subject in this paper is to find core specifications that can resolve the problem of an inverse coupling between void reactivity and plutonium breeding. We have adopted a tight lattice BWR core and a neutron streaming channel in order to achieve both the breeding characteristics and the negative void reactivity effect. In the paper, we named this BWR core a breeding BWR (BBWR). The objective of this study is to clarify the core characteristics of the BBWR.

At first, we assumed the diameter of the BBWR core should be approximately the same as that of conventional BWR cores so as to suppress plant construction costs. The common way to enhance the conversion ratio of the light water reactor is to adopt tight lattice fuel assemblies. A low water to fuel volume (W/F) ratio provides a hard neutron spectrum, and the possibility of breeding increases, while the void reactivity coefficient tends to be positive. It is known that the decrease of the core height will improve the void reactivity in a hard spectrum core through the design experience of the fast breeder core. But the deduction of the core height leads to the increase of core diameter even in a tight lattice core, which has a relatively large number of pins per unit area. Therefore we have introduced a new core concept in order to improve the void reactivity coefficient under the restriction of core diameter by adopting a neutron streaming channel.

The height of the BBWR core, 1.6 m, is decreased to less than half that of the conventional BWR, and partial length fuel assemblies whose active fuel length is about half that of the normal fuels are arranged by one-third of the whole core. The streaming channel located at the upper part of the partial assembly will enhance axial neutron leakage when the void fraction is increased, the neutrons of which originally have leaked out through the side of the normal assemblies and the top of the bundle of the partial assemblies. The cavity can in the streaming channel not only provides a streaming path for the leaked neutrons from the fuel, but also expels water in the channel, which leads to a larger breeding ratio. In order to make the spectrum of BBWR core hard, we have adopted a triangular tight lattice with thin stainless steel cladding pins in a large square channel box where the overall water to fuel ratio is reduced to about 0.5.

Our analysis – including a void coefficient evaluation with Monte Carlo code – shows that the BBWR core with an axial blanket thickness of 40 cm achieves a breeding ratio of 1.04 and a void coefficient of $-1 \times 10^{-4} \Delta k/k$ per 1% void increase.

Introduction

The Liquid Metal Fast Breeder Reactor (LMFBR) has been developed as the type of the next generation power reactors in several countries, including Japan, for a long time. Today, the development schedule is being reassessed in these countries due to difficulties of economical or technological consideration.

If economical or technological considerations are the main issue, the types of near future reactors are similar to current reactors and are of the same technology based on the current experience. In this respect, the water cooled reactor could be considered as a possible candidate. In particular, BWR has an advantage in breeding of fissile materials as boiling in water decreases the amount of moderator to induce a hard spectrum that is essential to breeding.

In BWR cores, tight-lattice configurations result in a very hard neutron spectrum and less than 1% of the neutrons have energies below 1 eV. Although this neutron spectrum could allow breeding characteristics when MOX fuels are applied like LMFBR, the increase of η value for ^{239}Pu and the fast-fission effect in ^{238}U made the reactivity of the core increase when the void fraction of the core is increased. The major concern in this paper is to find core specifications that can resolve the problem of an inverse coupling between void reactivity and plutonium breeding. So, in the paper, we named the core breeding BWR (BBWR). We have adopted a tight lattice BWR core and a neutron streaming channel in order to achieve both the breeding characteristics and the negative void reactivity effect.

In the paper, we also assess the characteristics of BBWR when it was applied to an innovative fuel cycle system named BARS, which stands for BBWR with an advanced recycle system. In the BARS, the light-water reactor spent fuel is recycled as a fuel for BBWR through oxide dry processing and the vibro-pac fuel fabrication. The fuel provided from the dry processing is contaminated by rare earth (RE) elements, which have been produced as fission products and some amount of them are accompanied with heavy metal elements in the process.

The objective of this study is to clarify the core characteristics of the BBWR both in the conventional recycle system like Purex without RE contamination, and in the BARS with RE contamination.

BBWR core description

At first, we assumed the diameter of the BBWR core should be approximately the same as that of conventional BWR cores so as to suppress plant construction costs. The common way to enhance the conversion ratio of the light water reactor is adopting tight lattice fuel assemblies [1]. A low water to fuel volume (W/F) ratio provides a hard neutron spectrum. In general, when the neutron spectrum becomes hard, then the η value increases and the possibility of breeding increases.

On the other hand, the void reactivity coefficient has a tendency to be positive, because the spectrum becomes harder and η increases more when void fraction increases. It is known that a decrease in the core height will reduce the void reactivity in a hard spectrum core through the design experience of the fast breeder core. But the deduction of the core height leads to the increase of core diameter even in a tight lattice core, which has a relatively large number of pins per unit area. Therefore we have introduced a new core concept in order to improve the void reactivity coefficient under the restriction of core diameter by adopting a neutron streaming channel described below.

Figure 1 shows a core profile of BBWR in a large BWR plant (reactor thermal output of 3 926 MWt, core diameter of 5.2 m, core height of 1.6 m). The height of the BBWR core is decreased to less than half that of the conventional BWR, and partial fuel assemblies whose active fuel length is about half that of the normal fuels are arranged by one-third of the whole core as shown in Figure 2.

The number of assemblies were determined under the criteria that the maximum linear heat generation rate should not exceed about 400 W/cm. The streaming channel located at the upper part of the partial assembly will enhance axial neutron leakage when void fraction is increased, the neutrons of which originally have leaked out through the side of the normal assemblies and the top of the bundle of the partial assemblies as shown in Figure 3. The cavity can in the streaming channel not only provides a streaming path for the leaked neutrons from the fuel, but also expels water in the channel, which leads to a larger breeding ratio. Figure 4 shows the cross-section view of a BBWR fuel. In order to make the spectrum of the BBWR core hard, we have adopted a triangular tight lattice in a large size channel box where the water to fuel ratio is reduced to about 0.5. Gap water between the channel box has also been expelled using a control rod with a follower in which helium gas is filled up as shown in Figure 5. A water removal plate attached on the outer side of channel box (shown in Figure 4) also helps to decrease W/F ratio. A set of typical BBWR core specifications is displayed in Table 1. For the purpose of increasing fuel volume fraction, stainless steel (SUS) has been applied to the material of claddings and channel boxes because its strength is higher than that of zircaloy, while the neutron absorption penalty is small due to a harder spectrum. These concepts enable a W/F ratio as low as 0.49 in the BBWR core.

Core characteristics for MOX fuel without rare earth

Figure 6 shows a BBWR neutron spectrum compared with an LMFBR spectrum, where both results have been calculated by the method shown in Table 2 [2]. Since the whole shape of the spectrum indicates the feature of a fast spectrum, the FBR core analysis method will provide adequate results. Table 3 shows an evaluated η value of ^{239}Pu in BBWR, which suggests breeding is possible under appropriate conditions.

The breeding ratio at the equilibrium cycle in four batch refuelling core was analysed by R-Z two-dimensional diffusion burn-up calculation with the JENDL-3 library as shown in Table 2.

At first, we assumed that the MOX fuel loaded to BBWR has no RE contamination. Pu isotope composition was assumed to be that of the LWR discharged fuel shown in Table 4. The axial blanket fuel was assumed to use depleted uranium that is located at both ends of the core except at the top of the partial assemblies.

Burn-up characteristics of the BBWR core (shown in Table 1) were analysed under the condition that the core average void fraction be 60% and the fuel smear density be 90%, in addition to the condition for fuel composition mentioned above.

In Table 4, the Pu enrichments of load fuel were determined to be critical at the end of equilibrium cycle (EOEC). The enrichment of the outermost assemblies has been set higher than the other fuels aiming at radial power flattening, while no axial enrichment distribution has been adopted.

The breeding ratio at the middle of equilibrium cycle (MOEC) is 1.04, and this indicates the possibility of breeding in BBWR. It is considered that the small burn-up reactivity of 1.4% $\Delta k/k$ is due to the large internal conversion ratio.

Figure 7 shows the dependency of breeding ratios on some different core parameters of BBWR [3]. It suggests that the breeding ratio largely depends on the void fraction, the thickness of the axial blanket and the fuel smear density, while the breeding ratio can exceed unity under the appropriate conditions.

The power peaking factor of the BBWR derived from Table 4 is about 1.9, which indicates a relatively flat power shape due to its fast spectrum, while the burn-up of 49 GWd/t shows as large as that of conventional BWR.

The void coefficient of BBWR has been evaluated with a three-dimensional Monte Carlo code MVP [4] and the JENDL-3 library as shown in Table 5.

In the Monte Carlo calculation, the whole core was modelled assuming axial void distributions, where the flow rate of each type of assembly was determined to meet the criteria for the minimum critical power rate ratio of 1.3 under the assumption that the power and flow rate of the partial assembly are half of the normal assembly. The resultant void fraction in core average at the rated power is 61%, which is approximately the same as that of the breeding ratio calculation conducted in Table 4. The void reactivity coefficient was derived from the reactivity between the rated power and 50% power. Figure 8 shows the cross-section model of the streaming channel in the calculation. The gap between the channel box and cavity can was set as a variable.

The result shown in Table 6 indicates that a negative void coefficient can be achieved by optimising the streaming channel structure.

Core characteristics for MOX fuel with rare earth

As mentioned in the introduction, in the oxide dry processing of BARS, RE elements in the FP which contaminate the recycled fuel may deteriorate the core characteristics such as the breeding ratio. Here it is assumed the above BBWR was adopted as a core in BARS [5].

Table 7 presents the fuel composition when the load fuel is fabricated for BBWR. Pu enrichment was again determined to be critical at EOEC. The fraction of RE was taken as a variable depending on the decontamination factor, which is the ratio of initial RE to residual RE in the process. Two-dimensional diffusion burn-up calculations were conducted to obtain the burn-up characteristics.

The results presented in Figure 9 and Figure 10 suggest that the effect on the Pu enrichment and breeding ratio by the RE contamination is small when the DF (decontamination factor) is greater than 5.

Conclusions

BBWR (breeding BWR) core, which can be adopted to the BARS (BBWR with advanced recycle system), has a fast neutron spectrum and a neutron-leakage-enhanced core configuration, and BBWR can achieve both breeding characteristics and a negative void coefficient when the detailed design parameters are optimised.

Acknowledgements

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Table 1. Core specifications of BBWR

Item	Unit	BBWR	Current ABWR
Power	MWe	1 356	1 356
Steam temperature	°C	287	287
Reactor pressure	MP	7.2	7.2
Core equivalent diameter	m	5.2	5.2
Core height	m	1.6 (normal assembly) 0.8 (partial assembly)	3.7
Assembly numbers	–	208 (including 132 normal assemblies and 76 partial assemblies)	872
Number of fuel pins	Assembly	658	62
Pin lattice type		Triangle	Rectangular(8 × 8)
Pin diameter	mm	11.2	12.3
Cladding thickness	mm	0.3 (SUS)	0.86 (Zr)
Pin gap	mm	1.3	4.0
Pin pitch	mm	12.5	16.3
Blade width of control rod (CR)	mm	250	250
CR thickness	mm	8	8
Inner width of channel box	mm	303	134
Bundle pitch	mm	317	155
Flow area to fuel area ratio		0.49	3.1

Table 2. Analysis method for burn-up characteristics

Item	Condition
Cross-section library	JENDL-3
Code	Two-dimensional RZ diffusion and burn-up calculation Homogeneous model
Number of energy groups	25
Burn-up calculation model	Two-dimensional RZ Uniformly scattered model Control rod at fully withdrawn position
Pu enrichment adjustment	Reactivity at EOEC: 0.2% Δk

Table 3. η value of BBWR

Item	BBWR	Typical LMFBR
²³⁹ Pu capture cross-section	1.4	0.5
²³⁹ Pu fission cross-section	3.1	1.9
Emission neutrons per fission	2.9	2.9
η	2.0	2.3

Table 4. Burn-up characteristics of BBWR

Item	Unit	BBWR
Void fraction	%	60
Fuel smear density	%	90
Refuelling scheme	–	1 year × 4 batches
1) Load fuel Pu enrichment		
Inner assembly	%	16.6
Outermost assembly	%	19.8
2) Burn-up reactivity swing	% Δk/k	1.4
3) Breeding ratio (MOEC)		
Core		0.92
Axial blanket		0.12
Total		1.04
4) Core average discharge fuel burn-up	GWd/t	49
5) Maximum linear heat rate		
BOC		
Inner assembly	W/cm	391
Outermost assembly	W/cm	391
EOEC		
Inner assembly	W/cm	397
Outermost assembly	W/cm	367
6) Core average linear heat rate		
BOEC	W/cm	210
EOEC	W/cm	209
7) Heavy metal inventory	t	164

*1) Pu isotope composition: ($^{238}\text{Pu}/\text{Pu}$)($^{239}\text{Pu}/\text{Pu}$)($^{240}\text{Pu}/\text{Pu}$)($^{241}\text{Pu}/\text{Pu}$)($^{242}\text{Pu}/\text{Pu}$) = 0.027, 0.479, 0.303, 0.096, 0.085; (Am/Pu) ($^{241}\text{Am}/\text{Am}$) = 0.01, 1.0

*2) U isotope composition: ($^{235}\text{U}/\text{U}$)($^{238}\text{U}/\text{U}$) = 0.002, 0.998

Table 5. Analysis method for void coefficient

Item	Condition
Cross-section library	JENDL-3
Code	MVP
Method	Continuous energy Three-dimensional Monte Carlo
Model	Initial core Fuel and CR region: homogeneous model Streaming path: heterogeneous model
Number of neutron histories	500 000

Table 6. Void reactivity coefficient of BBWR

Gap between cavity box and channel box (cm)	Void reactivity ($\times 10^{-4}$ Δk/k/%void)
10.0	-1.61
8.0	-1.13

Table 7. Initial condition of fuel components

Nuclide	Initial weight fraction ¹	Weight fraction in load fuel
U	94.2%	Adjusted by enrichment ²
Pu	1.0%	Adjusted by enrichment ³
Minor actinide(MA)	0.13%	Entirely removed ⁴
Rare earth (RE)	1.3%	Dependent on DF/accompanied with Pu
Noble metal(NM)	1.6%	Dependent on DF/accompanied with U ⁵
Other FP	1.7%	Entirely removed

¹ Weight fraction in discharged LWR fuel.

² ²³⁵U: 0.2wt% (corresponds to depleted U).

³ Pu isotope composition: 5 year cooling/2 years after reprocessing

⁴ It is assumed that MA is not contaminated.

⁵ It is assumed that NM is accompanied with U when DF is the same as that of RE.

Figure 1. BBWR core profile

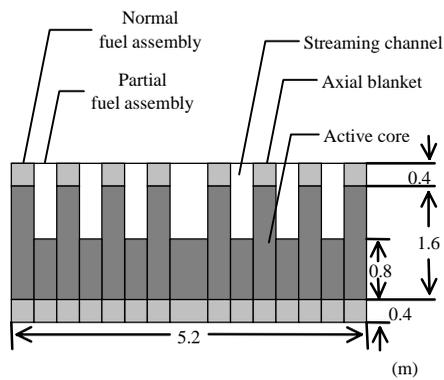


Figure 2. BBWR core layout

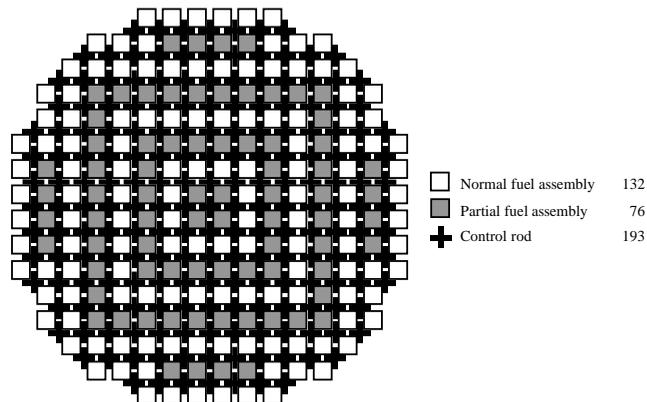


Figure 3. Profile of assemblies

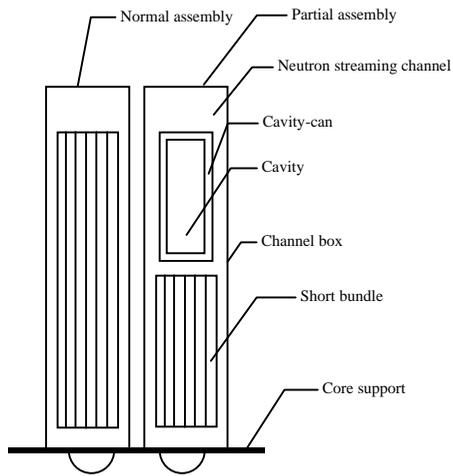
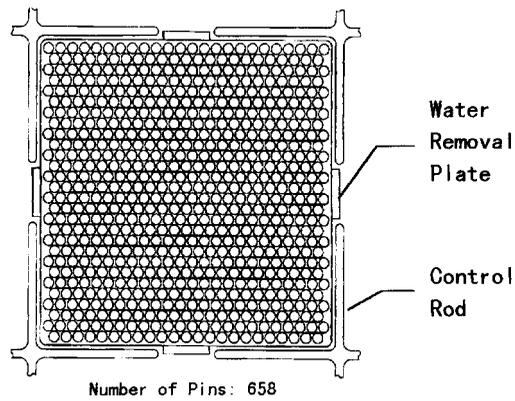


Figure 4. Cross-section view of BBWR assembly



Number of Pins: 658

Figure 5. Profile of control rod

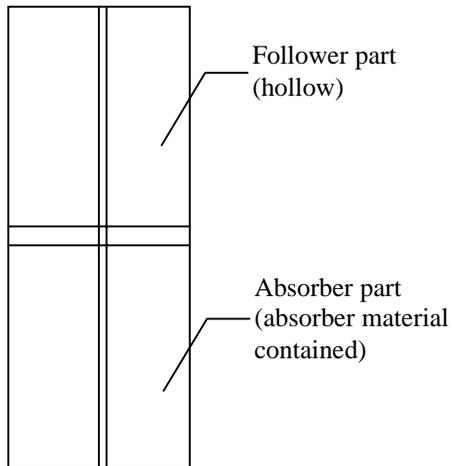


Figure 6. Neutron flux spectrum

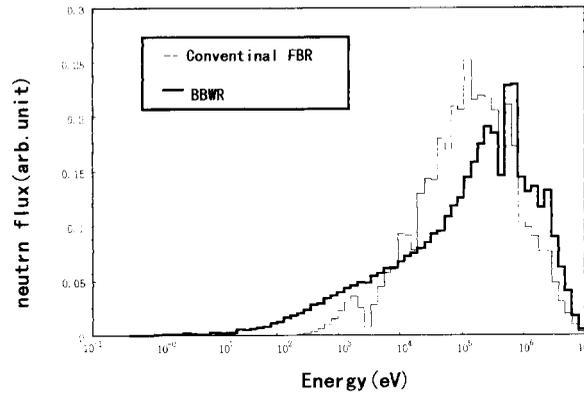


Figure 7. Core parameters and breeding ratio

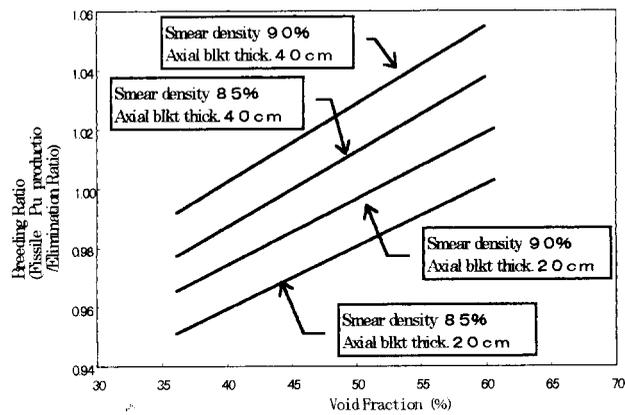


Figure 8. Cross-section view of streaming channel for Monte Carlo calculation

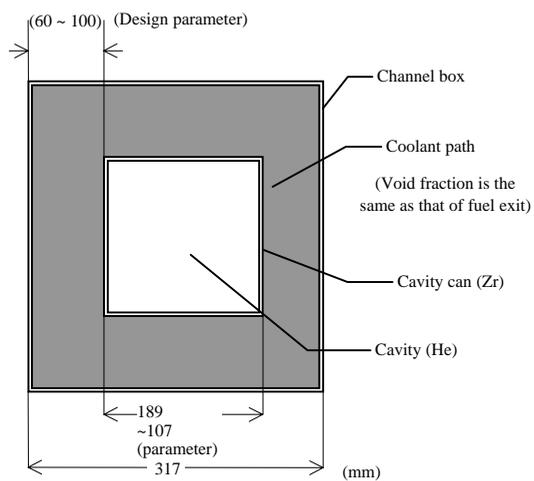


Figure 9. Pu enrichment in BARS

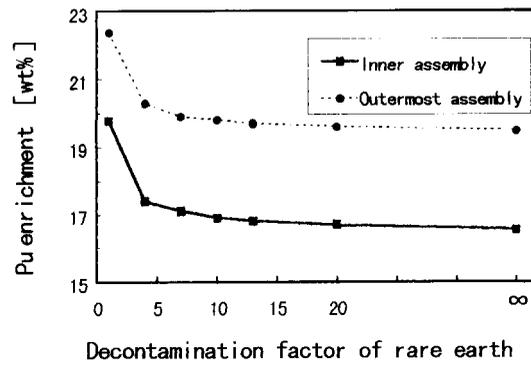
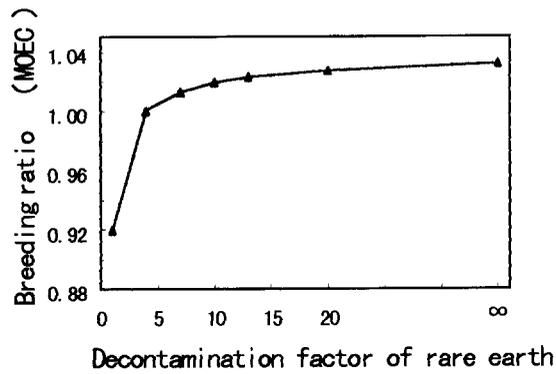


Figure 10. Breeding ratio in BARS



CONCEPTUAL CORE DESIGN OF A RESOURCE RENEWABLE BWR AND LONG-TERM ENERGY SUPPLY

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Abstract

An adaptation of the BWR has been conceived which demonstrates yet another aspect of the superiority of the BWR, this time as an LWR which has the potential to breed and to consume multi-recycled plutonium. The concept is referred to as the Resource Renewable Boiling Water Reactor (RBWR). The RBWR may be characterised as a BWR which operates with mixed (depleted uranium and plutonium) oxide fuel within the epithermal spectrum, and which has a breeding ratio of 1.0 and a negative void coefficient. The RBWR uses a short core (less than 1.5 m active core length). The shorter nuclear core should have a desirable effect upon nuclear island economics and capital equipment cost reductions. The RBWR is also effective in ensuring actinide transmutation and conducive to nuclear non-proliferation.

Introduction

Many people continue to consider oil as an inexhaustible resource, believing that the necessary quantities of it will remain available at low prices. This belief exists because of plentiful oil production and the return of oil prices in real terms to their level before the oil crises in recent years. Awareness of energy problems has worn off. In view of the basic trend toward ease in the supply of natural uranium in recent years, and the fact that there are still technical problems to be overcome before FBRs start commercial operation, LWRs will continue to be a viable alternative. In Japan, the tentative target for commissioning of a disposal site for high-level radioactive waste is the 2030s, 2045 at the latest. In a situation in which a new international order is being sought in the aftermath of the collapse of the Cold War structure, the existence and handling of plutonium, including its peaceful use, has become a major point of interest as regards concern about proliferation of nuclear weapons.

Uranium resources, however, can only operate current LWRs for about 100 years as shown in the following section. When development and utilisation of nuclear energy first began, there existed great expectations that it would not be an energy resource for only 100 years, but would become an alternative energy resource bequeathed to future generations from those who just happened to be living in an age of plenty, using an invaluable resource that it took hundreds of millions of years of the earth's history to produce.

An adaptation of the BWR has been conceived which demonstrates yet another aspect of the superiority of BWRs this time as an LWR which has the potential to breed and to consume multi-recycled plutonium. The concept is referred to as the Resource Renewable Boiling Water Reactor (RBWR) [1]. The RBWR may be characterised as a BWR which operates with mixed (depleted uranium and plutonium) oxide fuel within the epithermal spectrum, and which has a breeding ratio of 1.0 and a negative void coefficient. The RBWR uses a short core (less than 1.5 m active core length). This shorter nuclear core should influence nuclear island economics and capital equipment cost reductions. The RBWR should also be effective in ensuring actinide transmutation and be conducive to nuclear non-proliferation.

Scenario of long-term energy supply

A scenario for long-term energy supply is shown in Figure 1. Fifteen million tonnes are used as the quantity of natural uranium resources in a long-term assessment of LWR plants including reasonably assured, estimated assured and speculative resources. Three million tonnes of enriched uranium can be obtained from 15 million tonnes of natural uranium. About 30 tonnes of enriched uranium can operate a 1 GWe current LWR for about one year, and so three million tonnes of enriched uranium can operate one thousand (1 000) 1 GWe current LWRs for only one hundred years. Consequently about 15 million tonnes of depleted uranium and 15 000 tonnes of fissile plutonium will be left for us. If an RBWR having a breeding ratio of 1.0 and 10 tonnes of internal and external fissile plutonium inventory can be realised, fifteen hundred (1 500) 1 GWe RBWRs can be operated for 10^4 years. This provides for a scenario of long-term energy supply by the combination of current LWRs and RBWRs. The RBWR is thus a recycle-oriented BWR utilising all uranium resources with plutonium from LWRs as catalyst meaning that the total amount of fissile plutonium does not change during the life of the fuel bundles.

Change of plant power capacity and required natural uranium are shown in Figure 2. It is assumed for simplicity that the increasing rate of the total plant capacity world-wide will increase

from 1976 to 2050 by a rate of 20 GWe per year and a plant capacity of 1 500 GWe will be maintained after 2050. The effective operation for the past 20 years, considering the plant capacity factor, is close to the line drawn for the total plant capacity. The RBWR will be introduced in 2011 in this scenario. The number of current LWRs contributing to the total plant power capacity gradually decreases and all of the current LWRs will be replaced with RBWRs by the year 2200.

Required natural uranium is shown by the dashed line and amounts to 15 million tonnes by 2080 in the case of once-through operation in current LWRs. On the other hand, the total amount of required natural uranium to 2200 will be kept below 15 million tonnes by the combination of current LWRs and RBWRs, and fifteen hundred (1 500) 1 GWe RBWRs will continued to be operated using 15 000 tonnes of fissile plutonium and 15 million tonnes of depleted uranium for 10^4 years after 2200.

Design targets and concepts

Design targets and concepts are shown in Figure 3. The first design target is to realise long-term energy supply under the condition that power generation cost and safety are at ABWR levels. Other chief concerns, such as elimination of actinides disposal and non-proliferation, will be dealt with later. A hexagonal tight lattice fuel bundle instead of the current square lattice, coolant with high void fraction and Y-type control rods instead of cruciform control rods can reduce the effective water-to-fuel volume ratio to about 0.3. A mixed oxide fuel lattice, where plutonium is fed to depleted uranium with an effective water-to-fuel volume ratio of less than 0.3, can realise a breeding ratio of more than 1.0. Shortened tight lattice fuel bundles generating the same thermal power as that of an ABWR [2] can decrease plutonium inventory per unit power and the total number of power plants will be increased under limited plutonium. A breeding ratio of 1.0 and increasing the total plant capacity are essential to long-term energy supply. The same rated power, burn-up, diameter of reactor pressure vessel and cladding material as those of ABWR can keep maintaining plant construction cost at ABWR level for the RBWR. With regard to fuel cycle cost an increase in the manufacturing cost of mixed oxide fuel bundles will be cancelled by eliminating the natural uranium and enrichment costs. Enhancement of neutron leakage in the axial direction by the shortened core and the swing of the power distribution by axially zoned or axially heterogeneous fuel bundles make the void coefficient negative. Holding the core exit quality under 40% is essential to enclose radioactive corrosion products and radioactive materials generated from the fuel rod in the case of fuel failures in the pressure vessel. The distillation effect in the primary coolant system is an important merit of BWRs which cannot be expected from a steam-cooled breeder reactor.

Conceptual designs

The main design parameters of the RBWR in the equilibrium core and specifications of fuel bundle and core-averaged void distribution are compared with those of an ABWR in Table 1 and Figure 4, where RBWR-HO is the homogeneous core using axially two zoned fuel bundles minimising plutonium inventory, RBWR-B2 is the high burn-up core with average exposure of discharged fuel bundle in the equilibrium core of 70 GWd/t, and RBWR-AC is the core recycling minor actinides with plutonium.

These nuclear cores have 1 350 MWe rated electric power, 7.2 MPa core pressure and 2.88 m core radius. The 223 Y-type control rods with three wings are inserted into gaps among the 720 hexagonal fuel bundles to constitute the nuclear core. Followers consisting of zircaloy are attached to

control rods to prevent water from going into spaces after rods are withdrawn. The triangle fuel rod lattice is effective in decreasing the ratio of water to fuel while keeping cooling ability. The core height of the RBWR-HO is 52 cm to which the axial blankets, 25 cm for the upper one and 20 cm for the lower one, are attached. The core coolant flow is 2.6×10^4 tonnes/h with subcooling of 10°C at the entrance and 34 w/o steam quality at the core exit which keeps the distillation function, while confining radioactive corrosion products to the pressure vessel, even in a transient process.

Lattice pitches of fuel bundles are 199.3 mm for the side with the control rod and 194.4 mm for the side without one. The channel box of the fuel bundle is hexagonal with 193.6 mm length for facing sides and 2 mm thickness. Control rod wings are 7.5 mm thick and the gap between a wing and the channel box of a fuel bundle is 1.6 mm. Fuel bundles for the side without control rods make contact through pads on the channel box, and there is the space 0.8 mm wide between channel boxes. The 271 fuel rods are arranged in a triangular lattice of 11.38 mm pitch and 1.3 mm space to constitute the fuel bundle which has fuel rods fixed with four ring-type spacers keeping 1.05 mm gap between the outermost fuel rod and the channel box. Fuel rods for the equilibrium core of the RBWR-HO contain a mixture of PuO_2 and UO_2 and have a 8.63 mm outside diameter. Bundle averaged fissile plutonium enrichment of 12 w/o in the upper part of the bundle and 10 w/o in the lower part is required. In the RBWR-HO an inventory of 4.4 tonnes per 1 GWe of fissile plutonium is required.

A breeding ratio of 1.01 is achievable with a exposure of 45 GWd/t. This ratio is defined as the number of atoms of fissile plutonium left in discharged fuel bundles per fissile plutonium in initial fuel bundles. The internal breeding ratio – the number of fissile plutonium atoms left in the core part of discharged fuel bundles per atoms in initial fuel bundles – is 0.90. This large internal breeding ratio results in a small net percentage fissionable material burn-up in the core and, therefore, a small reactivity swing between refuellings. The loading pattern of fuel bundles in the equilibrium core as shown in Figure 5 adopts the zone loading and periodic boundary condition of 120 degrees in the azimuthal direction due to flattening of the radial power peaking to 1.20. The core coolant flows in the lower blanket with 10°C subcooling and gives a void fraction of about 30% at the bottom of the core due to heating in the lower blanket and that of about 80% at the top of the core. Axial power peaking factor including blankets of 1.72, maximum linear heat generation rate of 13.3 kW/ft and MCPR of 1.32 in hailing power can be achieved.

In an operating BWR, about 85% of the fission reactions occur in the thermal energy region, whereas the medium fission energy is about 9 keV in the RBWR-HO, and the reaction rates in the resonance region are comparatively large. Therefore, while a conventional BWR has a Doppler coefficient of $-1.6 \times 10^{-5} \Delta\text{k/k}/^\circ\text{C}$, the value in the RBWR-HO is $-3.7 \times 10^{-5} \Delta\text{k/k}/^\circ\text{C}$, or about two times as large. The RBWR-HO has a void coefficient of $-0.5 \times 10^{-4} \Delta\text{k/k}/\%\text{void}$ compared with $-7.0 \times 10^{-4} \Delta\text{k/k}/\%\text{void}$ for the conventional BWR and so the thermal margin is increased relatively, for example, in the event of pressure elevation or temperature lowering of coolants.

Calculation methods

One of the remarkable features of the RBWR is that the effective water-to-fuel volume ratio is smaller than that of current BWRs, so that the calculation method for microscopic epithermal cross-sections and the nuclear data library for resonance energy range are important in nuclear characteristic evaluation. For precise numerical evaluation of fuel bundle designs, the VMONT [3], in which a vectorised Monte Carlo neutron transport method is coupled with the burn-up calculation,

has been developed. The C/E values for k-infinity and reaction rate ratio of ^{238}U capture to ^{239}Pu fission are shown in Figure 6. The applicability of the VMONT code to tight lattice configurations was confirmed using PROTEUS experimental data [4,5].

For thermal hydraulic analysis, the critical power correlation for the tight lattice configuration gains increasingly in importance. So, the modified CISE critical power correlation [6], which is based on the critical quality-boiling length correlation developed by CISE [7], has been developed for the application range. Figure 7 compares experimental data and calculation results, in which the former data were taken from an experiment on the critical power of the closely packed lattice fuel bundles at Bettis Atomic Power Lab [8]. The proposed correlation could reproduce the experimental data with a standard deviation error of 8%. Transient behaviours of a BWR involve a complex interplay between the inherent system thermal-hydraulics and neutronics and the active components. The transient behaviours were evaluated by a lumped-region model for in-reactor component thermal-hydraulics coupled with a point reactor neutron kinetics model, followed by one-dimensional single channel analysis to estimate flow transient in a fuel bundle. More precise analyses by a three-dimensional nodal kinetic code are now being planned. The modified CISE critical power correlation was used for thermal margin calculations.

Transient and safety analyses

The proposed designs of the RBWR have a negative void coefficient. The typical transients were analysed and the ΔMCPR , that is, the decrease in thermal margin during transients was evaluated. In the evaluation, the characteristics of the active components in the RBWR were assumed to be those of the ABWR. For example, internal pumps are utilised as recirculation pumps instead of jet pumps in current BWRs. Figure 8 shows the results of transient analyses. The maximal ΔMCPR of the RBWR-HO was 0.10, nearly the same as for current BWRs. The fuel integrity can be maintained because the MCPR was more than 1.3 in the rated power normal operation. The maximum heat flux and the maximum vessel pressure during transients were confirmed as maintained at the limiting values for current BWRs. From a safety perspective, a high pressure flooder break accident, which seems to be the most severe loss of coolant accident (LOCA) for the RBWR, was also analysed. Figure 9 shows that the peak cladding temperature in LOCA was 672°C for the RBWR-HO, less than the limiting value ($1\ 200^\circ\text{C}$) of current BWRs.

Actinide transmutation

The design concept of RBWRs should play an important role from the viewpoint of storage of long-life actinide elements. In the second phase of the RBWR development, other actinide elements as well as plutonium are recycled together taking one step forward from only plutonium recycling in the first phase. Figure 10 shows that accumulation of ^{237}Np (half-life is about 2×10^6 years) reaches equilibrium at the amount of 40 kg per 1 350 MWe RBWR-AC. This result means that the equilibrium state of ^{237}Np between production rates and consumption rates is kept in the RBWR-AC under the condition that 40 kg of ^{237}Np generated in current BWRs is fed to the plutonium-enriched depleted uranium fuel bundles in the initial core of the 1350 MWe RBWR-AC, and a new recycling system can be realised with LWR technology, in which plutonium and other actinides are confined to only nuclear reactors and reprocessing plants.

International nuclear fuel centres and nuclear non-proliferation efforts

The quantity of fissile plutonium in a spent fuel bundle of the RBWR with a breeding ratio of 1.0 is nearly equal to that in a fresh bundle. By establishment of several international nuclear fuel centres comprised of a fabrication plant and a reprocessing plant, it becomes possible to avoid domestic and international transfers of plutonium except for MOX fuel bundles between a centre and the RBWRs. Furthermore, the RBWR can be operated with more than 35% ²⁴⁰Pu content under the equilibrium condition of plutonium multi-recycling as shown in Figure 11. Consequently the RBWR is expected to be conducive to nuclear non-proliferation.

Conclusion

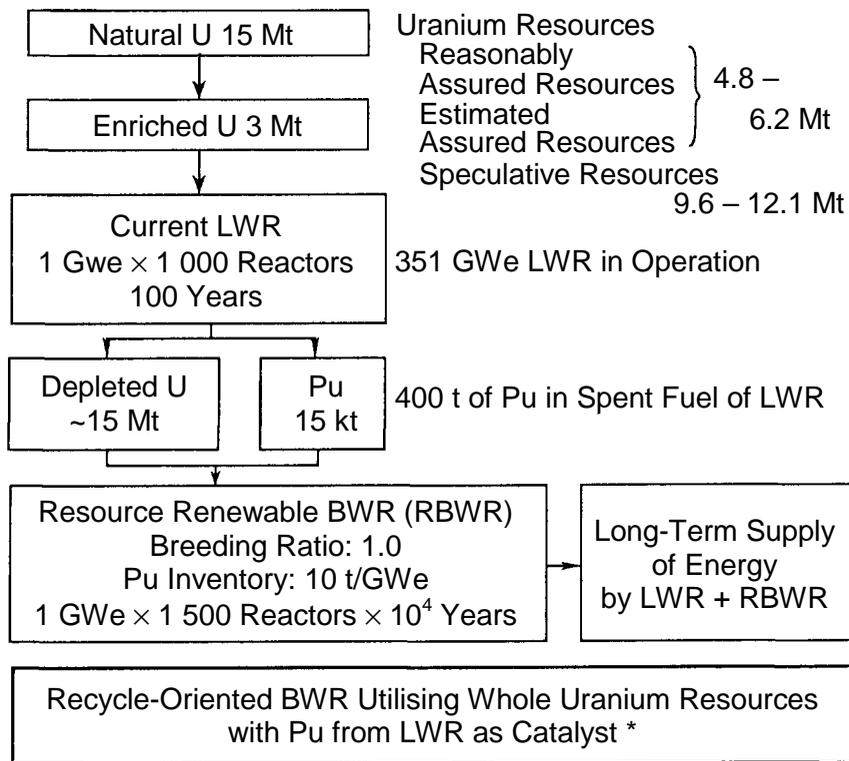
The conceptual designs of the 1 350 MWe RBWR-HO, and RBWR-B2 were proposed, which secure long-term supply of energy at ABWR levels of power generation cost and safety. The RBWR, attaining a breeding ratio of 1.0 and a negative void coefficient, is realised by the combination of a hexagonal tight lattice fuel bundle, a core-averaged void fraction of 65%, Y-type control rods, a reduction in core height, swing of the power distribution by axially zoned or axially heterogeneous fuel bundle, and so on. Fifteen hundred (1 500) 1 GWe RBWRs can be operated for 10⁴ years using 150 000 tonnes of fissile plutonium and 15 million tonnes of depleted uranium, which would remain as a result of operating one thousand (1 000) 1 GWe current LWRs for 100 years. Since the steam quality at the exit of the core is kept to 40% or less, the materials to be used are the same as those of BWRs now under operation and a distillation function by boiling can be maintained to confine radioactive materials within a pressure vessel. The RBWR-AC is designed for recycling actinides together with plutonium in order not to leave long-lived radioactive waste for future generations. Furthermore, a combination of the RBWRs and international fuel centres integrating manufacturing and reprocessing is conducive to nuclear non-proliferation.

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Figure 1. Scenario of long-term energy supply



* Constant Fissile Pu in Period of Burn-up

Figure 2. Change of plant power capacity and required natural uranium

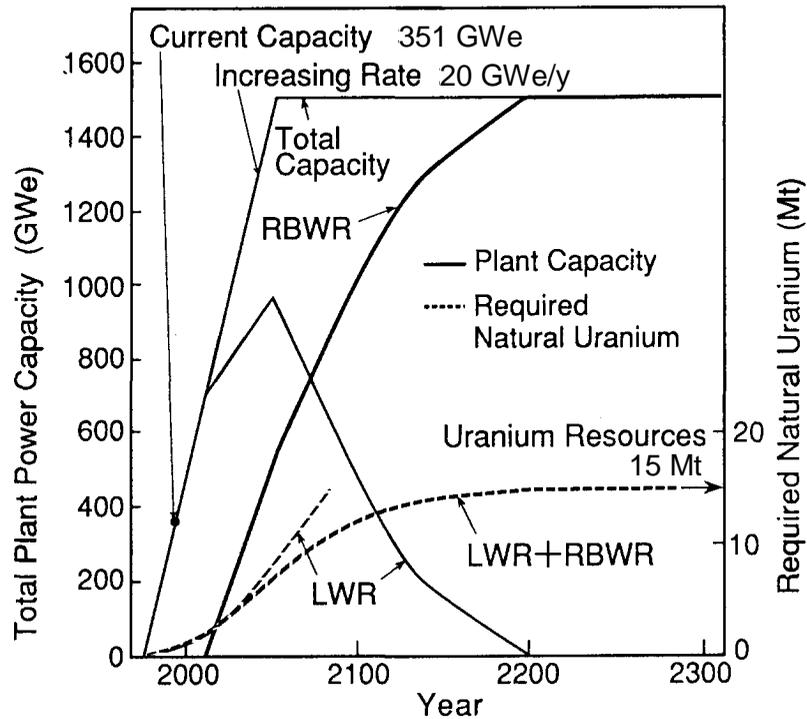


Figure 3. Design targets and concepts of RBWR

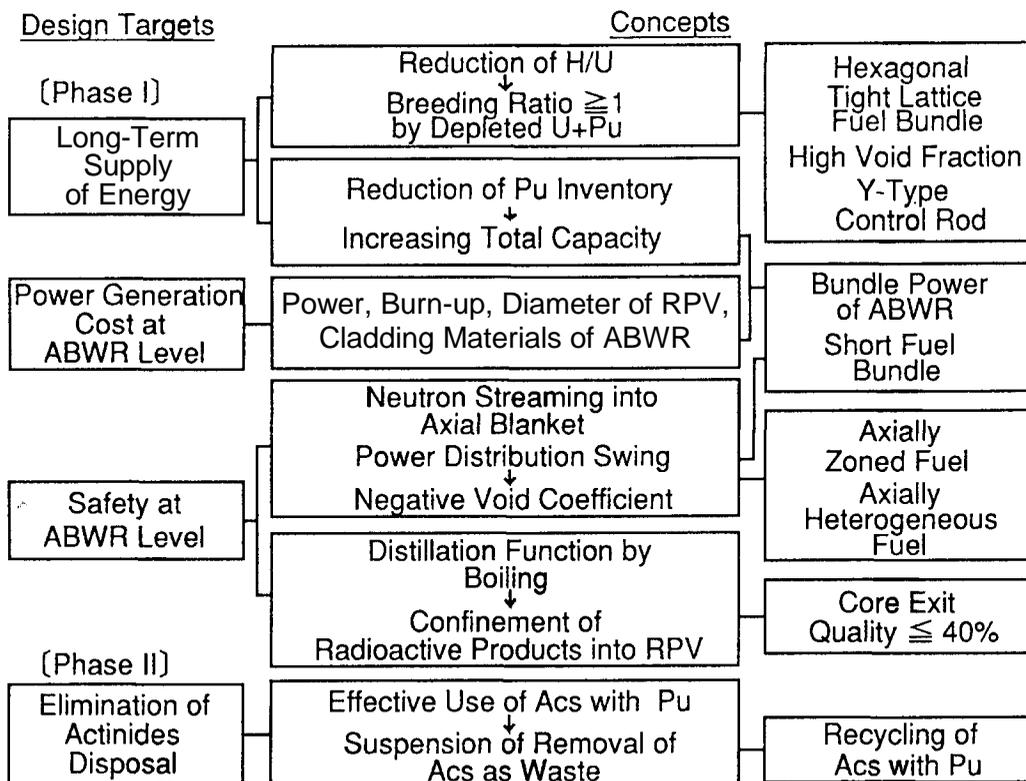


Figure 4. Specifications of fuel bundle and core-averaged void distribution

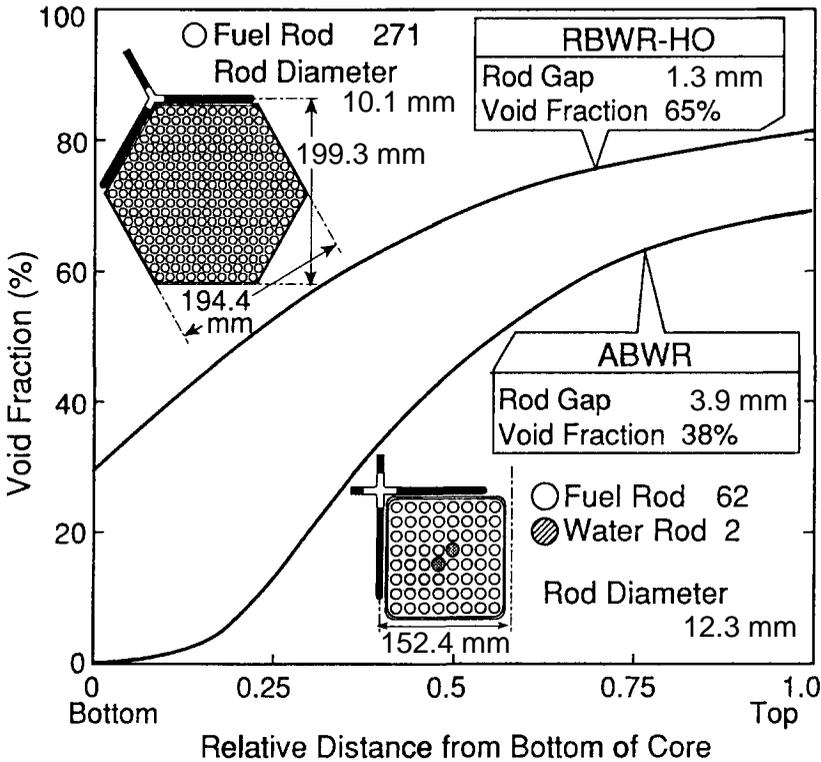


Figure 5. Fuel loading pattern

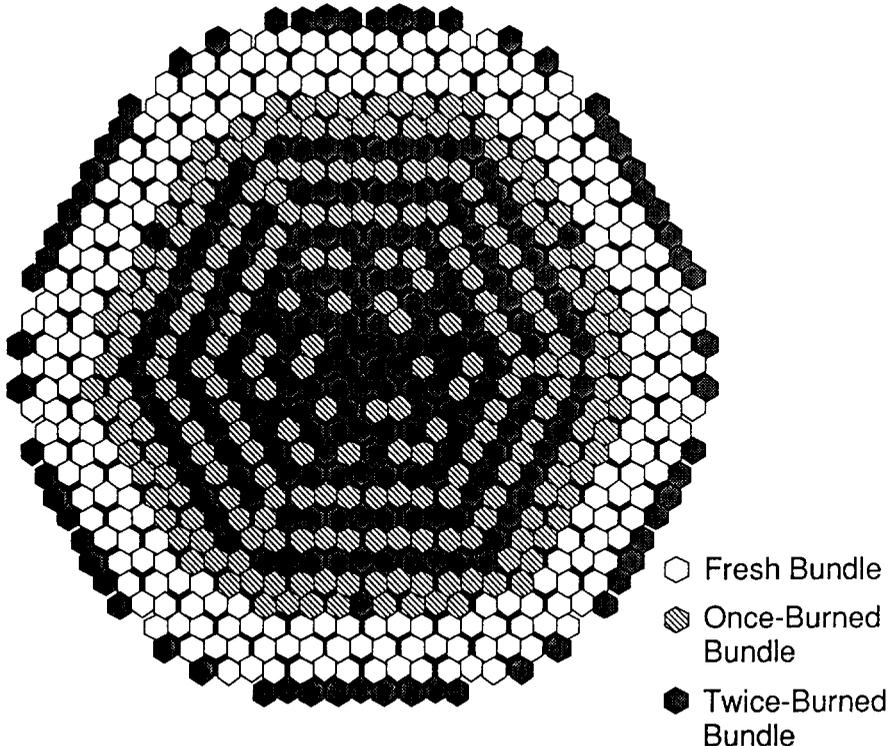


Figure 6. Comparisons of experimental and calculated k-infinity and reaction rate ratios

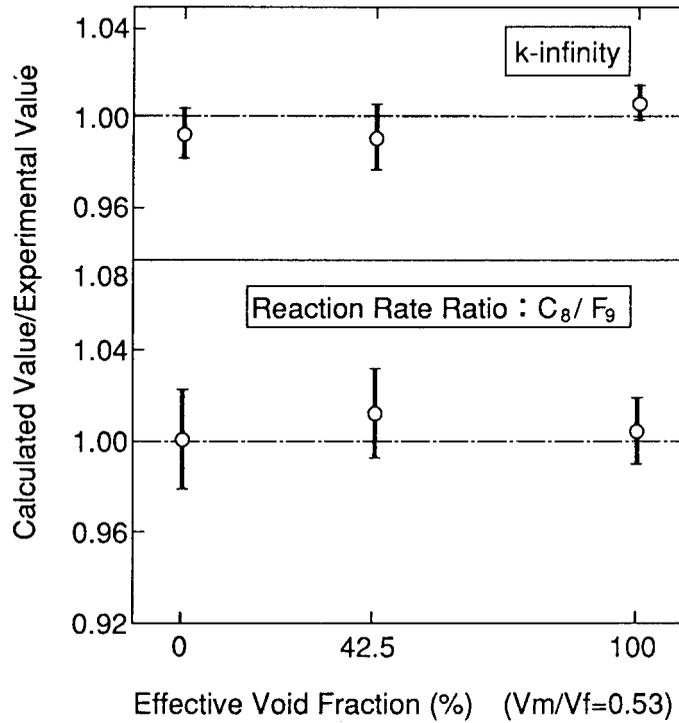


Figure 7. Comparison of calculated and experimental critical powers

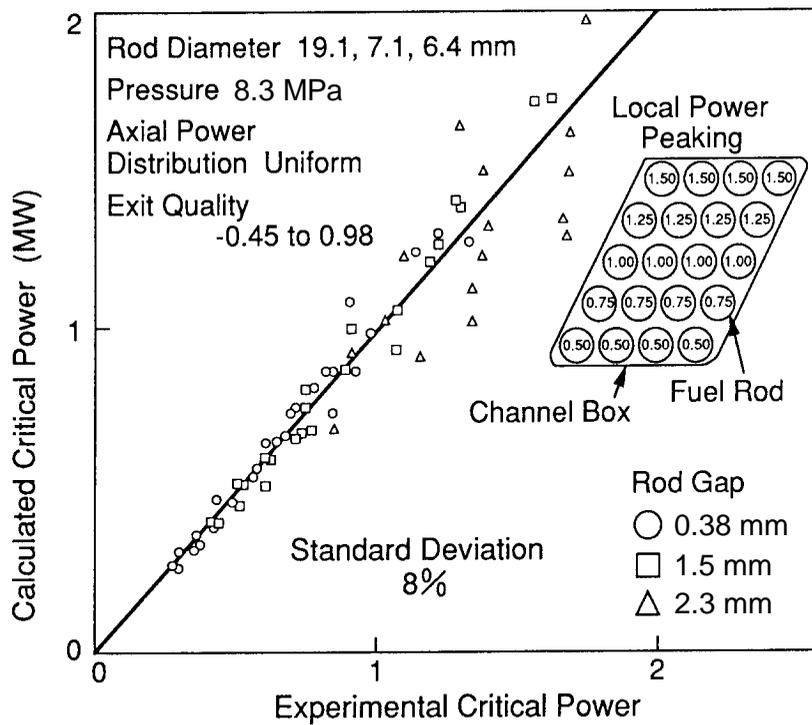


Figure 8. Thermal margin during typical abnormal transients

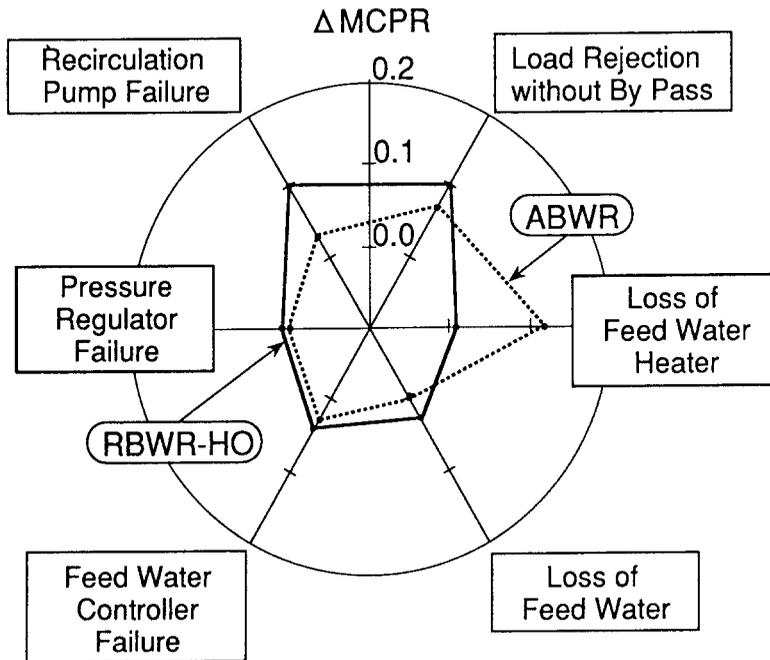


Figure 9. Peak cladding surface temperature for loss of coolant accident

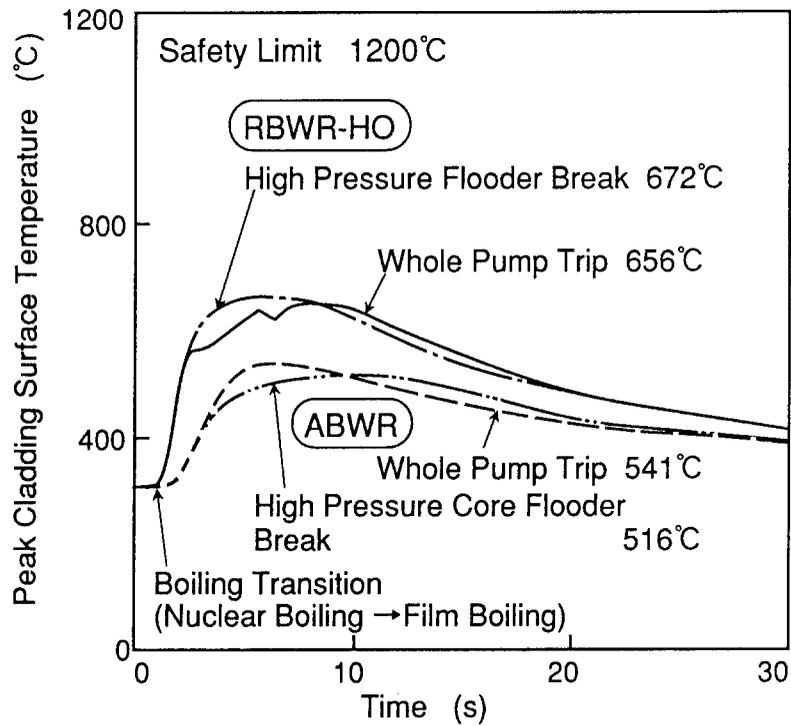


Figure 10. Accumulation of ^{237}Np

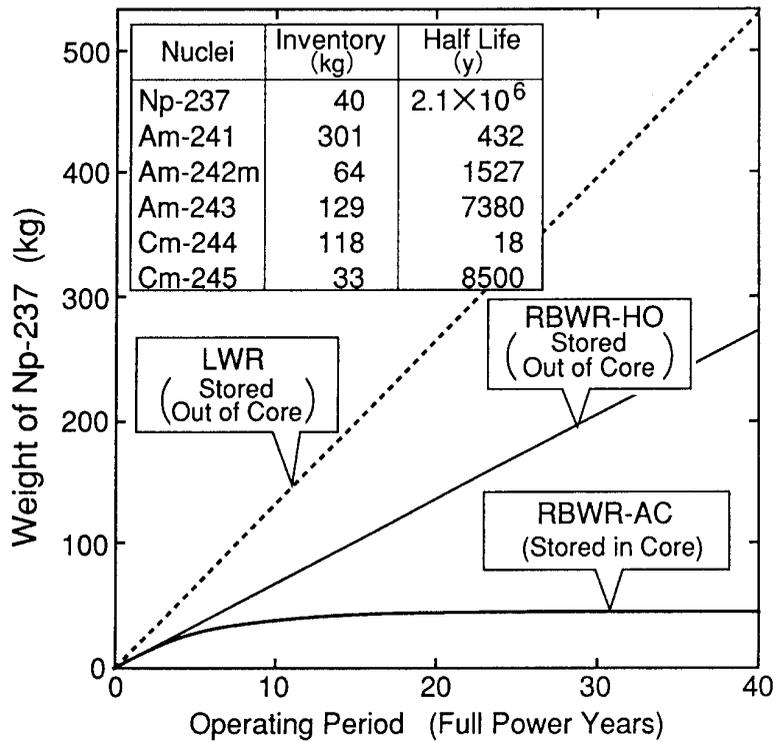


Figure 11. Non-proliferation efforts and Pu isotope content

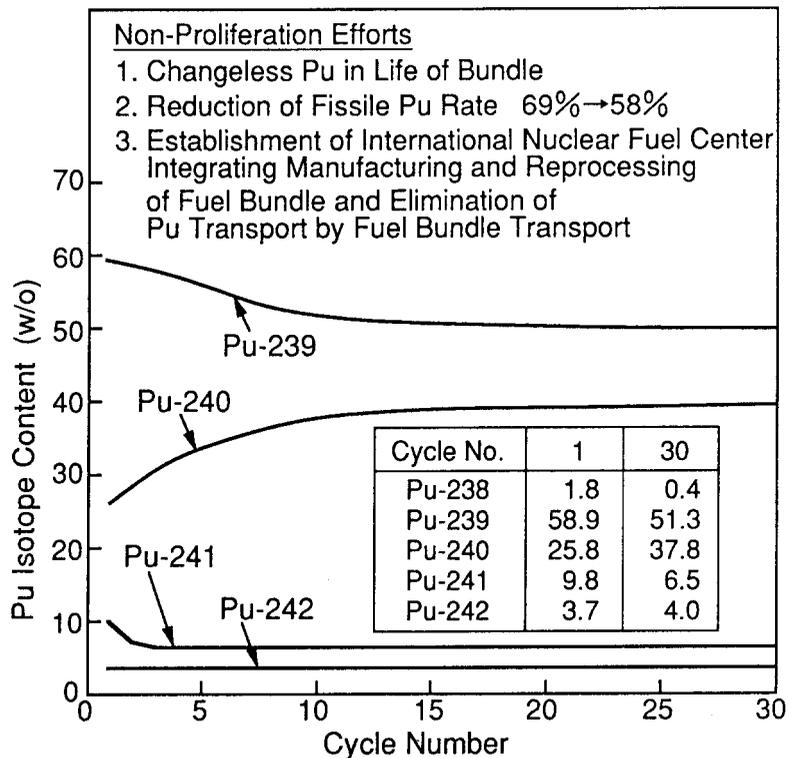


Table 1. Specifications and core performance

Item		RBWR-HO	RBWR-B2	RBWR-AC	ABWR																						
Electric Power	MWe	1356	1356	1356	1356																						
Dome Pressure	MPa	7.2	7.2	7.2	7.2																						
Core Outer Radius	m	2.88	2.88	2.88	2.69																						
Number of Fuel Bundles		720	720	720	872																						
Burn-up	GWd/t	45	70	45	38																						
Core Active Height ¹	m	0.52	0.70	0.68	3.71																						
Coolant Flow Rate	10 ⁴ t/h	2.6	3.2	2.9	5.2																						
Core Exit Quality	%	34	28	31	14.5																						
Average Void Fraction	%	65	59	64	38																						
Core Pressure Drop	MPa	0.15	0.16	0.19	0.17																						
Fissile Pu Enrichment	%	10.8	10.7	10.3	3.6																						
Fissile Pu Inventory ²	t	4.4	5.8	5.5	...																						
Breeding Ratio		1.01	1.01	1.01	...																						
Max. Linear Heat Generation Rate ³	kW/ft	13.3	13.1	13.3	12.5																						
MCPR ³		1.32	1.40	1.32	1.30																						
Void Coefficient	10 ⁻⁴ Δ $\frac{k/k}{\%void}$	-0.5	-0.5	-0.5	-7.0																						
Axial Pu Distribution	<table border="1" style="display: inline-table; vertical-align: middle;"> <tr> <td>w/o</td> <td>cm</td> </tr> <tr> <td></td> <td>cm</td> </tr> <tr> <td>w/o</td> <td>cm</td> </tr> </table>	w/o	cm		cm	w/o	cm	<table border="1" style="display: inline-table; vertical-align: middle;"> <tr> <td>12</td> <td>17</td> </tr> <tr> <td>10</td> <td>35</td> </tr> </table>	12	17	10	35	<table border="1" style="display: inline-table; vertical-align: middle;"> <tr> <td>18</td> <td>22</td> </tr> <tr> <td>DU</td> <td>29</td> </tr> <tr> <td>18</td> <td>19</td> </tr> </table>	18	22	DU	29	18	19	<table border="1" style="display: inline-table; vertical-align: middle;"> <tr> <td>13.5</td> <td>27</td> </tr> <tr> <td>DU</td> <td>18</td> </tr> <tr> <td>13.5</td> <td>23</td> </tr> </table>	13.5	27	DU	18	13.5	23	
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13.5	23																										

1: Upper and Lower Blanket 25, 20 cm 2: For 1 GWe 3: Hailing
 DU: Depleted Uranium

PROCESS DEVELOPMENT FOR DUPIC FUEL FABRICATION

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Abstract

Direct use of spent PWR fuel in CANDU reactors (DUPIC) fuel cycle technology is currently under development at KAERI in co-operation with Canada, USA and the newly joined IAEA. The DUPIC concept, based on the idea that the spent PWR fuel can be directly refabricated into a reusable fuel for CANDU reactors, has a lot of benefits in terms of proliferation resistance, savings of uranium resources and reduction of spent fuel arising from PWR and CANDU, etc. KAERI has developed DUPIC processes and equipment suitable for a hot cell environment. The main activities in IMEF hot cell are almost ready for fabrication of DUPIC fuel starting in early 1999. Prior to the main campaign, powder characterisation and sintering behaviour on a small scale will be carried out at the end of 1998. In this paper, the status of the DUPIC research programme at KAERI is described.

Introduction

DUPIC fuel cycle technology has been under development at the Korea Atomic Energy Research Institute (KAERI) since the early 1990s in co-operation with Canada, the USA, and more recently, the newly joined IAEA [1,2]. Before the experimental verification of DUPIC fuel technology started, a feasibility study on DUPIC fuel was performed. The results of the feasibility study on the DUPIC fuel cycle were that the oxidation and reduction of oxide fuel (OREOX) process was selected as the most promising fabrication option, and that the safeguarding of the DUPIC process was possible.

Under the international co-operation framework for the experimental verification of DUPIC fuel, KAERI has been developing the processes and equipment to fabricate DUPIC fuel, while Atomic Energy Canada Limited (AECL) has developed the processes to fabricate several DUPIC fuel elements. KAERI is also performing a compatibility analysis of DUPIC fuel with CANDU reactors jointly with Canada. For the safeguards technology development, KAERI has been developing the safeguards system and non-destructive assay method with the USA, in which IAEA shows much interest.

The overall fabrication process can be divided into decladding of spent PWR fuel rods, OREOX process, pelletisation, rod fabrication and bundle assembling. Off-gas treatment and fuel inspection processes should also be included. Due to the highly radioactive characteristics of DUPIC processes, each piece of equipment should be designed and maintained in a remote manner. KAERI has completed the development and cold tests of the equipment, and plans to begin the fabrication of DUPIC fuel elements in early 1999.

In order for DUPIC fuel to be qualified ultimately as a licensable fuel, its irradiation behaviour should be analysed and established. Overall irradiation plans for DUPIC fuel, including pellet irradiation, are scheduled at the HANARO research reactor for the end of 1999. Through a series of irradiation and PIE examination, the feasibility and the performance of the DUPIC fuel will be verified experimentally.

Characteristics of DUPIC fuel cycle

Over the years nuclear energy has become more important, especially for countries that have little energy resources and small area. In contrast to its importance, the nuclear field has several key issues to be considered, such as disposal of spent fuel, nuclear weapons proliferation, etc.

The basic concept of the DUPIC fuel cycle comes from the idea that in the spent PWR fuel the fissile remnants are enough to burn again in CANDU reactors which use natural uranium. All spent PWR fuel can be reused in CANDU reactors in the case where the ratio of PWR and CANDU reactors is adequate. Even though the basic idea is simple, there are many questions that remain to be addressed. Reactor compatibility, remote manufacturing technology, safeguarding systems and performance evaluation are among the key issues.

In the early stage of the DUPIC programme, seven options of fabrication methods were profoundly analysed. Five options are simple reconfiguration methods without decladding, and two options are OREOX and the vibrational packing method. Though reconfiguration methods allow easy fabrication of DUPIC fuel, they showed a lot of difficulties with regard to reactor physics and fuel performance. As a result of the feasibility study, OREOX was selected as the most promising option. OREOX has extreme merits in the areas of reactor compatibility and performance, but is more difficult to fabricate.

The DUPIC process has strong proliferation resistance because it does not separate radioactive fission products as well as plutonium from spent PWR fuel. Plutonium bearing fuel is very attractive from the viewpoint of peaceful consumption of plutonium. Combining the merits of proliferation resistance and reuse of spent PWR fuel makes the DUPIC fuel cycle one of the most attractive fuel cycles for the near future.

In order to manufacture DUPIC fuel, the spent PWR fuel assembly is disassembled and decladded to recover the spent fuel materials. The fuel fabrication processes are shown schematically in Figure 1. The recovered spent fuel materials are treated by repeated oxidation and reduction processes to produce the microcracks in the powder so as to increase the powder reactivity and are subject to additional powder milling to make the powder finer for the purpose of increasing powder sinterability. Once the resintered powder feedstock is prepared, the DUPIC fuel fabrication process is almost similar to the conventional CANDU fuel fabrication process, except that all fabrication processes should be performed in hot cells due to their intrinsic radioactivity. Although volatile and some semi-volatile fission products will be removed during the fuel fabrication processes of oxidation, reduction and sintering, the bulk of the radioactive stable solid fission products will remain in the DUPIC fuel. This causes all the fabrication processes to be highly radioactive, thereby requiring heavy shielding throughout the operation. Although the maturity of fuel fabrication technology for conventional CANDU fuel has been well established, the remote fabrication and quality control technology required for DUPIC fuel fabrication requires further development.

The spent PWR fuel with a discharge burn-up of about 35 000 MWd/T is composed of about 0.9 wt.% of fissile uranium, 0.6 wt.% of fissile plutonium and about 3 wt.% of fission products. Even though the retained fission products in DUPIC fuel act as absorber materials, the burn-up of DUPIC fuel could be more than 15 000 MWd/T, about twice that of conventional CANDU fuel. However, DUPIC fuel has distinct features with regard to conventional CANDU fuel, such as (1) high fissile content; (2) high plutonium content; (3) fuel composition variation due to initial enrichment and burn-up history of spent PWR fuel; and (4) radioactivity due to residual fission products.

One of the advantages of the DUPIC fuel cycle is the waste reduction of the whole fuel cycle. The fabrication process wastes are mainly metallic forms of the PWR assembly skeleton, the cladding hulls and the trapped volatile and semi-volatile fission products, which are much smaller in volume compared to corresponding wet reprocessing process due to the characteristics of the dry process. The spent fuel of about three PWR reactors can be fabricated to supply DUPIC fuel for one CANDU reactor of equivalent reactor capacity, even though the optimum reactor ratio would depend on the discharge burn-up of PWR fuel. This concept is displayed in Figure 2. Considering the doubling of the burn-up of DUPIC fuel in CANDU reactors, the overall radioactive waste arising can be reduced by about 35% in comparison with the once-through fuel cycle. And the amount of uranium resources needed for CANDU reactors can be saved and replaced by the spent PWR fuels. The fact that Korea has both PWRs and CANDUs indicates a strong motivation to develop DUPIC fuel. Also, we believe that all PWRs and CANDUs can be connected not only in Korea, but also in the world.

International co-operation

In general, research using spent nuclear fuel receives focal attention from the safeguards and proliferation viewpoints, regardless of its objectives and the research processes. For the DUPIC fuel development programme, the safeguardability and transparency of the research were the primary considerations from the beginning. Therefore, the DUPIC programme has been developed jointly by KAERI, AECL and the USA so as to strengthen both the technical capability and transparency of the

research. While the fuel process technology, reactor physics and safety analysis are mainly being developed by the AECL and KAERI, the safeguards technology is being jointly developed by KAERI and the USA. Moreover, the IAEA became the fourth partner in the DUPIC programme to bolster safeguards matters. Thus, the DUPIC research programme is a desired model for international co-operation for the promotion of technological improvement and concurrently maintaining the transparency of the research.

While KAERI has developed the fuel bundle manufacturing technology, AECL has succeeded in fabricating several DUPIC fuel elements at the Whiteshell Laboratory and is going to irradiate the elements at NRU soon.

When DUPIC fuel cycle technology is shown to be safe and reliable in performance and economically competitive, it will become an alternative fuel cycle for the future and will enhance prospective co-operation scenarios between adjacent LWR and CANDU countries.

DUPIC fuel development at KAERI

KAERI has been preparing the required fabrication and test facilities for its main fabrication campaign starting in early 1999. The existing research facilities including the hot cell facilities of the Post-Irradiation Examination Facility (PIEF) and the Irradiated Material Examination Facility (IMEF), the HANARO research reactor, and other fuel laboratories will be utilised with minimum modification for the fabrication of prototypical fuel. From the middle of 1999, several DUPIC fuel pellets and elements will be fabricated, and a series of DUPIC fuel irradiation tests will begin at the HANARO research reactor to compare the behaviour of DUPIC fuel pellets with conventional uranium oxide pellets. Following the pellet irradiation, element and bundle irradiation tests will be scheduled to gather more information on the behaviour of DUPIC fuel in reactor conditions.

DUPIC powder characterisation and sintering behaviour study

Before manufacturing DUPIC fuel, KAERI has planned to study powder characteristics and sintering behaviour of spent PWR fuel on a small scale at the end of 1998. Its results will be used for controlling process parameters in manufacturing DUPIC fuel in 1999. Decladding equipment, an OREOX furnace, a tube type sintering furnace and a hand press were installed in the PIEF hot cell. Doors and inserting equipment of both furnaces are modified to operate easily with remote manipulators. The press was designed as compactly and simply as possible. A simple off-gas treatment system was also installed to prove the efficiency of the process developed by KAERI.

DUPIC fuel manufacturing plan

The IMEF hot cell that is planned to fabricate DUPIC fuel was constructed as an inspection hot cell of irradiated materials. Therefore, some modification for fabrication of DUPIC fuel was carried out. Roof service room and rear doors were amended, and some utility lines and penetration lines for electricity and signal lines were modified.

The DUPIC fuel manufacturing process was developed by KAERI using simulated DUPIC fuel. An overall flowsheet of the DUPIC process is shown in Figure 1. The main processes are decladding, OREOX, powder treatment, sintering, grinding, welding and inspection. KAERI plans to start the fabrication of DUPIC fuel in early 1999. Over 20 items of manufacturing equipment were designed in

a remote manner and are now under cold testing. The decladding process and equipment with twin rolls were developed by KAERI. In the OREOX furnace it was decided to use a commercially available furnace from LINDBURG. The doors and inserting equipment of the OREOX furnace were modified to be suitable for hot cell work. The sintering furnace was specially designed with remote manner and was installed in the IMEF hot cell and tested up to 2 000°C. Grinding equipment was designed with remote and dry manners. Other process equipment was manufactured and made ready for fabricating DUPIC fuel. The off-gas treatment system will be installed after its functionality has been proved during the PIEF study. Detailed inspection of by-products will be planned during the verification stage. After all processes are qualified using spent fuel feedstock, a quality control system will be developed systematically.

Reactor physics

The reference DUPIC fuel composition was selected as that of the spent fuel with a discharge burn-up of 35 000 MWd/T and 3.5% initial enrichment. Compatibility with existing CANDU reactors includes reactor physics, fuel handling and radiation protection. A study on the reactor physics, such as power distribution, reactivity perturbation effect, reactor control system, regional overpower protection, CHF and CCP margins was carried out [3]. Works for commercialisation will be done in the near future.

Fuel performance

DUPIC fuel has very distinguished characteristics compared to the other uranium dioxide fuel. The main difference is that the DUPIC fuel contains fission products and plutonium. This affects the physical and chemical properties of the fuel as well as its nuclear physics. The most important properties are thermal conductivity and fission gas release of DUPIC fuel. To reveal the properties and to gather important information on the irradiation behaviour of DUPIC fuel, DUPIC pellets will be irradiated at the HANARO research reactor. DUPIC elements and bundles will then be irradiated so as to demonstrate the performance of the fuel.

Safeguards system

Recently, the issue of safeguards has become an important factor to be considered when selecting a fuel cycle option. The DUPIC fuel cycle is regarded as excellent in terms of safeguards because there is no separation of fission products and plutonium during its processes. Also, it is difficult for the diversion due to its intrinsic radioactivity and heavy shielding.

KAERI has developed a new neutron counter (DUPIC Safeguards Neutron Counter: DSNC) with the USA for measuring sensitive materials. Its function was successfully demonstrated at KAERI in 1998. The idea and the utility of this neutron counter are excellent to the point that the IAEA has expressed interest in this project. KAERI has also been developing a near real-time safeguards system for the DUPIC process using a neutron monitor.

Conclusions

1. DUPIC fuel cycle technology has the multiple advantages of reducing the accumulation of spent fuel, increasing the efficiency of uranium resource utilisation and of possessing proliferation resistant characteristics.

2. KAERI is performing a co-ordinated research programme to experimentally verify the performance of DUPIC fuel in co-operation with the AECL and the USA. The DUPIC research programme is a typical example of international research programmes for nuclear fuel cycles which efficiently harmonise the expertise of the participating parties while maintaining the transparency and safeguardability of the research project.
3. KAERI will fabricate DUPIC fuel in early 1999 and has plans to irradiate DUPIC fuel to verify the DUPIC fuel performance.

Acknowledgements

KAERI's DUPIC programme is sponsored by the Korean government's nuclear R&D plan.

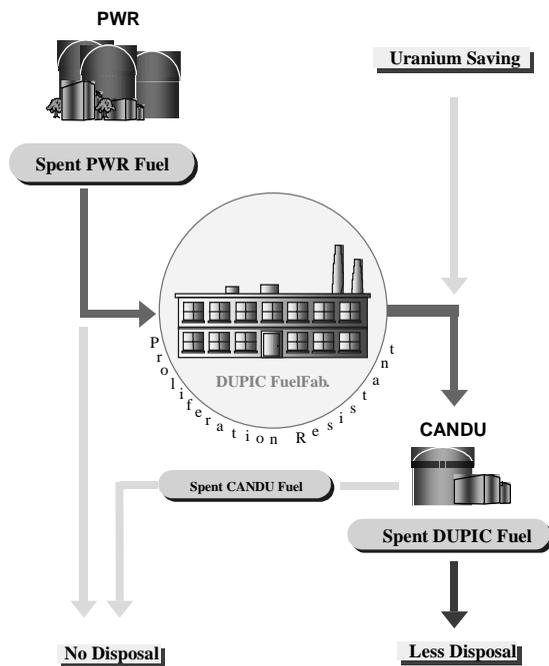
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Figure 1. DUPIC fuel fabrication processes



Figure 2. DUPIC fuel cycle concept



THE LR-0 REACTOR POSSIBILITIES FOR MOX TYPE FUEL PINS RESEARCH

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Abstract

In the first part a short description of the LR-0 reactor (dimensions, power, used fuel, working regimes) is given, including the experimental possibilities generally and the short scope of the solved problems. The second part describes information about the experimental possibilities, including a programme concerning measurements using MOX fuel.

Introduction

New types of fuel for power reactors have their own procedure of testing, licensing and interpretation, and may have positive as well as negative effects. It is known that the use of MOX fuel is not a cheap solution, also because spent fuel requires different handling and (especially under our conditions) also a different spent fuel dry container. The difference in distance between the vessel wall and the core, in addition to geometrical differences (square versus hexagonal) are of relevance when comparing Western-type PWRs and VVERs. This difference determines the neutron fluence and thus the reactor lifetime; this is relevant for MOX fuelled cores which produce a larger vessel fluence due to a harder neutron spectrum.

The result of measurements in critical facilities consists not only of the information about the neutron fluxes in the ideal core, but also the set of information about the sensitivity of measurements to the geometrical and material characteristics; the latter information is difficult to obtain in cores of power reactors. PWR and VVER reactors have differences in materials, in-core instrumentations, regulations and other parameters. Additionally, the process of core calculation and its verification use slightly different approaches; consequently the designers of VVER are calling for the traditional set of experimental data enabling to understand how measurements and calculations are carried out at LR-0. A change of fuel will lead to changes in the storage capacities and in the licensing of spent fuel storage.

There exist several sets of data from the so called “cold” or room temperature pinwise power distributions and fluxes and corresponding integral data from the various testing cores. The main set is without any doubt that of the TIC set [1] which is now openly and widely used. This was completed by the critical assemblies in the Kurchatov Institute for tight or other regular lattices: 1) small core, big core practically enabling one to one measurement of VVER-1000 cores for the fluxes, kinetics and power shape analyses; and 2) intermediate core in the NRI Řež (the largest critical facility in operation) – enabling to measure at the assembly level and on the pin level. This is especially useful for spent fuel storage simulation as well as simulation of pressure vessel fluence and the reflector influence.

Various measurements were carried out which are described in the following. The process of data evaluation was accompanied by original procedures such as the height reactivity coefficient through the capillarity effects, the upper and lower axial buckling correction and its changes through the boron sedimentation on the upper dry and wet part of the rods through longer measurement cycles.

Experience has also been gained with a core in which two types of lattice are inserted (D_2O and H_2O), and how to use the results for simulating power reactor cores. Even though today Monte Carlo methods and modern computers provide new quality calculations for design, knowledge based on experiences is still badly needed.

An overview of the possibilities offered by LR-0 for the MOX type cores is thus presented.

The fuel amount presently available is about 65 special shortened VVER-1000 type assemblies (about one-half of the full VVER-1000 core) – all dismantlable and some of them enabling measurements inside the rods. The fuel is equally shared between the Czech Republic and Russia.

The LR-0 reactor description

The design of the reactor makes it possible and easy to rearrange the reactor core and to modify the operational regime according to the requirements of an experiment. The aluminium reactor vessel is situated in a concrete shield and covered with mobile shielding platforms. The bottom cylindrical part has a diameter of 3.5 m and a height of 6.5 m. The dimensions enable to realise full scale experiments (in the case of VVER type reactor in the r-direction only) [2].

The basic types of fuel assemblies used are the shortened dismountable models of the VVER-1000 and VVER-440 assemblies. The height of the fuel filling in element is only 1 250 mm. The fuel enrichment varies from 1.6 to 4.4% of ^{235}U . There possibility also exists to design and operate multi-zone cores with an inner (inserted) core to be investigated driven with LR-0 standard assemblies. The LR-0 reactor provides a wide range of benchmarking possibilities with an accurate reproducibility of the parameters.

LR-0 experimental advantages

Experimental advantages of the LR-0 reactor include:

- VVER and PWR related radially full scale experiments;
- flexible rearrangements of the core;
- flexible operation;
- a special supporting plate which enables the triangularly symmetric assembly arrangement with an arbitrary pitch;
- the capability to design and operate multi-zone cores, i.e. substituted cores, with an internally inserted part in hexagonal or square geometry (driven by LR-0 standard assemblies);
- modelling neutron field parameters of power reactors;
- wide range of benchmarking possibilities, with high reproducibility of the benchmark design parameters;
- wide range of measurement techniques including equipment and experienced staff.

Review of projects

A list of projects would include:

- *Pinwise flux distribution measurements.* Radial and axial fission density distributions in the fuel assemblies, e.g. the model of VVER-440 control assembly type with and without partially inserted absorption part, VVER-440 and VVER-1000 mock-ups, compact spent fuel storage lattices, burnable absorber study (Gd and CrB_2), experiments with VVER-440 profiled fuel assembly.

- *Compact spent fuel storage.* Experimental verification of the criticality safety with relation to design parameters and different absorbing materials.
- *VVER-440 and VVER-1000 mock-ups.* Measurements of the space-energy distribution of the neutron flux for RPV dosimetry purposes – in the vicinity of the pressure vessel model (from the core boundary to the biological shielding).
- *Space kinetics experiment.* Benchmark data for space-time kinetics code qualification.
- *Core parameters.* Integral and safety parameters, such as control and shutdown rods integral and differential efficiency, critical water level, water level reactivity coefficient.
- *Experiment with new design fuel assembly.* Influence of new profiled fuel assembly and spacer-grid material on the pinwise spatial flux density distribution
- *VVER-440 control assembly influence.* Measurement of pinwise flux distributions, with and without partially inserted control assembly model.
- *Burnable absorber study.* Power distribution measurements with Gd and CrB₂ absorbers.

Review of the solved problems

Problems already addressed include:

- Control rod and burnable absorber (Gd, CrB₂) benchmark.
- Benchmark experiments for pressure vessel reactor dosimetry.
- Basic reactor physics core parameters measurement in mock-up core.
- Spent fuel storage benchmark.
- Control rod influence benchmark.
- Experiment with new designed fuel assembly.

The experimental possibilities for MOX type fuel pin research

It should be possible to obtain the licence to use MOX fuel in the LR-0 reactor. The appropriate measures as regards the non-proliferation and safeguard aspects, radiological protection and nuclear safety will have to be taken, but these measures are related to available time and financial resources only, and not to the technical problems of the reactor operation or fuel storage and investigation.

An experimental programme in two stages is proposed. The first one concerns the measurements of the basic parameters, the measurements of δk_{eff} , energy distribution, neutron spectrum influence (reaction rates measurements); oscillation measurements with samples are intended at this stage.

The second type of measurement is more complex, and consists of the boron acid effectiveness and control elements effectiveness measurements, the dosimetry of the pressure vessel of the VVER or Western-type PWR reactors using MOX fuel and the problems of compact storage of the fresh and spent fuel.

Basic neutron-physical experiments

- Measurements of critical values of the height in individual configurations and measurements of δk_{eff} caused by the presence of MOX fuel rods.
- Measurements of power distribution (in all cores: radially in the cores higher than 650 cm, and also axially).
- Measurements of weights of the individual absorbing rods and that of their groups (it is anticipated that there will be six absorbing rods concentrically arranged with the pitch 3); these measurements will be complemented with measurements of the radial power distribution in the vicinity of the absorbing rods.
- Measurements of reaction rates, using a wide range of the activation neutron detectors (absolute measurement), comparison with the configuration without MOX fuel rods.

Experiments with asymptotic core

- Measurements of the power density and neutron fluxes in the mixed uranium-plutonium cores.
- Measurements of the boric acid efficiency for the reactivity compensation.
- Reactivity excursion using small specimens of pure plutonium isotopes and minor actinides so as to more accurately determine their neutron-physical characteristics (oscillation methods).
- Measurements inside MOX fuel rods (foils, track detectors); additional analysis of the safety measures and technical arrangements of the measurements is required.

Full-scale measurements (nearly the same as power reactor dimensions)

- Determination of the efficiency of the reactor control and emergency clusters for different variants.
- Verification of compact storage facilities for fresh and spent fuel and of transport casks, including their design and licensing.
- Dosimetry of reactor pressure vessels for VVER reactors with MOX fuel.
- Space kinetics of the MOX cores.

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FIRST CRITICALITY OF LWR-PROTEUS: A NEW PROGRAMME OF INTEGRAL EXPERIMENTS FOR CURRENT, ADVANCED AND INNOVATIVE LWR FUELS

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Abstract

The recently achieved first criticality of the rebuilt PROTEUS facility marks the start of a new experimental reactor physics programme at the Paul Scherrer Institute in Switzerland. The programme, known as LWR-PROTEUS, will address issues of current concern to the Swiss nuclear industry, in particular the validation of reactor physics codes as applied to modern LWR fuels. In addition to investigations for the existing power plants (e.g. detailed power distributions and burn-up related effects), the project provides the possibility for the execution of additional special experiments related to innovative fuels. Following a general introduction to LWR-PROTEUS, this paper briefly describes the planning of such a special experiment, namely the measurement of relative reactivity effects for sample rods of mixed oxide and inert matrix Pu-fuels in a representative PWR spectrum.

Introduction

Improved fuel utilisation, increased plant output/flexibility and more efficient management of the back-end of the fuel cycle are important goals in the further development of LWR technology. These are already being partly realised in current-day plants through the implementation of increasingly complex fuel assembly designs. Such developments are reflected most strongly in the case of BWRs, the combination of demands such as high-burn-up, longer operating cycles and flatter power distributions having led to assembly designs lying well outside the available integral database. For PWRs, the challenges, qualitatively speaking, are somewhat different – larger than desired safety margins currently being necessary, for example, in relation to burn-up and Pu recycle issues.

In the context of the above, the launching of a new programme of “driven” integral experiments at PSI’s zero-power PROTEUS facility has been marked by the recently achieved first criticality of the rebuilt facility. The experiments are being conducted in close collaboration with the Swiss utilities, the principal aim being to reduce the existing uncertainty limits on power distributions and reactivity effects in modern LWR fuel assemblies. There are essentially two parts to the currently scheduled three-year experimental period, the first related more specifically to BWRs and the second more to PWRs.

In the first phase [1], the desired integral database is to be obtained by carrying out measurements on actual full-size BWR assemblies, i.e. before these are introduced into a commercial power reactor. As already indicated, the second part of the experimental programme will be conducted employing a test zone characteristic of a modern PWR core, with the emphasis being placed upon measurements involving high burn-up fuel. Whereas radial and axial power distributions across the central fuel assembly will be of greater importance than reactivity-effect measurements in the BWR experiments, the inverse will be the case for the PWR part of the programme. It is in the latter framework that certain “special” experiments are also being planned to obtain integral data of relevance to innovative fuel designs, such as inert matrix fuels (IMF) for Pu incineration [2].

The current paper provides a general introduction to the LWR-PROTEUS programme and reports briefly on the recent first criticality and results of the commissioning tests of the first configuration. In addition, planning calculations are presented for a series of relative reactivity worth measurements planned in a reference PWR test configuration. These comprise values predicted for IMF and mixed oxide (MOX) fuel rods currently being fabricated at PSI for irradiation tests in the Halden reactor (as per their present nominal specifications [3]).

Layout of the rebuilt facility

The PROTEUS facility, shown in its newly rebuilt form in Figure 1, has undergone significant changes to accommodate the new experiments. The reconfigured system comprises the following distinct zones (from the centre moving outwards; see Figure 2):

Test zone. For the first phase, nine full-size BWR elements of the type SVEA-96+ (ABB Atom) are located in a 3×3 arrangement in an aluminium test tank containing water or polyethylene as moderator. Each SVEA 96+ element comprises 96 fuel pins arranged on a square pitch around a central water canal. The ^{235}U enrichment varies both axially and radially in the range 2-5% and there are 16 Gd-containing pins in each element. Since the elements are some 4.5 m in length and the fuel in the driver regions somewhat less than 1 m

in height, the test tank can be driven axially to enable investigations along the whole length of the (axially heterogeneous) test elements. This is a unique aspect of the experiments and is only possible because of the unusual layout of the facility.

During the BWR-related part of the programme, a number of different test zone conditions will be investigated. These will include different (simulated) moderator densities, presence of (fixed) absorber blades (Hf and B_4C) and asymmetric loadings of elements (simulated channel bowing).

Figure 1. A general cut-away view of the as-built LWR-PROTEUS facility showing the 3×3 arrangement of BWR elements in the vertically drivable test zone

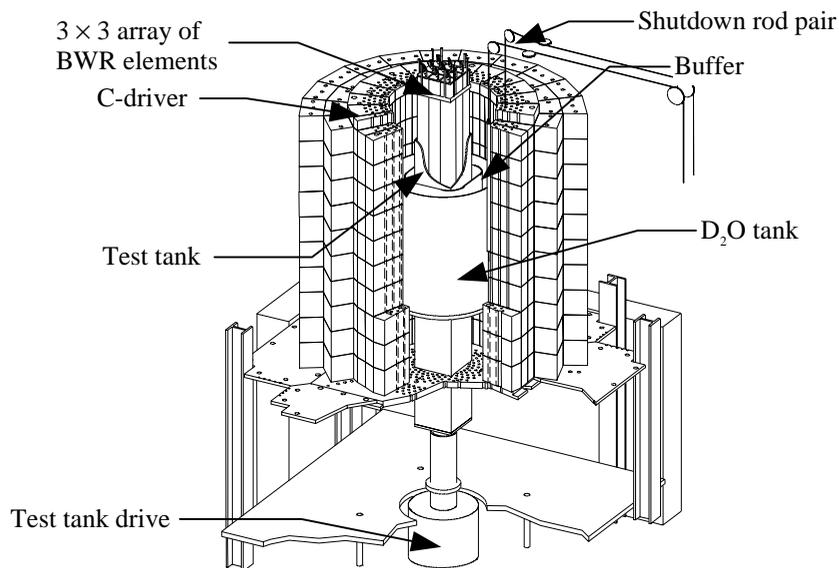
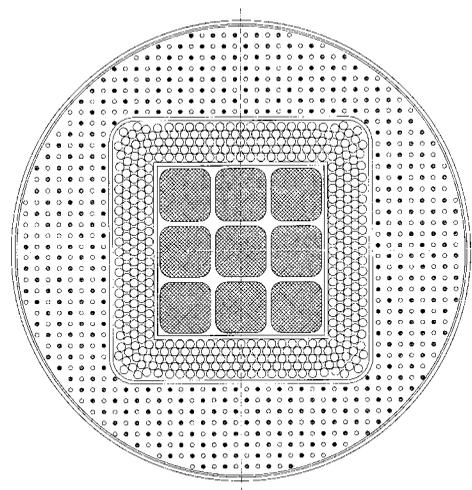


Figure 2. Plan views of LWR-PROTEUS, showing the test, buffer and driver zones



- *Natural uranium buffer.* This serves to minimise the effect of the thermal driver regions on the harder test-zone spectrum. It consists of tightly packed natural uranium metal rods in air.
- *D₂O zone.* This serves both as a driver region and as a neutron bridge acting to maintain sufficient thermal flux in the graphite driver, which in turn guarantees sufficient worth for the safety/shutdown rods. It is fuelled with 5% enriched UO₂ fuel rods.
- *Graphite driver/reflector.* This contains driver fuel, safety, shutdown and control rods and the nuclear instrumentation. The fuel is also 5% enriched UO₂.

Physics of the LWR-PROTEUS system

The configuration described above is a so-called “driven system”, in which an otherwise subcritical test zone is made critical by the surrounding driver zones. The advantages of driven systems can be summarised as follows:

- A smaller amount of test fuel is required.
- A large range of test zone conditions (including $k_{\infty} < 1$ states) can be investigated by changes in the driver loading alone, thus avoiding undesirable perturbations to the test zone which would influence the measurement conditions and thus affect the interpretability of the results.
- The necessary reactor control and instrumentation equipment (usually perturbing from the experimental viewpoint) can be located in the outer driver regions, thereby avoiding disturbance of the test lattice.

This type of system does, however, have some limitations, for instance on the reactivity of the test zone. A highly reactive test zone can reduce the importance of the driver regions to such an extent that the control and shutdown systems have too little worth, which was a problem in the early design phase of LWR-PROTEUS. The problem was overcome by changes in the D₂O driver loading and by an increase in the number of shutdown rods from 8 to 16. The flux distribution for the final design of the system with a 0% voided test zone is shown in Figure 3. Clearly identifiable is the large thermal peak in the driver regions and the effect of the buffer in hardening the spectrum entering the test zone. The effectiveness of the buffer is further indicated in Figure 4, in which the spectral index of the PROTEUS test zone (ratio of fast to thermal flux) is compared with that of a so-called single zone critical arrangement in which the test zone is artificially expanded in the radial direction to achieve criticality.

First criticality and commissioning of LWR-PROTEUS

The rebuilt facility became critical for the first time on 17 November 1998 (i.e. shortly after ARWIF'98), and this has marked the start of low power operation of the facility (up to 1% of the maximal facility power of 1 kW). The criticality predictions overestimated the system reactivity by between 0.5% and 1.5% in k_{eff} . Considering the complexity of the whole-reactor arrangement, this is considered to be satisfactory agreement. The discrepancy does not, in any case, have any influence

Figure 3. Thermal, fast and total neutron fluxes in the various radial zones of LWR-PROTEUS

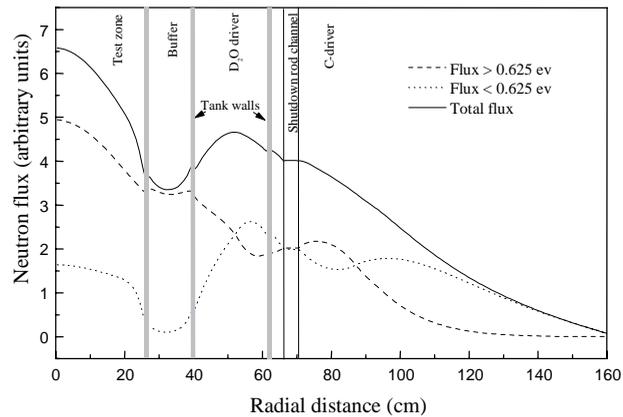
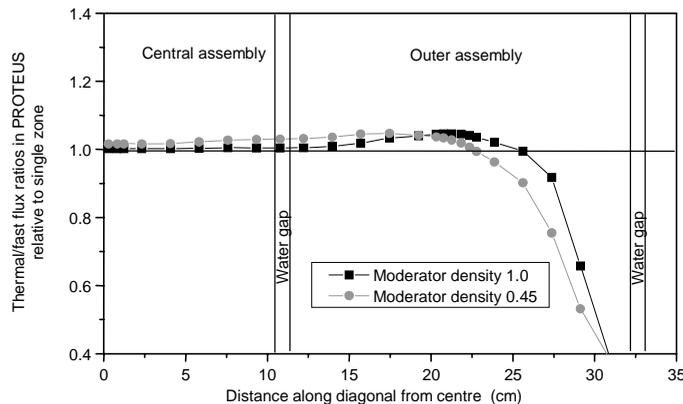


Figure 4. Special index variations in LWR-PROTEUS relative to a single zone system



on the reliability of the investigations to be conducted in the test zone. All safety parameters have now been measured and have been shown to lie within the required limits. Following the completion of the remaining commissioning measurements, the reactor power will be increased in a stepwise fashion to its maximal licensed level.

LWR-PROTEUS with a PWR core

In parallel to the planning of experiments for the first phase, certain calculations have also been carried out for the second phase in which a PWR-type test zone will be created. The general nature of most PWR fuel (higher average enrichment as compared to BWR fuel, no burnable absorber, etc.) means that, in the absence of soluble boron in the moderator, the reactivity of PWR lattices is significantly higher than that of BWR lattices (typical cold k_{∞} values of 1.45 compared with 1.1 for BWR lattices). Because, for safety reasons, it was decided at the outset to avoid the use of soluble boron in PROTEUS, an alternative approach has been taken in which the outer regions of the test zone are poisoned with fixed absorber rods containing B_4C pellets. Figure 5 shows the current design of the planned PWR test zone which, assuming the characteristics of fuel from one of the operating Swiss nuclear power plants, has been shown to satisfy the conflicting requirements of sufficient

Figure 5. Changes in test zone from Phase 1 to Phase 2 (quarter test zone)

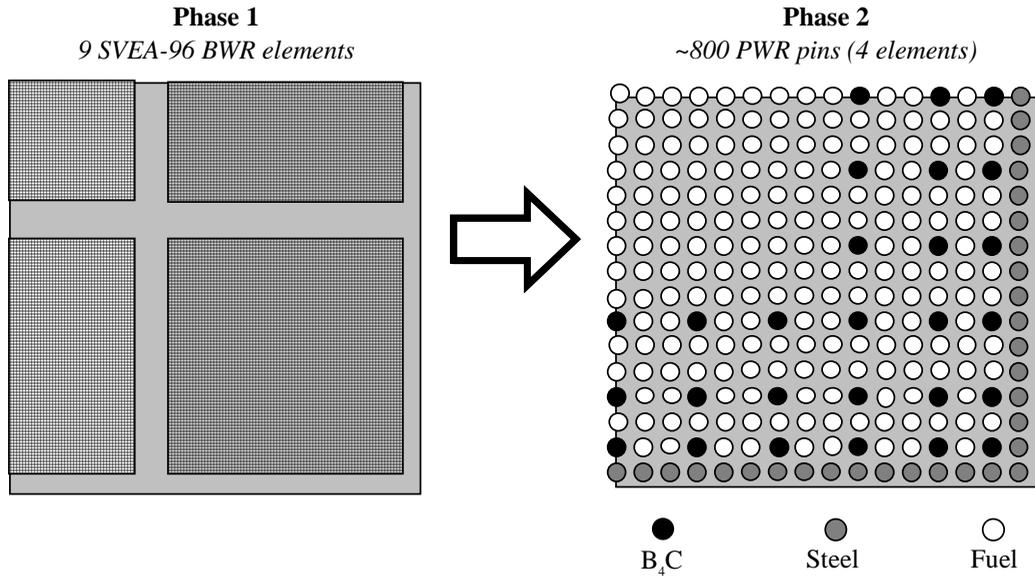
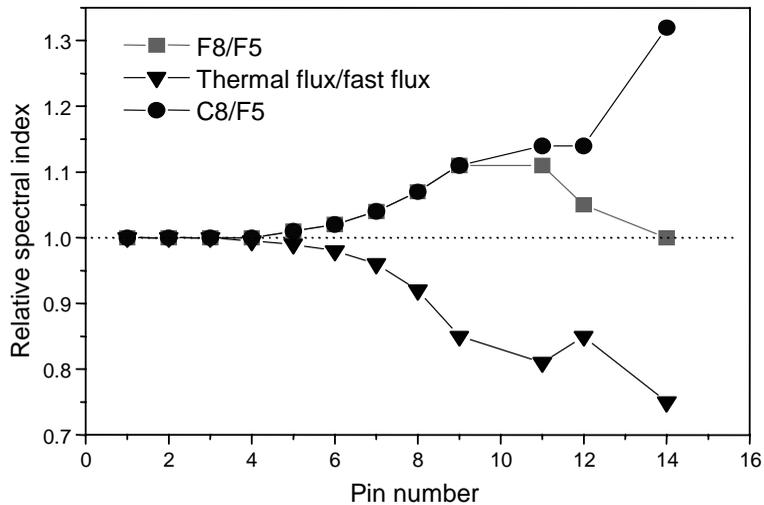


Figure 6. Spectral index of proposed PWR test zone



shutdown margin and a suitably sized unperturbed test zone. Figure 6 shows the spectral properties of the new test zone, in which a central region of about 9×9 size can be clearly identified as having a well defined equilibrium spectrum. It is planned to carry out reactivity effects measurements with high burn-up pins in the centre of this region.

Planning calculations for additional experiments to assess the reactivity effects of innovative fuels

For the test zone layout described above, and in addition to the planning for the mentioned burn-up related measurements, two-dimensional calculations were carried out to estimate reactivity worths for a range of different innovative fuel rod types (see Table 1). The calculations were

Table 1. Reactivity effects of replacing the central fuel rod in the PWR test zone by different innovative fuel rod types

	Composition of sample rod (g.cm ⁻³)					Reactivity		
	U	Pu	Am	Zr	Y	Er	ϵ ^a	ϵ ^b
MOX 1	8.45	0.76	0.0				-3.00	
MOX 2	8.45	0.78	0.033				-4.70	
IMF 1		0.9	0.0389	3.345	0.335	0.36	-11.2	-10.8
IMF 2		0.9		3.345	0.335	0.36	-9.7	-9.3
IMF 3		0.93		3.328	0.335	0.38	-9.6	-9.3

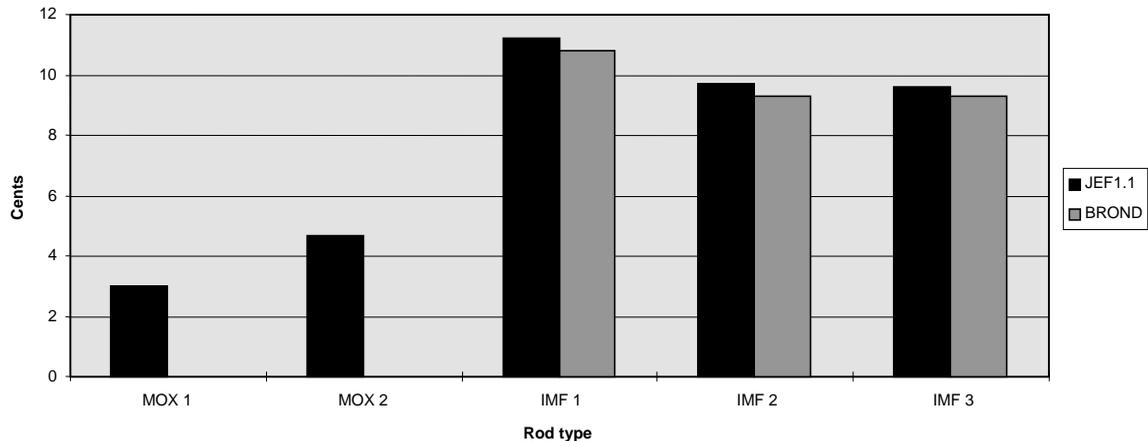
^a JEF1.1

^b BROND-2

performed with a 2-D model of the full PROTEUS reactor, using the code BOXER of the ELCOS system [4] in conjunction with a well established broad group structure for LWR applications (24 groups, of which 8 thermal groups below 1.3 eV). For each calculation, the central fuel rod of the test zone was replaced by the sample rod to be investigated and the reactivity change observed. To simulate the surrounding neutronic environment of the single rod (consisting of either MOX or the PuO₂/Er₂O₃ inert matrix fuel of interest [2]), the cell with the standard (4.3 wt% enriched) UO₂ fuel was calculated in the fundamental mode spectrum using white reflection boundary conditions, its outgoing partial current serving as boundary condition for the cell with the sample fuel under consideration. The cross-section library was based upon the Joint European File, Rev. 1 (JEF-1.1) [5]. In addition, and in the framework of a data sensitivity study for the burnable poison, the data for the erbium isotopes were also taken from the Russian file BROND-2 [6].

The calculated reactivity effects are given in the last two columns of Table 1 and are also plotted below in Figure 7. It is seen that the signals to be obtained are of the order of a few cents. Furthermore, for the IMF rods, the differences observed in the use of JEF and BROND data for erbium are seen to be of the order of 0.5 cent.

Figure 7. Calculated reactivity effects of the various sample rods



From past experience at PROTEUS, it is known that reactivity effects of up to 10 cents can be measured with a 1σ uncertainty of 0.1% (i.e. 0.01 cent). On this basis, the observed differences between the BROND and JEF datasets, for example, can clearly be resolved by the planned measurements.

Summary

With first criticality achieved on 17 November 1998, a new series of experiments has been launched at the PROTEUS facility, with the aim of providing an up-to-date validation base for LWR design and core analysis tools.

The first phase of the programme principally involves the investigation of power distribution and reactivity effects in advanced BWR fuel assemblies. A second phase, currently being planned, will address PWR-related fuel cycle issues.

In addition to the commercial goals, the project provides an appropriate framework for special experiments in which physics aspects of innovative fuels can be addressed.

Acknowledgements

The support of the Swiss Nuclear Utilities (UAK) and EGL/KKL is gratefully acknowledged. Valuable contributions to the LWR-PROTEUS programme have been made by a number of persons. In particular, we would like to thank: R. Brogli of PSI, S. Helmersson and M. Gustavsson of ABB Atom, W. Färber and R. Jacot-Guillarmod of EGL, J. Axmann, R. Stamm'ler and S. Borressen of Studsvik Scandpower, as well as the staff of the technical department (B8) of PSI and members of the PROTEUS operational team.

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CONCEPTUAL DESIGNING OF WATER-COOLED REACTORS WITH INCREASED OR REDUCED MODERATION

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Abstract

Conceptual designing of some kinds of advanced reactors is being performed at JAERI. One of them is a water-cooled MOX fuelled reactor with a high burn-up of around 100 GWd/t and a long operation cycle of around three years, aiming at economical advantages, reduction of spent fuel production and Pu deployment. Another is a water-cooled very high conversion, i.e. breeding, reactor with a conversion ratio of more than 1. This is so as to establish a long-term energy supply with the experienced water-cooled reactor technology. This paper presents some results from these conceptual design studies on water-cooled reactors. For the high burn-up reactors, a PWR-type design with increased moderation and a BWR-type design with reduced moderation have been proposed. Our current design values of initial Pu (fissile) content are in the range of 11-12 w/o. For the high conversion reactor, a feasibility study is in progress on a heavy water-cooled PWR-type reactor with a significantly reduced moderator to fuel volume ratio of around 0.5. At present, a heterogeneous design with some radial blankets seems to be promising to attain both the high conversion ratio of more than 1 and the negative void coefficient simultaneously.

Introduction

Conceptual designs of some types of advanced reactors are being undertaken at the Japan Atomic Energy Research Institute (JAERI). One of them is for high burn-up and long cycle operation achievement with a water-cooled full MOX core. Our design goals for this are 100 GWd/t burn-up and a three-year cycle length with the full MOX core. This is intended to be one of promising options to reduce spent fuel production, refuelling frequency and Pu accumulation, resulting in economical advantages under the current perspective of extended water-cooled reactor utilisation in Japan.

For PWR-type reactors, we are proposing designs with somewhat increased moderation. That is, the ratio of the moderator to the fuel (V_m/V_f , referred to as the “moderation ratio” hereafter) ranges between 2.5 and 3. This selection is based on some parametric surveys for the moderation ratio effects on burn-up, reactivity coefficients, the DNBR and so forth [1]. In the framework of this study, we have also made another design for 60 GWd/t burn-up and a two-year cycle core as an intermediate step for the above design considering faster and easier realisation of this type of concept.

For the BWR-type reactors, on the other hand, we are proposing designs with slightly reduced moderation, i.e. a moderation ratio around 2, compared to the current ABWR design, whose moderation ratio is around 3. This difference in the direction of the moderation ratio is based on the larger margin of control rod equipment in the current BWR-type reactors.

Another conceptual design has recently been performed with a significantly reduced moderation ratio for a very high conversion ratio achievement. Our design goals for this are a conversion ratio more than 1 and a negative void coefficient simultaneously. For the PWR-type reactors, for example, a feasibility study for the core with the gap width between rods of around 1 mm using the heavy water as the moderator is under investigation, and some promising results have been obtained.

High burn-up and long operation cycle PWR-type reactors

Cell burn-up characteristics parametric survey

In order to achieve a much higher burn-up and a longer operation cycle than is currently the case, it is beneficial to use MOX fuel because of its lower reduction in reactivity during burn-up. Figure 1 shows one example for burn-up behaviours of reactivity comparing 5%-enriched UO_2 fuel case with 14 w/o Pu content full MOX cases in four different V_m/V_f values. This clearly indicates a much lower reduction in reactivity for any full MOX case. This figure also shows that a burn-up of 100 GWd/t may be attained with a Pu content of 14 w/o and a V_m/V_f of more than 2.5, if a three-batch refuelling scheme is introduced, and that the increase in attainable burn-up becomes smaller as V_m/V_f becomes larger.

These results were obtained by cell burn-up calculations for a one-dimensional equivalent cylinder model with the SRAC code system [2]. The isotopic composition of the fuel used in the present calculations is given in Table 1. The base fuel material is assumed to be depleted U of 0.2% ^{235}U content.

The dependence of the soluble boron efficiency on V_m/V_f and Pu content is summarised in Figure 2. The boron efficiency is much smaller than that for the current UO_2 fuel core (about 8 pcm/ppm). This suggests that the necessary critical boron concentration would be about twice at the beginning of cycle (BOC). At any Pu content investigated, the boron efficiency increases proportionally with increase in V_m/V_f . It decreases when Pu content increases, and increases when burn-up increases.

Figure 1. Burn-up behaviours of reactivity for various Vm/Vf values at a Pu content of 14 w/o

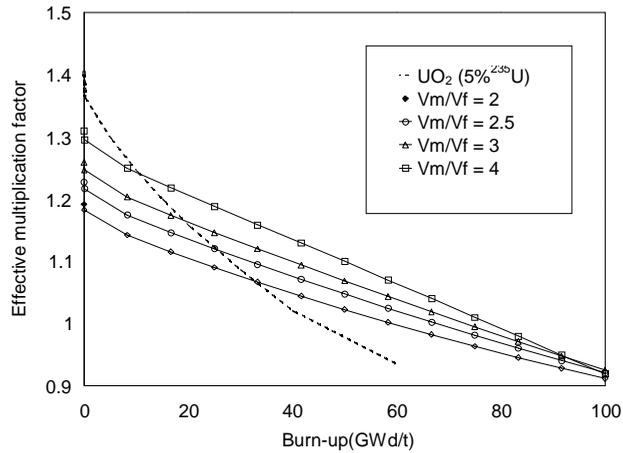
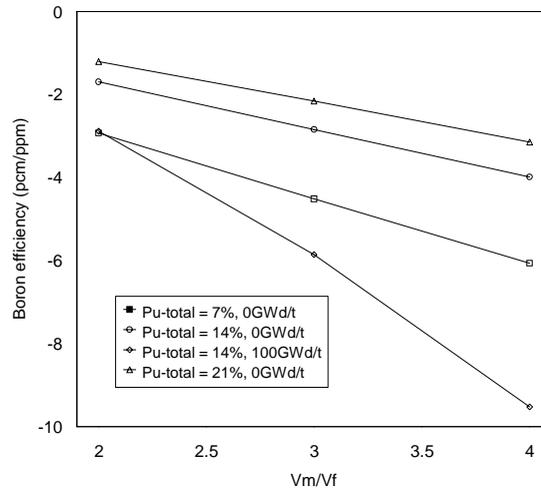


Table 1. Isotopic composition of fuel material

Pu (w/o)						Pu (w/o)	
238	239	240	241	242	²⁴¹ Am	Pu	Pu-fis.
2.94	52.60	23.74	11.67	7.24	1.81	1	0.6427

Figure 2. Dependence of soluble boron efficiency on Vm/Vf and Pu content



Dependence of the void coefficient on Vm/Vf at a void fraction of 90% and BOC is presented in Figure 3 for 14 w/o Pu content at the critical or no boron concentration. The void coefficient decreases in negative value with an increase in Vm/Vf. The dependence of the moderator temperature coefficient on Vm/Vf at BOC is also shown in Figure 4 for a Pu content of 14 w/o at the critical or no boron concentration. The coefficient is almost constant with no boron, whereas at the critical boron concentration it increases with increase in Vm/Vf. It should be noted it becomes positive for Vm/Vf value more than about 3.5. These results suggest the void and moderator temperature coefficients be negative for Vm/Vf value smaller than 3 at a Pu content of 14 w/o.

Figure 3. Dependence of void coefficient at 90% void on V_m/V_f at BOC for 14 w/o Pu content

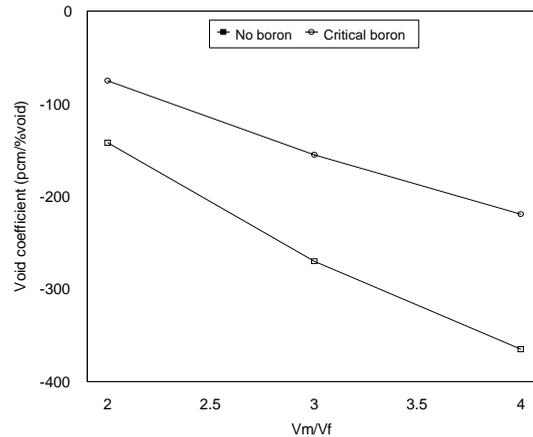
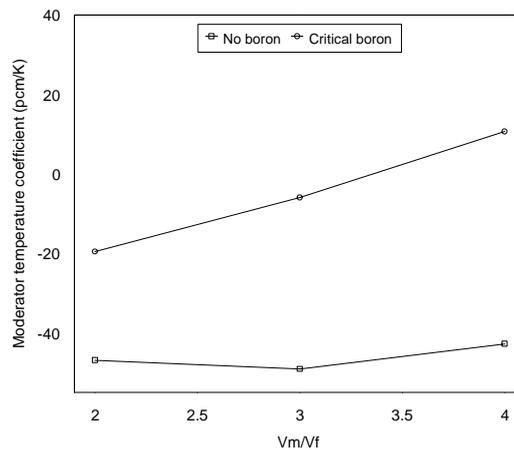


Figure 4. Dependence of moderator temperature coefficient on V_m/V_f at BOC for 14 w/o Pu content

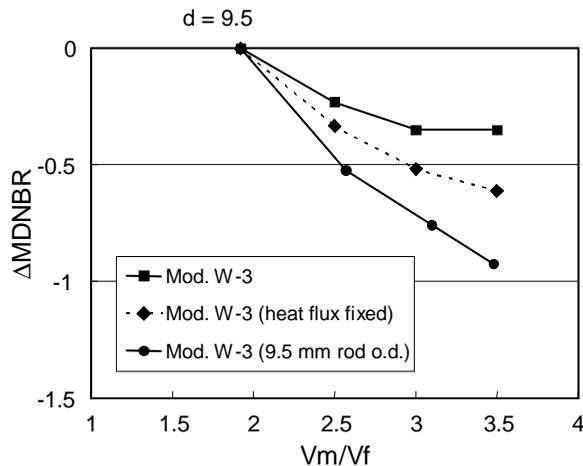


Core specifications

For the full MOX core design, some investigations have already been performed. Tochiyama *et al.* [3] proposed a design for the advanced PWR with a conventional V_m/V_f value of about 2. For this design, the fissile Pu content is 5.9 w/o. The maximum assembly burn-up and the cycle length are 47 GWd/t and 13.5 EFPM, respectively, with a three-batch refuelling scheme. Barbraut [4] and Girieud *et al.* [5] proposed some designs with increased moderation aiming at efficient consumption of Pu and sufficient control worth. V_m/V_f values are 2.5 and 4.0, respectively. The former attains a burn-up of 60 GWd/t with a Pu content of 9% and a four-batch refuelling scheme. The latter attains a burn-up of 55, 66 and 60 GWd/t with a Pu content of 7.5% and a four-batch refuelling scheme, 9.7% and three-batch, and 9.9% and two-batch, respectively. Their cycle lengths are 12, 18 and 24 months, respectively. Our design goals, however, are different from theirs and require a much higher burn-up and longer cycle length. That is, a 100 GWd/t burn-up and a three-year cycle length, although 60 GWd/t burn-up and a two-year cycle core is also investigated as an intermediate step of the above design, taking into consideration the faster and easier realisation of this type of concept.

Based on the above information, it has been clarified that higher moderation is more beneficial from the reactor physics point of view, resulting in higher burn-up, larger control worth, the smaller Pu content necessary and so forth. On the other hand, higher moderation generally causes some difficulties such as reduction of core power density, increase in core diameter, reduction of DNBR and so on. Figure 5 shows calculation results for the V_m/V_f effect on the reduction of minimum DNBR (MDNBR) from that for the current 17×17 type assembly case under normal operating conditions. The subchannel analysis code COBRA-IV-I and the modified W-3 DNB correlation were used for the calculations. This indicates MDNBR decreases significantly with an increase in V_m/V_f , keeping the fuel rod diameter the same. When we reduce the fuel rod diameter, the reduction in DNBR is much smaller and some increase in the average linear power density might be possible fixing the heat flux from the fuel rods as shown in the figure. However, an increase in the average linear power density causes a decrease in the cycle length, and hence, we decided not to increase the average linear power density in our design. Including these, the appropriate range of V_m/V_f for our design is considered to be between 2.5 and 3.0. From the reactor physics point of view, the value of 3.0 is better, whereas from the thermal design point of view, 2.5 is better.

Figure 5. Dependence of MDNBR reduction on V_m/V_f



One more point is the limit for the reactor pressure vessel inner diameter, which is assumed to be 5.2 m as in the APWR design for 1 400 MWe. Although, at present, the system for JAERI's passive safety reactor JPSR [6] with 600 MWe output is considered to be used with the concerned core, extension to the power output of 1 100 MWe should be considered to be possible. From this point, a lower V_m/V_f value is appropriate. Considering all the above information, we selected $V_m/V_f = 2.6$ as the first core specification investigated, resulting from the pitch of 13.8 mm with rods of 9.5 mm in diameter. Detailed specifications of the fuel assembly and the core are summarised in Tables 2 and 3, respectively.

The specifications of the assembly are almost the same as that of the existing 17×17 , except for the difference in rod pitch. The average linear power density is 16 (kW/m) and is about 10% reduced. This is for extension of the cycle length and an increase in MDNBR. Specifications of the control rod clusters are summarised in Table 4. A larger control rod in diameter is adopted using natural B_4C as the absorber. The fuel loading pattern is "out-in" type, to establish the lowest radial power peaking factor, as shown in Figure 6.

Table 2. Specification of fuel assembly

Assembly	Vm/Vf	2.6
	Type	17 × 17
	Dimension (mm)	235 × 235
	Number of fuel pins	264
	Number of thimble tubes	24 + 1
	Rod pitch (mm)	13.8
Fuel rod	Rod diameter (mm)	9.5
	Cladding thickness (mm)	0.57
	Cladding material	Zircaloy-4
	Fuel gap width (mm)	0.085
Fuel pellet	Pellet diameter (mm)	8.19
	Fuel material	MOX
	Density (%T.D.)	95

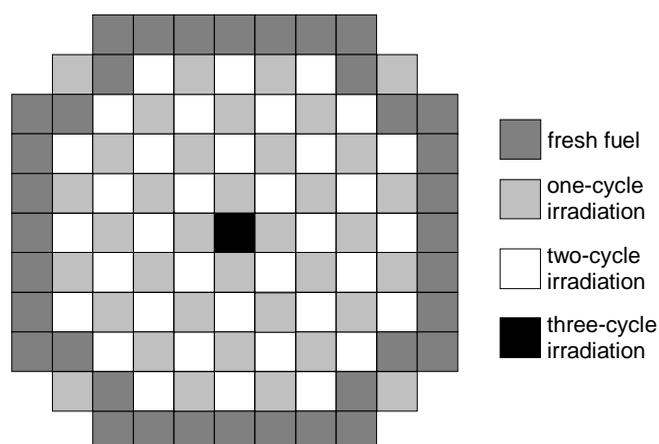
Table 3. Core configuration

Number of batch	3
Thermal output (MWt)	1 685
Electric power (MWe)	573
Number of fuel assemblies	109
Equivalent core diameter (cm)	277
Effective core height (cm)	366 (12 ft.)
Geometrical buckling (cm ⁻²)	3.7521E-4
Average linear heat rate (kW/m)	16.0
Average power density (kW/l)	76.5
Coolant condition	
Inlet temperature (C)	289
Outlet temperature (C)	325
Pressure (atm)	152

Table 4. Configuration of control rod clusters

Outer diameter of thimble tube (mm)	13.4
Thickness of thimble tube (mm)	0.4
Material of thimble tube	SUS-316
Control rod outer diameter (mm)	10.9
Control rod cladding thickness (mm)	0.5
Cladding material	SUS-304
Absorber	Natural B ₄ C
Absorber density (%T.D.)	80

Figure 6. Core cross-section and fuel loading pattern



Core burn-up characteristics

Based on the core specifications presented above, core burn-up calculations have been performed with the SRAC-COREBN code [2]. The results obtained are summarised in Table 5. The necessary contents of Pu-fissile are 12.1 and 7.0 for 100 and 60 GWd/t cases, respectively. Cycle lengths are 33.3 and 19.9 EFPM, respectively. Radial power peaking factors are 1.31 and 1.20, respectively, with an additional local peaking factor of 1.11. Critical boron concentrations are 4 200 and 2 800 ppm, suggesting adoption of 40% enriched boron in ^{10}B . The moderator temperature and void coefficients are negative and no burnable poison is necessary. Reactivities for hot shutdown and the control rod worth are summarised in Table 6. A shutdown margin of 2% is assumed. Necessary numbers for control rod clusters are 40 and 36 out of 109 assemblies for 100 and 60 GWd/t cases, respectively.

Although no burnable poison is necessary in the present designs, the effects of gadolinium (Gd_2O_3) and erbium (Er_2O_3) on reduction of power peaking factor are investigated. They are assumed to be homogeneously distributed in the fuel. Based on the results, most effective contents are 0.02-0.05 and 0.4-0.8 w/o for Gd_2O_3 and Er_2O_3 , respectively, achieving reduction of the peaking factor to the magnitude of about 0.1. The loading pattern effects on the necessary initial Pu content and the power peaking factor are under investigation.

For the obtained design, the DNBR evaluations under normal operating conditions have been performed [7]. Also, some typical safety analyses such as loss-of-flow accidents and loss-of-coolant accidents have been performed [8]. These results have shown that a sufficient safety margin exists in the present designs.

High burn-up and long operation cycle BWR-type reactors [9]

For BWR-type reactors, on the other hand, we started with the ABWR core design, assuming to use the ABWR system design. The fuel assembly is then based on the current 9×9 type design. That is, the diameter and pitch of the rods are 11.2 and 14.3 mm, respectively. By performing the single rod cell calculations, basic characteristics were investigated.

Table 5. Core characteristics of the equilibrium cycle

Average discharged burn-up (GWd/t)	60	100
Cycle length (EFPM)	19.9	33.3
Pu-fissile content (w/o)	7.0	12.1
Pu-fissile inventory for equilibrium cycle (kg)	1 140	1 990
Isotopic composition of discharged fuel (^{238, 239, 240, 241, 242} Pu)	4.8/34.1/30.5/17.1/13.6	5.7/33.9/30.1/16.1/13.5
Peaking factor		
F _{xy} (BOC-HFP)	1.33	1.45
F _{xy} *F _z (BOC-HFP)	1.69	1.94
F _{local} (BOC-HFP)	1.11	1.11
Maximum linear heat rate (kW/m)	29	34
Burn-up reactivity (%dk/kk)	9.8	10.6
Boron efficiency (pcm/ppm) (BOC/EOC)	-3.5/-4.1	-2.5/-2.9
Critical boron concentration (ppm) (BOC)	2 800	4 200
Moderator temperature coefficient at critical boron concentration (pcm/C) (BOC)	-26	-11
Moderator temperature coefficient at no boron (pcm/C) (BOC/EOC)	-61/-65	-49/-54
Doppler coefficient (pcm/C) (BOC/EOC)	-2.7/-2.8	-2.5/-2.6
Moderator void coefficient at critical boron concentration (pcm/%void) (BOC)		
0~10/ 0~40/ 0~90%void	-99/-147/-240	-43/-69/-22
Effective delayed neutron ratio (BOC)	0.0038	0.0036
Prompt neutron life time (10 ⁻⁶ sec) (BOC)	8.8	7.2

Table 6. Reactivity for hot shutdown and control rod worth

Burn-up (GWd/t)	60		100	
	BOC	EOC	BOC	EOC
Reactivity (%dk/kk)				
– Doppler	0.82	0.85	0.74	0.79
– Moderator temperature	1.09	1.17	0.88	0.98
– Reconstruction of neutron flux distribution	1.0	1.0	1.0	1.0
– Limit of insertion of control rod	0.5	0.5	0.5	0.5
– Shutdown margin	2.0	2.0	2.0	2.0
Total (%dk/kk)	5.41	5.51	5.12	5.27
Number of control rod clusters	36	36	40	40
Control rod worth with one-rod stuck condition (%dk/kk)	6.02	6.27	5.24	5.92

As the first step, the possibility of the uniform content distributions of Pu and gadolinium were investigated. The SRAC code was used for two-dimensional (XY) fuel assembly cell calculation. The results showed it is not feasible to use the uniform content distributions of Pu and gadolinium, because the local power peaking factor in the assembly becomes significantly large due to the presence of the water gap region for control rods resulting in strong non-uniform effects.

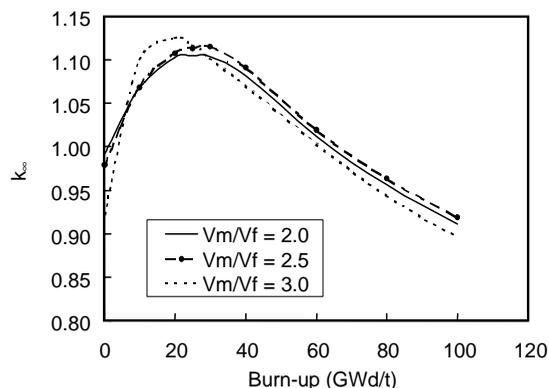
Then, some combinations of three to five different Pu and gadolinium contents in an assembly are used to reduce the local peaking factor. Also, the effects of V_m/V_f have been investigated by changing the number of water rods or the fuel rod diameter. The major parameters of three fuel assemblies investigated are summarised in Table 7. Values for V_m/V_f range from 2.0 to 3.0, which is the same value as for the current ABWR core design.

Table 7. Fuel assembly major parameters

V_m/V_f	Fuel rod number	Water rod number	Fuel rod diameter (mm)	Average Pu content (w/o)	Combination of $\text{PuO}_2/\text{Gd}_2\text{O}_3$ contents (w/o)
3.0	72	9	11.2	13.3	14/0.8, 12/0.8, 7/5
2.5	81	0	11.2	15.4	19/0, 12/1, 11/1, 11/6, 7/8
2.0	81	0	11.9	15.9	20/1, 12/1, 11/1, 11/6, 7/8

Figure 7 shows the burn-up behaviour of reactivity. All the cases can attain our goals with a four-batch refuelling scheme by adjusting Pu contents. Void coefficients have also been investigated and are negative. Local power peaking factors are in the range of 1.2-1.3 for all cases.

Figure 7. Burn-up behaviour of reactivity for three different V_m/V_f cases

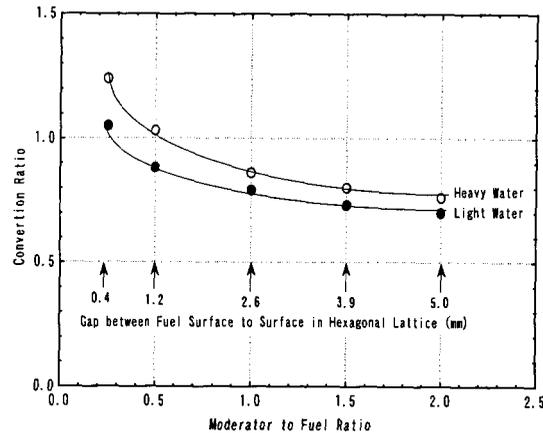


For the $V_m/V_f = 2.0$ case, three-dimensional core calculations were performed with the SRAC code and the BWR design code. Through these results, the main characteristics obtained in the fuel assembly cell calculations mentioned above have been confirmed to be good. Also, the shutdown margin was evaluated with the BWR design code for the same case. The results indicate enough shutdown margin even for the severest case of $V_m/V_f = 2.0$.

High conversion heavy water-cooled PWR-type reactor [10]

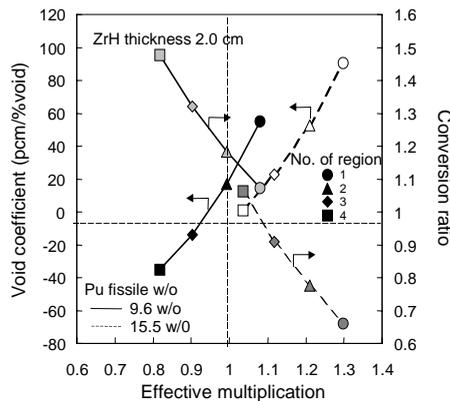
Another conceptual design has recently been undertaken with a significantly reduced V_m/V_f value for very high conversion ratio achievement. Our design goal for this is a conversion ratio more than 1. Figure 8 shows the results of a basic parameter survey on the conversion ratio with varying V_m/V_f for both the light water and the heavy water coolant cases. This indicates the conversion ratio becomes more than 1, if V_m/V_f is reduced to around 0.5 with the heavy water coolant. For this case, the gap width between rods is about 1 mm for 9.5 mm rods. This seems to be close to the attainable range. For the light water coolant, V_m/V_f should be around 0.25 to achieve a conversion ratio of more than 1. This value seems to be significantly difficult to achieve in a PWR-type reactor.

Figure 8. Dependence of conversion ratio on V_m/V_f and coolant water



For a tight lattice core, the void coefficient tends to become positive. This might be a serious problem to be overcome. To make the void coefficient negative, a short core is effective. Also, radial inner blankets have the effect to reduce it. If a zirconium hydride layer is placed between the fuel and the blanket regions, this effect increases with an increase in void. Therefore, this effect has been investigated in detail by increasing the number of the blanket region. Figure 9 shows the results and if there are blanket regions more than 4, the void coefficient might be negative. Based on this, core calculations have been performed for five radial regions case. For this case, the axial inner blanket is also introduced to 2.3 m effective length core. The calculated results satisfy requirements for the void coefficient, the conversion ratio and the effective multiplication. Although the burn-up is not high enough and about 45 GWd/t for the present case, these promising results encourage further study.

Figure 9. Dependence of void coefficient, conversion ratio and effective multiplication on number of region



Another important point is the thermal hydraulic difficulty. For this type of very tight lattice core, reflooding following the LOCA is expected to be significantly severe as well as the critical heat flux. Therefore, feasibility studies on these thermal hydraulic issues should be conducted simultaneously, especially including experimental investigation.

Summary

This paper presents some results from conceptual design studies on water-cooled reactors. One is for a high burn-up of 100 GWd/t and a long operation cycle of three years. A PWR-type design with increased moderation of $V_m/V_f = 2.6$ and a BWR-type design with reduced moderation of $V_m/V_f = 2.0$ have been proposed. For the high conversion reactor, a feasibility study is in progress on a heavy water-cooled PWR-type reactor with $V_m/V_f = 0.5$. At present, a heterogeneous design with some radial blankets seems to be promising to attain both a high conversion ratio of more than 1 and a negative void coefficient simultaneously.

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STUDY OF ADVANCED LWR CORES FOR EFFECTIVE USE OF PLUTONIUM

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Abstract

Advanced technologies of full MOX cores have been studied to obtain higher Pu consumption based on advanced light water reactors (APWRs and ABWRs). To this aim, basic core designs of high moderation lattice (H/HM: 5~6) have been studied with reduced fuel diameters in fuel assemblies for APWRs and those of high moderation lattice (H/HM: 6~7) with addition of extra water rods or reduced fuel diameters in fuel assemblies for ABWRs. The design and analysis of equilibrium cores show that nuclear and thermal hydraulic parameters satisfy the design criteria and the Pu consumption rate increases up to 47% for APWRs and up to 22% for ABWRs.

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Introduction

The recycling of plutonium in light water reactors is expected to continue for several decades. In Japan, the MOX fuel demonstration programmes have been performed successfully and reload size use of MOX fuel has been prepared for loading in 1999. The target of reload size use in the near future is about 1/3 of a core, and a full MOX core is also planned in an ABWR plant. As medium and long term development, core concepts have been studied for enhancing effective use of plutonium in LWRs. For this purpose, higher moderator cores have been studied in full MOX fuel cores based on ABWR and APWR core design [1-2]. NUPEC has been conducting this study on behalf of the Japanese Ministry of International Trade and Industry (MITI). This paper presents results of the core and fuel designs and core characteristics of equilibrium cycles of these advanced LWRs.

Design conditions and target of study

The study was performed based on cores of the advanced LWRs (APWRs and ABWRs) [3-4]. The basic specifications of those plants are shown in Table 1. The parameters in Table 1 and the power densities of cores have been conserved in this study.

Table 1. Basic design parameters

	APWR	ABWR
Rated thermal power	4 100 MW	3 926 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	17 × 17	9 × 9
Maximum burn-up	55 GWd/t	55 GWd/t

As an index to measure the efficiency of plutonium use, the “fissile plutonium consumption rate” was defined as follows:

$$\text{Fissile plutonium (Puf) consumption rate} = \frac{(\text{Loaded fissile plutonium} - \text{Discharged fissile plutonium})}{(\text{Loaded fissile plutonium})}$$

The target of the study is to increase this Puf consumption rate. Systematic sensitivity studies of the Puf consumption rate have been performed for the main design features of LWR cores. The results have shown that an increase in the moderation ratio of the cores is most effective to increase the Puf consumption rate.

In this study, two highly moderated assemblies with differently increased H/HM values were investigated and their performances were evaluated.

High moderation PWR

A preliminary fuel design study was performed to increase the core moderation ratio by two methods: (1) reducing the fuel pin diameter; and (2) replacing fuel pins by water rods. The influence of these two methods on in-assembly power distribution and thermal margin has been studied with

a 17×17 fuel assembly. As shown in Figures 1 and 2, the fuel design with reduced diameter has the advantage in both power distribution and thermal margin compared with the design applying the water rods.

Figure 1. Local power peaking of high moderation assemblies (PWR)

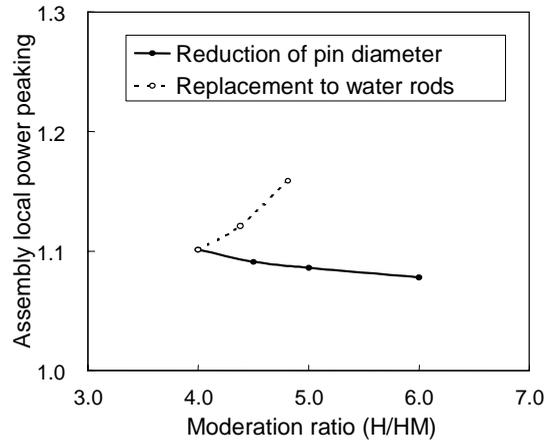
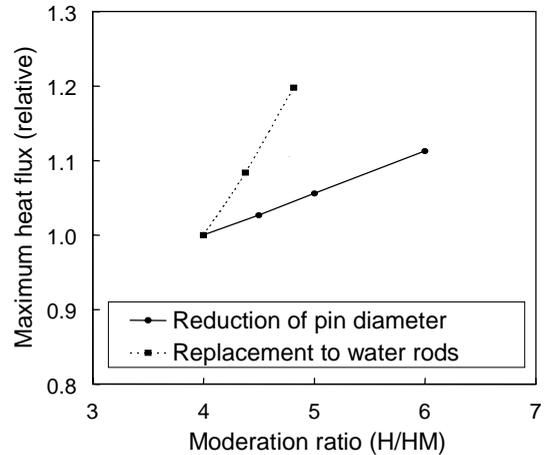


Figure 2. Maximum heat flux of high moderation assemblies (PWR)



While the fissile plutonium consumption rate increases with an increasing moderation ratio, the acceptable increase of heat flux is limited by thermal hydraulic margins. Therefore, the fuel design with a reduced diameter was selected to obtain the maximum effect. As for the first design, the moderation ratio was set to 5.0 as its hydrogen to heavy metal atomic ratio (H/HM) to maintain an adequate thermal hydraulic margin (highly moderated assembly-1). Additionally as the second design, H/HM was set to 6.0 to evaluate the feasibility of a highly moderated core (highly moderated assembly-2). In these designs, the fuel pin diameter is reduced from 9.5 mm (H/HM = 4.0) to 8.8 mm (H/HM = 5.0) or 8.4 mm (H/HM = 6.0) keeping the same fuel pin pitch.

The specifications of the fuel assembly are summarised in Table 2. The initial fissile plutonium enrichment is uniform in the assembly and is determined to meet the required cycle length. No burnable absorber is included in the assembly because they are not required in PWR full MOX cores. Table 2 shows the specifications of those fuel assemblies.

Table 2. Specifications of highly moderated MOX assembly (PWR)

	Reference 17 × 17 assembly	Highly moderated assembly-1	Highly moderated assembly-2
Moderation ratio (H/HM*)	4.0	5.0	6.0
Fuel pin diameter	9.5 mm	8.8 mm	8.4 mm
Assembly lattice	17 × 17	17 × 17 (same pitch)	17 × 17 (same pitch)
Number of thimbles	25	25	25
Fissile plutonium enrichment**	7.2 wt%	5.8 wt%	5.1 wt%
Matrix	Depleted uranium	Depleted uranium	Depleted uranium
Burnable absorber	–	–	–

* Atomic ratio of hydrogen to heavy metal.

** Enrichment is uniform in the assembly.

Under the condition of keeping the basic core design parameters shown in Table 1, two equilibrium core designs with each highly moderated assembly were performed. A full MOX core design with original 17 × 17 fuel assemblies was also performed as the reference core of these advanced designs. As for the reference full MOX core, the basic feasibility and its characteristics with the lower fuel burn-up limitation have already been studied and reported [5-7].

The major specifications of the high moderation cores are shown in Table 3. The effect of increasing H/HM is the decrease of required fissile plutonium enrichment for the same operation cycle length. The fissile plutonium enrichment for the high moderation cores-1 and -2 are 1.4 wt% and 2.1 wt% less than the reference core. On the other hand, the number of refuelling assemblies increases from 88 to 108 or 120 due to the decrease of fuel inventory of the core. The maximum assembly burn-up for both cores is less than the limitation (55 GWd/t).

Table 3. High moderation core specifications (PWR)

	Reference core	High moderation core-1	High moderation core-2
Moderation ratio (H/HM*)	4.0	5.0	6.0
Fissile plutonium enrichment**	7.2 wt%	5.8 wt%	5.1 wt%
Number of refuelling assemblies	88	108	120
Operation cycle length	15.5 EFPM***	15.5 EFPM	15.5 EFPM
Cycle burn-up	16.5 GWd/t	19.0 GWd/t	21.3 GWd/t
Maximum assembly burn-up	52.7 GWd/t	53.5 GWd/t	53.7 GWd/t

* Atomic ratio of hydrogen to heavy metal.

** Enrichment is uniform in the assembly.

*** Effective full power months.

Figure 3 shows depletion behaviour of power distribution. Since PWR full MOX core requires no burnable absorbers such as gadolinium and applies the same enrichment for all fuel pins, peaking factors of linear heat rate (FQ) and enthalpy rise (FΔHN) are well suppressed and change smoothly with depletion. The axial power distribution of full MOX cores are slightly shifted to the bottom of the cores compared with typical UO₂ cores. The axial offset of full MOX cores is more negative than typical UO₂ cores. Such power distribution characteristics provide additional DNBR margin for the reference core and these high moderation cores.

Figure 3. Depletion behaviour of peaking factors and axial offset (PWR)

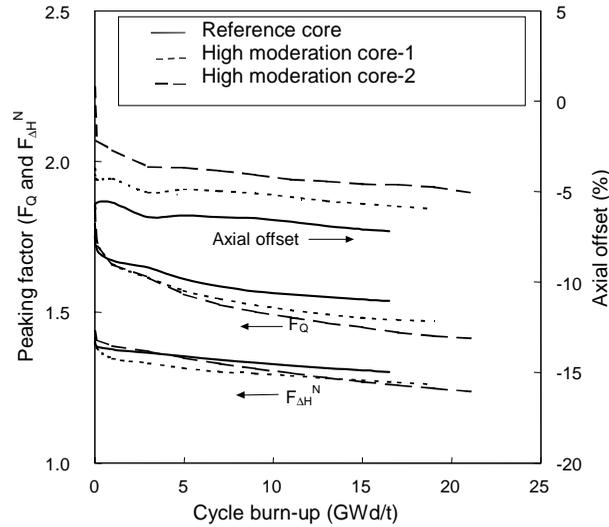
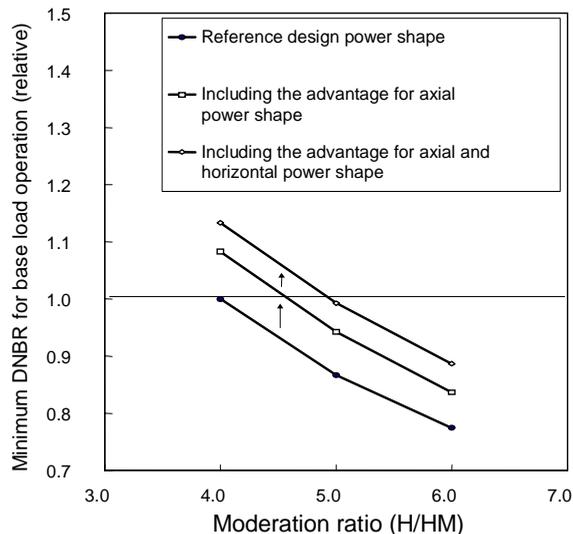


Figure 4 shows the DNBR analysis results for the highly moderated cores-1 and -2 with relaxed design power shapes considering the difference of power distributions between these cores and the typical UO_2 core. As the fuel pin diameter is decreased for high moderation ratio, the DNBR margin becomes smaller because of an increase of heat flux and a decrease of coolant flow speed caused by an increase in the channel flow area. These analyses show that such DNBR decreases can be recovered with these power distribution characteristics. The FAHN of the full MOX cores are 2-3% less than the typical UO_2 core. It provides about 5% additional margin for DNBR. The axial power distribution difference also provides about 10% additional DNBR margin. Those analyses indicate that the highly moderated core-1 (H/HM = 5) has the same DNBR margin with the typical UO_2 core, and the core-2 (H/HM = 6) has around 10% less DNBR margin. Such a reduction of the DNBR margin can be recovered by applying a fully generalised statistic thermal design method.

Figure 4. Power distribution effect on DNBR (PWR)



As for reactivity coefficients, the increase of H/HM affects the soluble boron worth. For the reference full MOX core (H/HM = 4.0), the boron worth is about 1/3 of the boron worth of UO₂ cores, but it increases about 50% to 100% with the increase of the H/HM to 5.0 (core-1) and 6.0 (core-2). This is favourable from the safety viewpoint.

The control rod worth decreases in the reference full MOX core for the harder neutron spectrum, so it is necessary to add control rods to stay at the required shutdown reactivity margin. The increase of H/HM has the beneficial effects of increasing the control rod worth and reducing the additional number of control rods. Figure 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 for core-1 and 77 for core-2, and is about 12 or 20 clusters less than that for the reference core. Using higher worth absorber such as enriched B₄C can reduce the required number of the clusters.

Figure 5. Reduction of required RCC number (PWR)

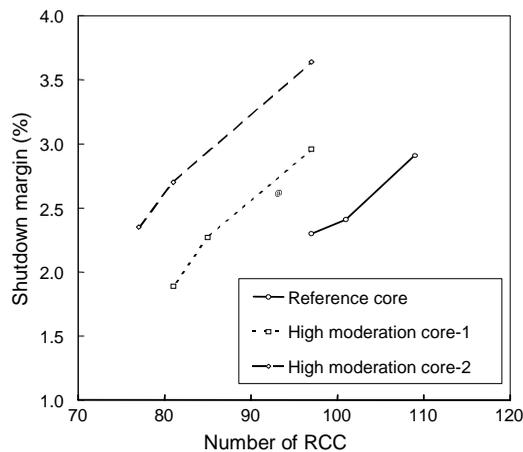


Figure 6 shows the fissile plutonium consumption rate and related parameters. With an increase in H/HM from 4.0 to 5.0 or 6.0, the consumption rate increases 24% or 47%.

Since these nuclear and thermal hydraulic characteristics of the high moderation cores are acceptable, the studied high moderation cores are feasible in PWRs and contribute to effective fissile plutonium consumption.

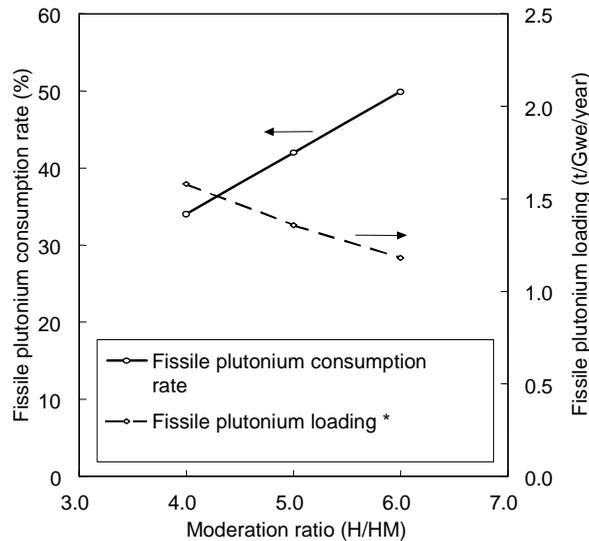
High moderation BWR

The BWR fuel design studies were done based on the 9 × 9 fuel assembly which was developed as the high burn-up “Step-III” fuel [8-9] 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, and the following results were reached:

- There is a small difference of fissile plutonium consumption rate and related parameters between these two designs for the BWR cores.
- Reducing the fuel diameter has more potential for increasing moderator ratio in terms of thermal margin, compared with replacing fuel pins by water rods.

Figure 6. Plutonium balance (PWR)

* Capacity factor is assumed to be 100%



Based on these results, the assembly with additional water rods was selected as the first design of high moderation assembly design for BWRs (highly moderated assembly-1), and the higher moderation assembly design of reducing fuel pin diameter was selected as the second design in this study (highly moderated assembly-2).

While the fissile plutonium consumption rate increases with the 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 Figure 7. To maintain adequate thermal margins, the assembly design with eight additional water rods was adopted as the highly moderated assembly-1, for which the H/HM value is 5.9, while that of the original 9×9 assembly is 4.9. The variation of MLHGR and MCPR were also evaluated with reducing the fuel pin diameter as shown in Figure 8. To satisfy thermal margins, the assembly design with a reduced fuel pin diameter from 11.2 mm to 9.8 mm was adopted as the highly moderated assembly-2, for which the H/HM value is 7.0.

The specifications of the fuel assembly are summarised in Table 4. The fissile enrichment and burnable poison design were determined to meet the discharge burn-up of 45 GWd/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 axial power peaking.

Under the condition of keeping the number and the size of fuel assemblies and control rods in ABWRs, two equilibrium core designs with each highly moderated assembly were performed. A full MOX core design with original 9×9 fuel assembly of the ABWR has been also conducted for the reference core of this advanced design. As for the reference core, the feasibility and core performance with lower discharge burn-up or different cycle have already been studied [10-11].

Figure 7. Variation of maximum linear heat generation rate (MLHGR) and minimum critical power ratio (MCPR) with moderation ratio by replacement of water rods (BWR)

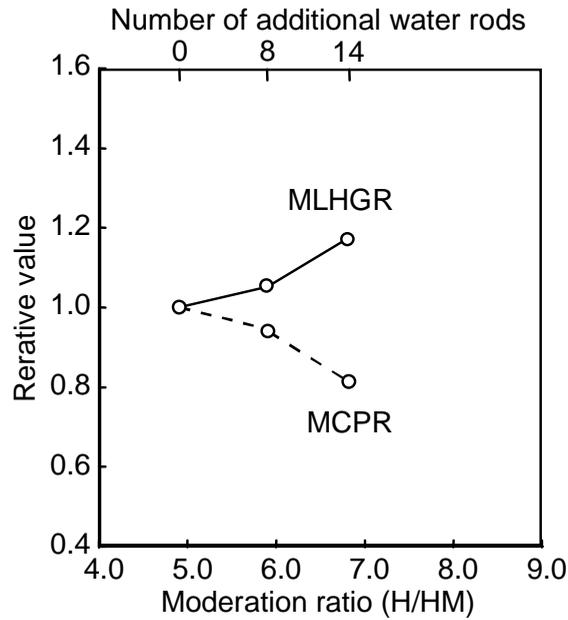


Figure 8. Variation of maximum linear heat generation rate (MLHGR) and minimum critical power ratio (MCPR) with moderation ratio by reduction of pin diameter (BWR)

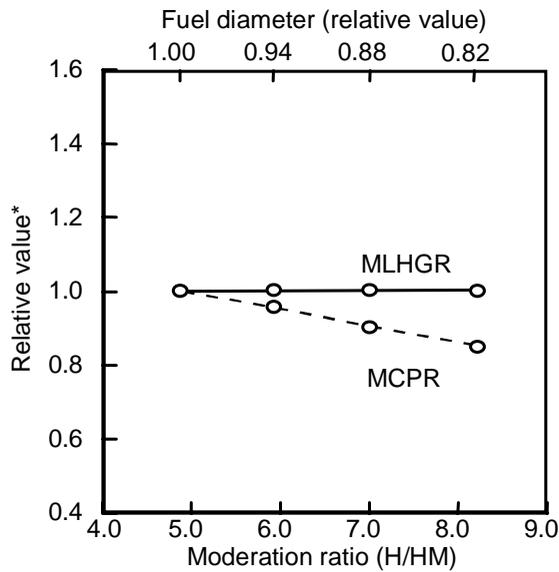


Table 4. Specifications of highly moderated MOX assembly (BWR)

	Reference 9 × 9 assembly	Highly moderated assembly-1	Highly moderated assembly-2
Moderation ratio (H/HM*)	4.9	5.9	7.0
Number of additional water rods	0	8	0
Fuel pin diameter	11.2 mm	11.2 mm	9.8 mm
Assembly lattice	9 × 9	9 × 9(same pitch)	9 × 9(same pitch)
Fissile material concentration**	5.0 wt%	4.6 wt%	4.7 wt%
MOX rod			
Assembly-averaged enrichment	3.6 wt%	3.3 wt%	3.5 wt%
Matrix	Depleted uranium	Depleted uranium	Depleted uranium
Gadolinium rod			
Gadolinium concentration	2.5/1.5 wt%***	3.0/2.0 wt%***	3.3/2.3 wt%***
Matrix	Enriched uranium	Enriched uranium	Enriched uranium

* Atomic ratio of hydrogen to heavy metal.

** Assembly-averaged concentration of ($^{235}\text{U} + ^{239}\text{Pu} + ^{241}\text{Pu}$).

*** Axial zoning of gadolinium concentration (lower/upper).

The specifications of the high moderation core are shown in Table 5. The fissile plutonium enrichment for the high moderation cores-1 and -2 can be reduced to 3.3 and 3.5 wt% from 3.6 wt% for the reference core, respectively. The number of reload fuel assemblies per an operational cycle increases for high moderation cores due to less fuel inventory. The average discharge burn-up is set to 45 GWd/t for all cores. The maximum assembly burn-up for all cores is less than 55 GWd/t, which is the same criteria for “Step-III” UO₂ fuel. The nuclear and thermal hydraulic performances were evaluated with a three-dimensional nuclear thermal hydraulic coupled core simulator. The calculated results on the cold shutdown margin (CSDM) are shown in Figure 9. The CSDM values for all MOX cores satisfy the design criteria without any modification of the control rods in the ABWR. The high moderation core-2 improves the CSDM about 2% Δk by increasing H/HM from 4.9 to 7.0.

Table 5. High moderation core specifications (BWR)

	Reference core	High moderation core-1	High moderation core-2
Moderation ratio (H/HM*)	4.9	5.9	7.0
Fissile plutonium enrichment**	3.6 wt%	3.3 wt%	3.5 wt%
Number of refuelling assemblies	232	264	300
Operation cycle length	15 EFPM***	15 EFPM	15 EFPM
Cycle burn-up	12.0 GWd/t	13.6 GWd/t	15.5 GWd/t
Average discharge burn-up	45.1 GWd/t	44.9 GWd/t	45.1 GWd/t
Maximum assembly burn-up	49.7 GWd/t	47.3 GWd/t	52.5 GWd/t

* Atomic ratio of hydrogen to heavy metal.

** Assembly-averaged initial enrichment.

*** Effective full power months.

The evaluated values of MLHGR and MCPR during the equilibrium cycle are presented in Figures 10 and 11. They show the satisfactory results comparing to the operational limits. The relative differences of MLHGR and MCPR between the high moderation core-1 and the reference core are

Figure 9. Cold shutdown margin of high moderation core (BWR)

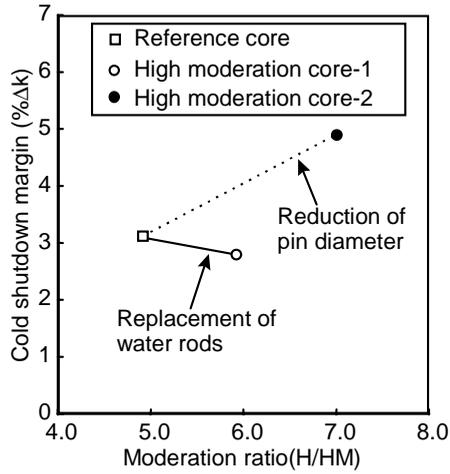


Figure 10. Maximum linear heat generation rate (MLHGR) of high moderation core (BWR)

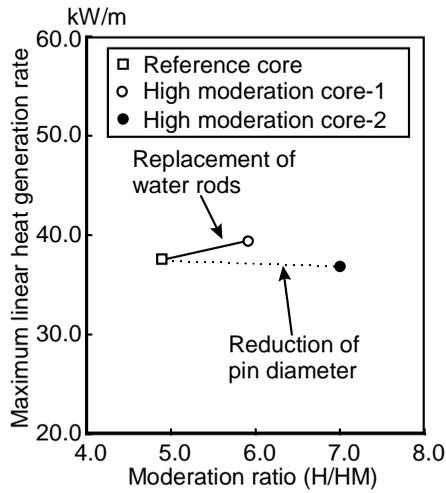
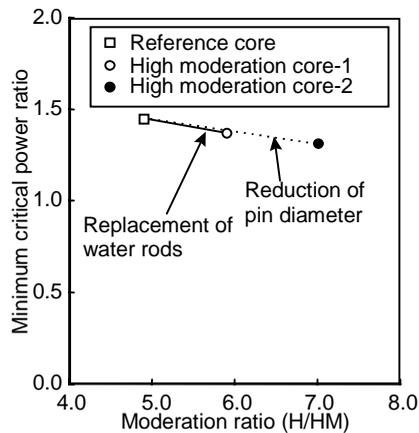


Figure 11. Minimum critical power ratio (MCPR) of high moderation core (BWR)



about 5%. And the MCPR decreases by about 10% by increasing H/HM 4.9 to 7.0. The loaded fissile plutonium and the fissile plutonium consumption rate for all MOX cores were evaluated and the results are presented in Figures 12 and 13. By increasing H/HM from 4.9 to 5.9 and 7.0, the consumption rate increases by about 17% and 22%, respectively.

Since these nuclear and thermal hydraulic characteristics of both high moderation cores are acceptable, the studied high moderation cores are verified to be feasible in the ABWRs and contribute to effective fissile plutonium consumption.

Figure 12. Fissile plutonium loading of high moderation core (BWR)

** Capacity factor is assumed to be 100%*

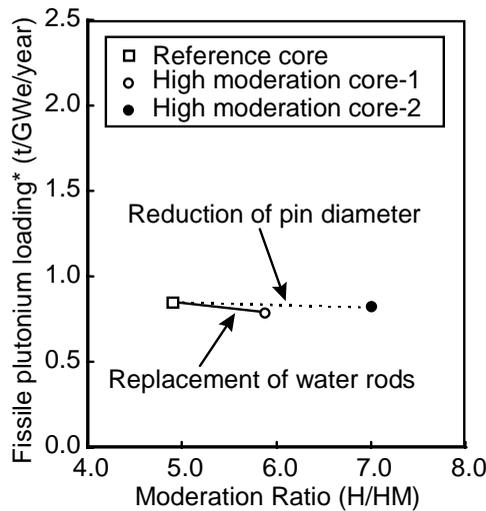
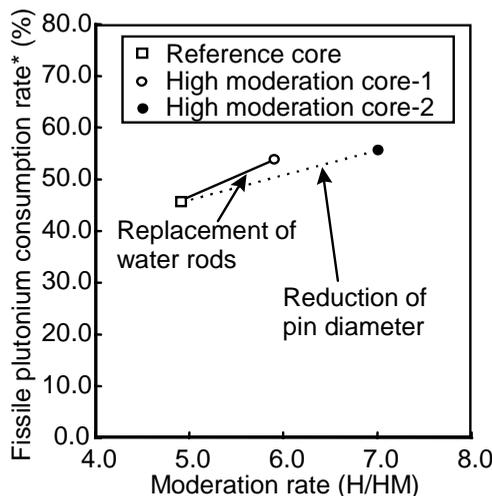


Figure 13. Fissile plutonium consumption rate of high moderation core (BWR)

** Capacity factor is assumed to be 100%*



Conclusion

Studies have been performed to develop advanced technologies for full MOX cores with high Pu consumption rates based on the advanced LWRs. To this aim, basic core designs of high moderation (H/HMs equal to about 5 and 6) have been carried for APWRs with reduced fuel diameters in fuel assemblies and those of high moderation (H/HMs equal to about 6 and 7) for ABWRs with the addition of extra water rods and reduced fuel diameters in fuel assemblies, respectively. The analysis of equilibrium cores shows that nuclear and thermal hydraulic parameters satisfy the design criteria and higher Pu consumption rates are obtained by increasing the moderation ratio.

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INVESTIGATION OF THE FUEL TEMPERATURE COEFFICIENT OF INNOVATIVE FUEL TYPES

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Abstract

The fuel temperature coefficient has been calculated for several fuel types designed for the transmutation of plutonium, ranging from standard (U, Pu) MOX fuel to innovative fuel types based on plutonium oxide mixed in an inert matrix. For all fuel types, the fuel temperature coefficient is large in magnitude and negative. For fuels containing uranium or thorium oxide, a large contribution to the FTC is due to the ^{238}U or ^{232}Th (about -2.2 pcm/K). In addition, quite a large contribution is present due to the ^{240}Pu (about -1 pcm/K), while the contribution due to ^{242}Pu is not larger than 0.3 pcm/K in magnitude. When the moderator-to-fuel volume ratio of (U, Pu) MOX fuel is enlarged from 2 to 4, the FTC decreases in magnitude due to a better thermalisation of the neutron spectrum and a correspondingly larger resonance escape probability.

When the fuel matrix is made of a neutron inert material, only the plutonium isotopes contribute to the FTC. For reactor grade plutonium, ^{240}Pu contributes about -1 pcm/k, while ^{242}Pu contributes -0.3 pcm/K. The odd fissile plutonium isotopes give either a positive or negative contribution to the FTC, dependent on the composition of the matrix. In case of weapons grade plutonium mixed in an inert matrix, the FTC ranges up to -1 pcm/K. Further research should assess the feasibility of such fuels based on thermal considerations.

Introduction

The burning of actinides is most effectively done in a fuel containing actinide elements mixed in an inert matrix. Under such conditions, no new plutonium and minor actinides are produced by neutron capture reactions in ^{238}U , as is the case in standard UO_2 fuel. Although fuels based on inert matrices are most promising in this respect, the characterisation of the inert materials will take a long time. Therefore, other innovative fuels are proposed for the burning of plutonium and minor actinides. Examples are fuels containing thorium oxide as a matrix material, a well known material with very good thermal properties, or standard MOX fuel (plutonium oxide mixed in uranium oxide) with an enhanced moderator-to-fuel ratio to limit the resonance neutron capture rate by the ^{238}U in the fuel. Of all these fuels, the material composition is different from that of standard UO_2 fuel, which will have a direct effect on the fuel temperature coefficient (FTC).

In this paper, the FTC is calculated as a function of burn-up for several fuel types which were proposed in the past to be used for the incineration of plutonium. To serve as a reference, calculations on standard UO_2 fuel have also been performed. The following innovative fuels were investigated:

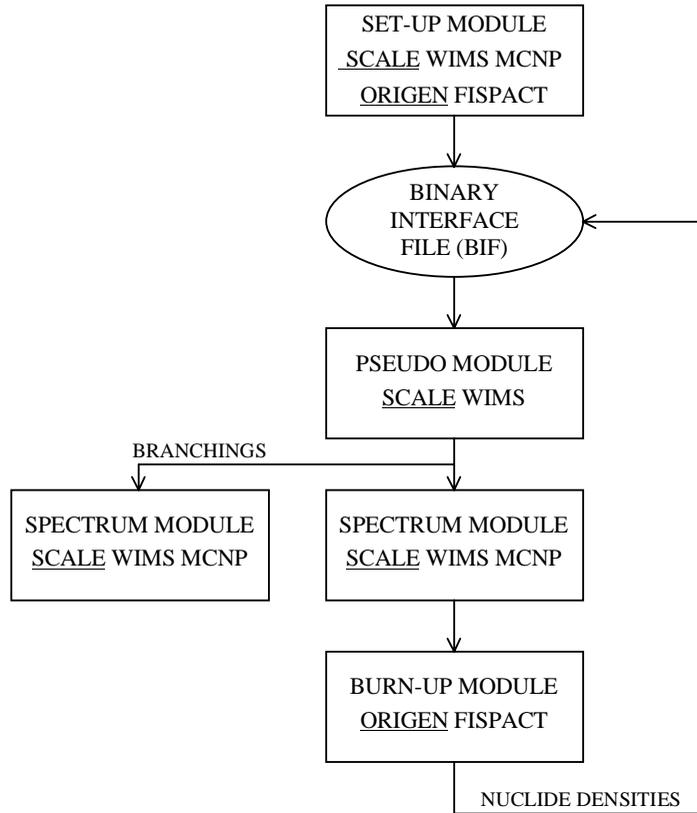
- (U, Pu) MOX fuel with a moderator-to-fuel ratio (MF) of 2 (Pu density 0.9 g/cm^3).
- (U, Pu) MOX fuel with a moderator-to-fuel ratio (MF) of 4 (Pu density 0.5 g/cm^3).
- Reactor grade (RG) Pu oxide mixed in an inert matrix (Pu density 0.5 g/cm^3).
- Weapons grade (WG) Pu oxide mixed in an inert matrix (Pu density 0.5 g/cm^3).
- Reactor grade (RG) Pu oxide mixed in an inert matrix with ^{10}B (Pu density 0.7 g/cm^3).
- Reactor grade (RG) Pu oxide mixed in an inert matrix with Er (Pu density 0.7 g/cm^3).
- Reactor grade (RG) Pu oxide mixed in thorium oxide (Pu density 0.6 g/cm^3).

Calculations

All burn-up calculations have been performed with the OCTOPUS burn-up and criticality code system [1]. This system is schematically shown in Figure 1. By means of the SCALE package, a spectrum calculation is performed and the multi-group resonance-shielded cross-sections are passed to the multi-group EAF library containing cross-sections for over 700 nuclides at infinite dilution with the same energy group structure. The calculated neutron spectrum is used to collapse the cross-sections of all these nuclides to one-group ones, which are subsequently passed to the burn-up code. After a burn-up step has been performed, this sequence of coupled spectrum and burn-up calculations is repeated. Information from one step to the other is transferred via a so-called binary interface file (BIF), containing all nuclide identifiers, densities and cross-sections for all burnable zones in the system. During the burn-up sequence, branching calculations can be performed with one or more input parameters changed. These branchings can be used to calculate reactivity coefficients and kinetic parameters as a function of burn-up. At each branching an adjoint spectrum calculation can be performed to calculate the contributions of individual nuclides to the FTC by means of first-order perturbation theory as applied by the VAREX code [2].

Figure 1. The OCTOPUS burn-up and criticality code system

The underlined codes are used for the calculations described in this paper



For the calculations on the UO_2 fuel and on the MOX fuels, the base design is a French N4 PWR with a moderator-to-fuel volume ratio of 2. An extensive description of all parameters used in the calculations is given in Ref. [3]. Here, it will be explained how the plutonium density in the fuel was determined. First a burn-up calculation was performed on UO_2 fuel with a fuel enrichment of 4.0%. From core calculations it is known that such fuel can reach a burn-up of 47.5 MWd/kgU. Knowing that the reactor runs in a five-batch mode and applying the linear reactivity model, the reactivity at the end of the third fuel batch equals that of the equilibrium core at the end of cycle. The k_∞ at the end of the third fuel batch equals 1.057. This value was used in the procedure to determine the plutonium density in the MOX fuel such that the k_∞ at the end of the third fuel batch with MOX fuel also equals 1.057. The boron concentration was determined such that the reactivity loss during the third fuel batch equals zero, implying that the linearly decreasing boron concentration compensates for the fuel depletion and the fission product build-up. Because the boron concentration and plutonium density depend on each other, these two parameters had to be determined iteratively.

For the calculations based on inert matrices and thorium oxide, the fuel design is based on the inert matrix fuel benchmark defined by PSI [4].

According to first-order perturbation theory and the following definition of reactivity change:

$$\delta\rho = \frac{k' - k}{k'k} \quad (1)$$

where k is the multiplication factor and k' the multiplication factor of the perturbed system. The reactivity change according to a perturbation of the cross-section reads [5]:

$$\delta\rho = \frac{\left\langle \Phi^*, \frac{1}{k} \delta F \Phi \right\rangle - \left\langle \Phi^*, \delta M \Phi \right\rangle}{\left\langle \Phi^*, F \Phi \right\rangle} \quad (2)$$

where Φ^* is the adjoint function, Φ the neutron flux, F the fission operator, M the transport operator, and δF and δM the changes of the fission and transport operators due to changes of the fission and absorption cross-sections. The latter changes are due to an increase of the fuel temperature.

Because the calculation of the fuel temperature coefficient is done by pin-cell spectrum calculations on an infinite lattice, the reactivity change can better be calculated by [6]:

$$\delta\rho = \frac{k' - k}{k} \quad (3)$$

Using this definition of reactivity change, Eq. (2) becomes:

$$\delta\rho = \frac{\left\langle \Phi^*, \frac{1}{k} \delta F \Phi \right\rangle - \left\langle \Phi^*, \delta A \Phi \right\rangle}{\left\langle \Phi^*, A \Phi \right\rangle} = \frac{1}{k} \frac{\left\langle \Phi^*, \delta F \Phi \right\rangle}{\left\langle \Phi^*, A \Phi \right\rangle} - \frac{\left\langle \Phi^*, \delta A \Phi \right\rangle}{\left\langle \Phi^*, A \Phi \right\rangle} \quad (4)$$

where the transport operator in the numerator has been replaced by the absorption operator, implying that there is no change in neutron leakage. Whether a nuclide gives a positive or negative contribution to the FTC depends on the change of the fission cross-section relative to that of the absorption cross-section. A nuclide with strong neutron capture resonances and no fission cross-section resonances will give a negative contribution to the FTC. However, fissile nuclides with both fission and capture resonances can give a positive or negative contribution to the FTC, dependent on the ratio between these two and on the total absorption rate in the system. For example, when a nuclide dominates the fission rate but not the absorption rate, the first term in Eq. (4) will dominate, which means that the nuclide gives a net positive contribution to the FTC.

Results for UO₂ fuel

In this paragraph, the results for standard UO₂ fuel are given as a reference. The FTC is shown in Figure 2 as function of burn-up, while the contributions of the individual nuclides are shown in Figure 3. The FTC increases as function of burn-up due to the contribution of ²⁴⁰Pu, which increases from zero to about -0.7 pcm/K at a burn-up of 47.5 MWd/kg. The contribution due to ²³⁸U remains constant during burn-up at a value of about -2.5 pcm/K. The irregular behaviour of the FTC during burn-up (see Figure 2) is caused by the variable boron concentration, which decreases linearly from about 1 000 ppm at the beginning of cycle until zero at the end.

Results for (U, Pu) MOX fuel

The results for MOX fuel with the same moderator-to-fuel volume ratio (MF = 2) are given in Figures 4 and 5. In an absolute sense, the FTC of MOX fuel is larger than that of UO₂ fuel with the

Figure 2. The FTC as a function of burn-up for UO₂ fuel

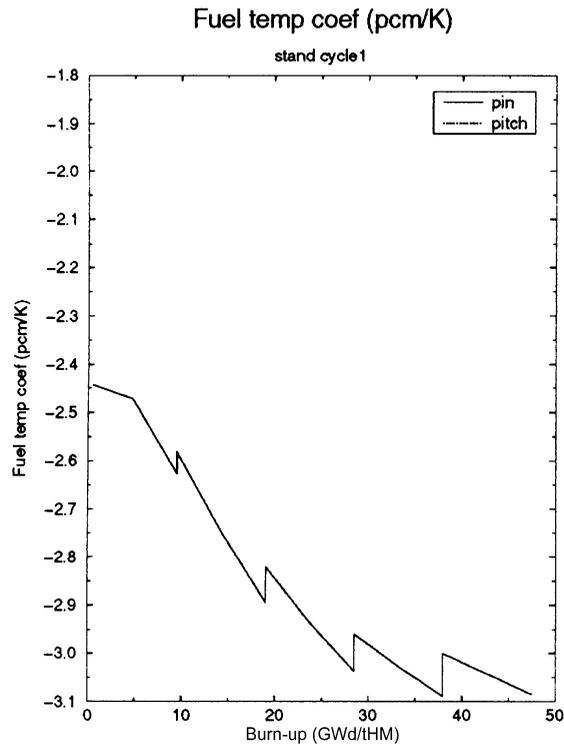


Figure 3. The contributions of individual nuclides to the FTC of UO₂ fuel

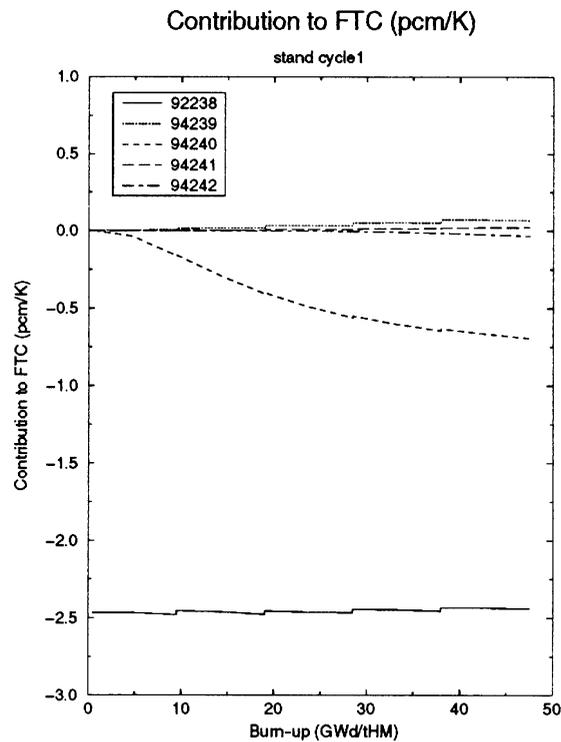


Figure 4. The FTC as a function of burn-up for MOX fuel with MF = 2

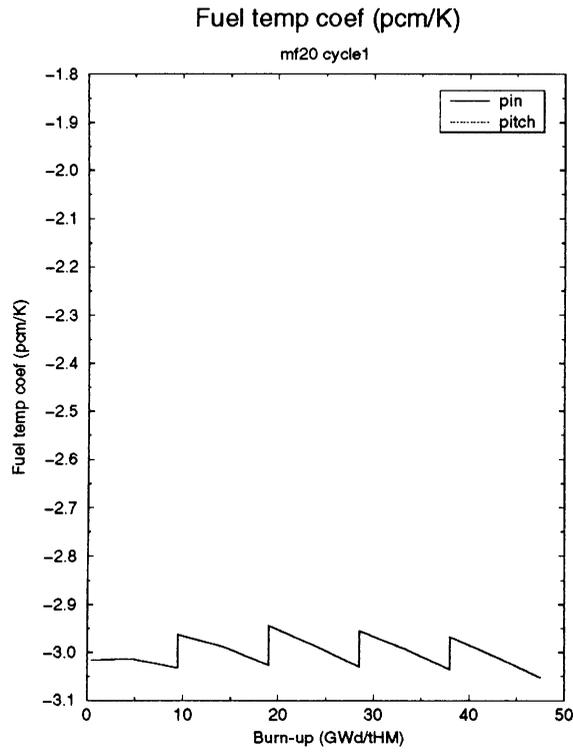
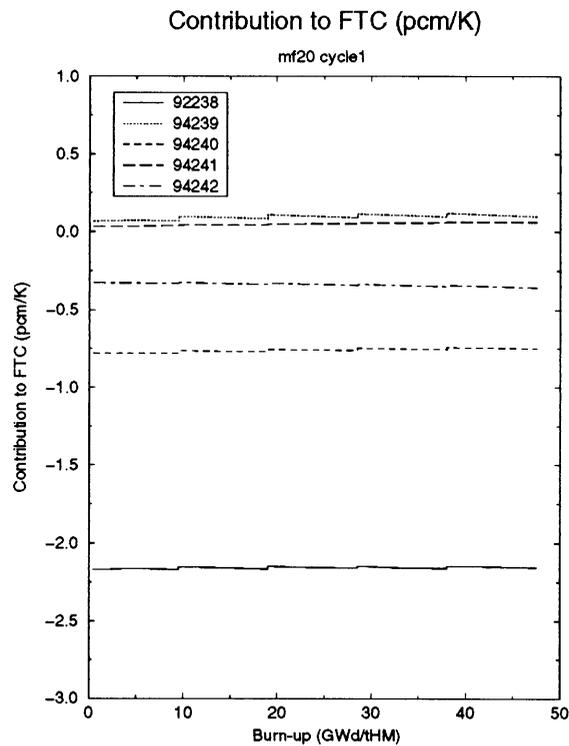


Figure 5. The contributions of individual nuclides to the FTC of MOX fuel with MF = 2



same MF ratio, which is mainly due to the extra contribution of the even plutonium isotopes. This is also seen in Table 1, which shows the contributions of the individual isotopes at BOL and at EOL. The contribution of ^{238}U is somewhat less in MOX fuel than in UO_2 , but this is compensated by the larger contributions of the ^{240}Pu and ^{242}Pu isotopes.

Table 1. The contributions of the individual isotopes to the fuel temperature coefficient (pcm/K)

The MF ratio for these fuel types equals 2

Isotope	Fuel type	BOL	EOL
^{238}U	UO_2	-2.5	-2.5
^{240}Pu	UO_2	0	-0.70
^{238}U	MOX	-2.2	-2.2
^{240}Pu	MOX	-0.75	-0.75
^{242}Pu	MOX	-0.30	-0.35

When the moderator-to-fuel ratio increases, the resonance absorption rate decreases. In Figures 6 and 7, the four factors of the well-known four-factor formula are shown as a function of burn-up. The fast fission factor ϵ decreases from about 1.8 for MOX fuel with MF = 2 to 1.2 for MOX fuel with MF = 4. This is compensated mainly by an increase of the resonance escape probability p from about 0.4 to 0.65. The thermal utilisation f and the number of neutrons released per thermal absorption in the fuel η do not depend much on the MF ratio. At the end of each fuel batch (burn-up interval of 9.5 MWd/kg), the thermal utilisation tends to unity due to the linearly decreasing boron concentration, indicating that at the end of each fuel batch, the parasitic neutron absorption by the soluble boron and cladding region is virtually zero.

Figure 6. The four factors as a function of burn-up for MOX fuel with MF = 2

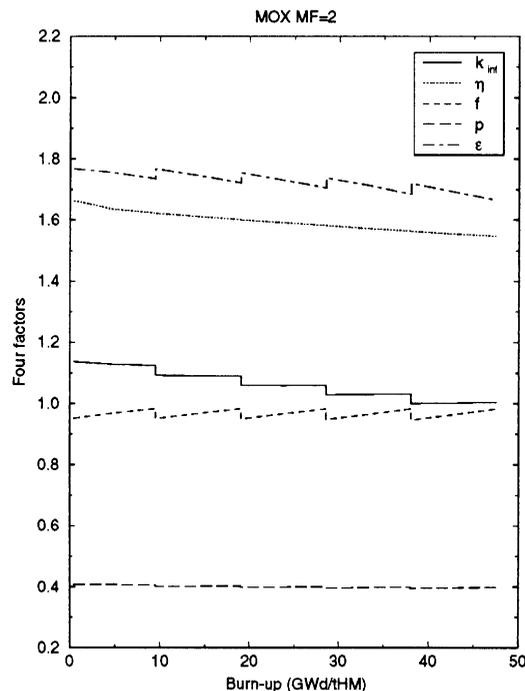
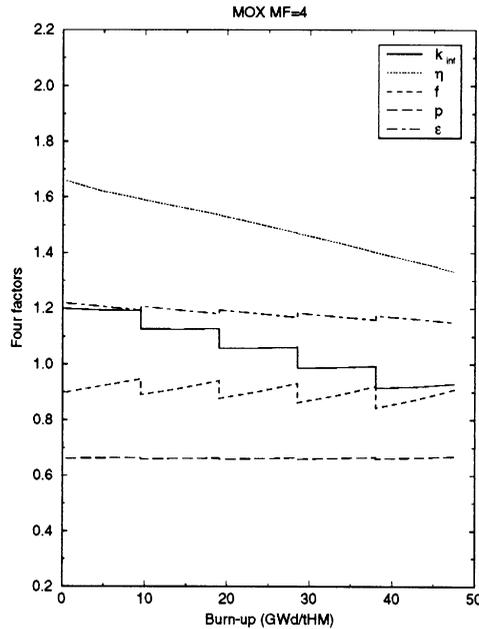


Figure 7. The four factors as a function of burn-up for MOX fuel with MF = 4



Figures 8 and 9 show the FTC and the contributions of the individual nuclides to the FTC as a function of burn-up for the MOX fuel with MF = 4. Reducing the fuel pin radius gives a larger FTC in magnitude than increasing the fuel pitch. This is in agreement with literature [7], where it has been reported that the maximal Doppler effect due to ^{238}U occurs at a Dancoff corrected mean chord length of about 0.05 cm, while this value equals about 1.2 cm in practice. (For a fuel pin, the Dancoff corrected mean chord length equals $2R/(1-C)$, where R is the fuel pin radius and C the Dancoff correction factor.)

Figure 8. The FTC as a function of burn-up for MOX fuel with MF = 4

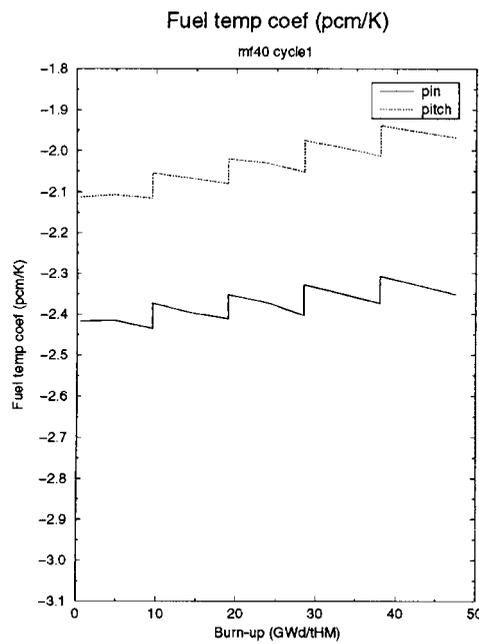
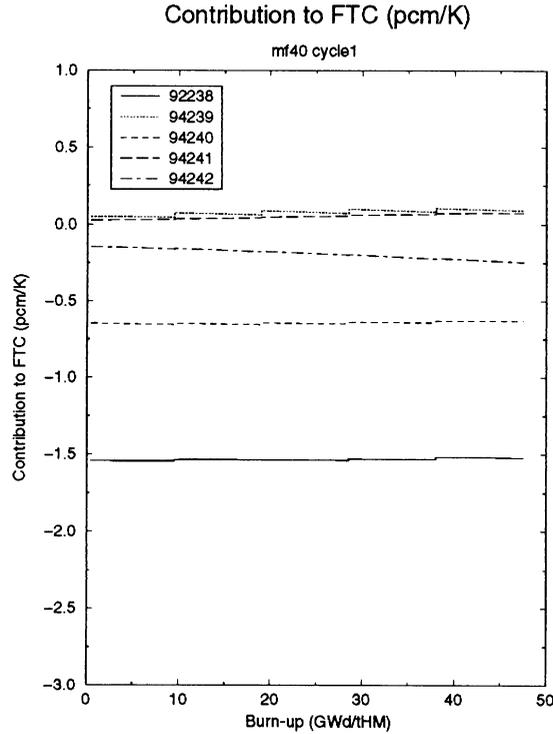


Figure 9. The contributions of individual nuclides to the FTC of MOX fuel with MF = 4



Results for inert matrix fuel

Several inert matrix fuels have been investigated:

- Reactor grade (RG) Pu mixed in an inert matrix (Al₂O₃-ZrO₂-MgO).
- Weapons grade (WG) Pu mixed in an inert matrix (Al₂O₃-ZrO₂-MgO).
- Reactor grade (RG) Pu mixed in an inert matrix with ¹⁰B as burnable poison (BP).
- Reactor grade (RG) Pu mixed in an inert matrix with Er as burnable poison.

In the first two cases, the plutonium density equals 0.5 g/cm³, while for the other two the plutonium density equals 0.7 g/cm³ to compensate for the extra neutron absorption by the matrix. More details about the composition of the fuels can be found in Ref. [4].

The FTC averaged over the fuel irradiation time up to 1 000 days are given in Table 2. Clearly, the difference between RG and WG plutonium is very small. In the first case, the FTC equals -1.2 pcm/K, in the second -1.05 pcm/K. This is due to the rather strong contribution of the ²⁴⁰Pu in both cases, as can be seen in Figures 10 and 11.

The k_{∞} as function of burn-up for the two fuels containing burnable poison (BP) are shown in Figure 12. For the fuel with erbium as BP, k_{∞} is almost constant up to about 750 irradiation days. For the fuel containing ¹⁰B, k_{∞} increases up to 500 irradiation days due to the fast depletion of the BP.

Table 2. The FTC of the IMF averaged over 1 000 irradiation days

Fuel type	Pu density (g/cm ³)	FTC (pcm/K)
RG Pu in inert matrix	0.5	-1.2
WG Pu in inert matrix	0.5	-1.05
RG Pu in inert matrix and ¹⁰ B	0.7	-1.0
RG Pu in inert matrix and Er	0.7	-1.3

Figure 10. The contributions of individual nuclides to the FTC of RG Pu mixed in IMF

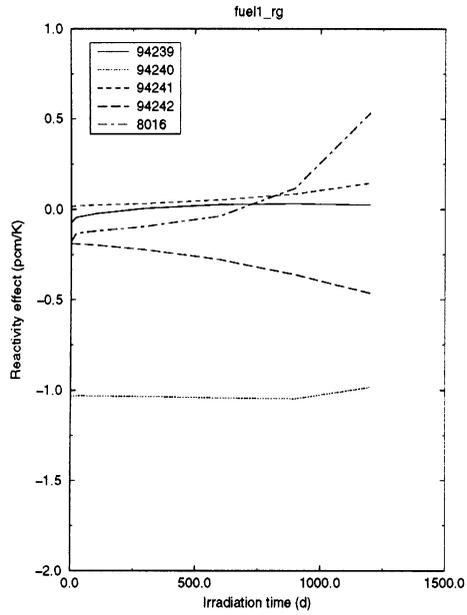


Figure 11. The contributions of individual nuclides to the FTC of WG Pu mixed in IMF

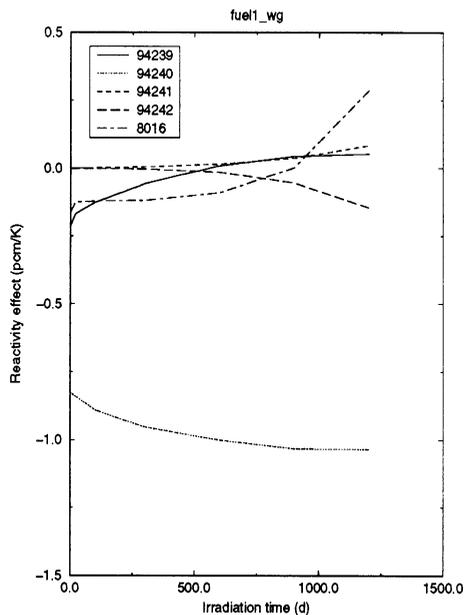
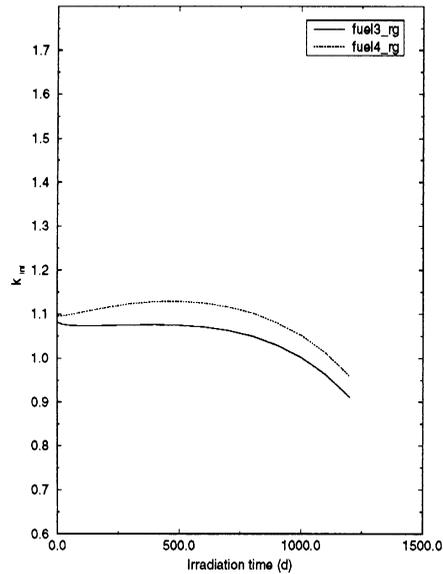


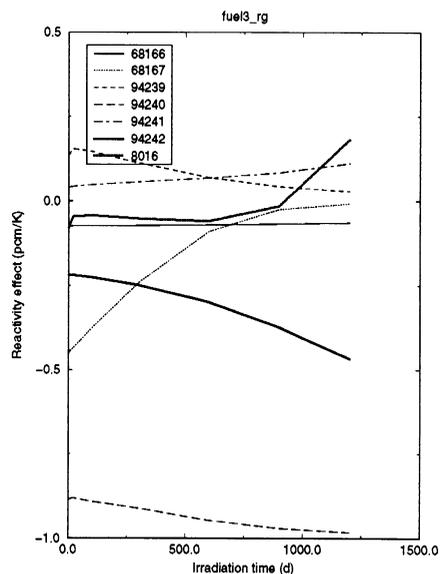
Figure 12. The k_{∞} as a function of burn-up for the inert matrix fuels with burnable poison

Fuel3_rg is the fuel with Er, fuel4_rg with ^{10}B



The erbium isotopes contribute to the FTC mainly due to the isotope ^{167}Er and to a lesser extent due to ^{166}Er . Note that erbium contains more isotopes, but that only ^{166}Er and ^{167}Er have nuclear data available in the JEF-2.2 data library. The contributions of the individual isotopes can be seen in Figure 13. The contribution of ^{167}Er equals about -0.4 pcm/K at BOL and virtually zero after about 600 days of irradiation. The contribution of ^{166}Er equals about -0.1 pcm/K during the whole irradiation. This is due to the much lower neutron capture integral of ^{166}Er (140 barns) compared with that of ^{167}Er (3 000 barns). During burn-up, the nuclear density of ^{166}Er decreases only with 13%, while that of ^{167}Er decreases from $9.8\text{E-}5$ to $1.1\text{E-}5$ barn $^{-1}\text{cm}^{-1}$. Due to the strong absorption by the BP, the contributions of the fissile plutonium isotopes ^{239}Pu and ^{241}Pu are positive.

Figure 13. The contributions of individual nuclides to the FTC in the IMF fuel with Er as BP



Results of (Th, Pu) MOX fuel

The last fuel investigated is the advanced MOX fuel containing plutonium oxide mixed in thorium oxide with a plutonium density of 0.6 g/cm³. Thorium is a fertile nuclide like ²³⁸U with an effective resonance integral comparable with that of ²³⁸U. Using the empirical relationship for the resonance integral of a cylindrical uranium-oxide fuel pin [5]:

$$I_{\gamma} = 4.15 + 26.6 \sqrt{\frac{S}{M}} \quad (5)$$

and for the resonance integral of a cylindrical thorium-oxide fuel pin [8]:

$$I_{\gamma} = -3.6 + 37.6 \sqrt{\frac{S}{M}} \quad (5)$$

where S is the surface of the fuel rod and M the associated mass, we find a value of about 23 barn for both fuel types with S/M ratio of about 0.5. In Table 3, the contributions of the individual nuclides to the FTC are given. Note that these values are very close to the ones of standard MOX fuel with MF = 2. Despite the fact that ²³²Th is fertile, a considerable amount of plutonium has been fissioned. The plutonium density in the fuel reduces from 0.6 to 0.16 g/cm³, which compares with the amount of plutonium transmuted in the inert matrix fuel without burnable poison, where the plutonium density decreases from 0.5 to 0.125 g/cm³. However, after 1 200 irradiation days, the amount of uranium (²³³U + ²³⁴U) produced equals about 0.08 g/cm³, which is as hazardous as plutonium. This effectively reduces the transmutation capabilities of fuels based on a thorium oxide matrix.

Table 3. The contributions to the FTC of plutonium oxide mixed in thorium oxide

Nuclide	Contribution to FTC (pcm/K)
²³² Th	-2.1
²⁴⁰ Pu	-1.0
²⁴² Pu	-0.2

Conclusions

The fuel temperature coefficient (FTC) has been calculated for several fuel types containing RG plutonium and for one fuel type containing WG plutonium. The fuels range from standard MOX fuel to innovative fuels based on inert matrices. For all of these fuels, the FTC is strongly negative. In the (U, Pu) and (Th, Pu) MOX fuels, the isotopes ²³⁸U or ²³²Th give about the same contribution (-2.2 pcm/K), which is only slightly less than the contribution of ²³⁸U in UO₂ fuel (-2.5 pcm/K). In the MOX fuels, there are additional contributions due to the even plutonium isotopes, which are at beginning of life lacking in the UO₂ fuel.

The inert matrix fuels have a much smaller FTC due to the missing contribution of ²³⁸U or ²³²Th. However, in all cases the FTC is larger than 1 pcm/k in magnitude, which can be sufficiently large. Surprisingly, this also holds for the case of weapons grade plutonium mixed in an inert matrix, and also for long irradiation times (> 1 000 days).

The odd fissile plutonium isotopes give a negative contribution to the FTC in case these isotopes dominate both the neutron capture rate and the fission rate. If they dominate only the fission rate, their contribution is positive (compare for example Figures 10 and 13). In many cases, the isotope ^{16}O gives a positive contribution to the FTC after long irradiation times when the neutron spectrum has considerably softened compared to the spectrum at beginning of life. This effect is due to upscattering.

At this stage it is difficult to say whether a FTC of -1 pcm/K for the inert matrix fuels is sufficient for safe operation or not. An extension of this work could be to perform thermal calculations to determine the fuel centreline temperature of the rods during normal operation and during accident situations, given the thermal conductivity and the heat capacity of the fuel material. Then the temperature margin to melting can be determined, which is a decisive parameter for the assessment of the feasibility of the fuel.

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URANIUM-FREE REACTORS

Chair: R.J.M. Konings

U-FREE Pu FUELS FOR LWRs – THE CEA/DRN STRATEGY

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Abstract

This paper presents the advancement of work at the CEA/DRN on inert matrices intended for PWR plutonium fuels. After an introduction reviewing the work programme in the French context, a summary is made of various aspects concerning materials, fuel rods, and assemblies. Research orientations mainly concern CERCER and CERMET as regards materials, whereas with respect to fuel rods, neutron studies have permitted selecting a spinel and a reference metal. The acquisitions of the TANOX studies and experiments have contributed to defining a CERMET as a potential candidate and delimiting accident studies for this cold fuel concept. A specific assembly has been proposed to allow using composite fuels in PWRs while retaining kinetic coefficients at acceptable values.

Introduction

In view of the fact that, a few years ago, France decided to pursue a fuel reprocessing policy, COGEMA, the French fuel cycle operator, committed itself to reprocessing (at La Hague, Marcoule, etc.). This has permitted the supply of quality MOX fuel that can be used at present in 16 French plant reactors (12 other authorisations granted by the safety authorities are pending). For reasons linked to core physics and neutronics, core loading is limited to 30% MOX.

The policy of the French utility, EDF, is to only reprocess the quantity of fuel required to supply the amount of plutonium it is permitted to use in its reactors.

It is obvious that the plutonium stream produced by 55 reactors and currently used in 16 reactors for 30% of their cores is far from balanced, and that a considerable stock of plutonium is accumulating in various storage vessels.

If general policy turns towards:

- Optimised utilisation of natural resources.
- A reduction of the volume, quantity and radiotoxicity of wastes.
- Increased resistance to proliferation.

And if, moreover, one reflects that the time required to effectively perfect a new fuel, which entails experimental irradiation tests, is about 20 years, it is urgent to envisage new programmes now, with the aim of defining the technical characteristics of the solutions considered with respect to objectives and the operating specifications of these new fuels.

One of the objectives (among others) which could be set in order to find a solution to the plutonium stream – while respecting the three criteria mentioned above – is to increase (100%) the fraction of plutonium fuel used in the reactors, while increasing cycle duration and, of course, the number of reactors using this fuel.

Studies have been initiated along these lines [1,2] with a view to defining cores with increased moderation so as to enhance the efficiency of the control systems and to reduce, in absolute values, the importance of the moderator coefficient, thus making a cold shock accident less deleterious.

However, this solution entails making significant changes to the reactor core and internals, while decreasing the volume power of the core and permitting 100% MOX loading.

Another line of study consists in considering, in addition to the generic specifications described above, that only minor modifications to the core are to be permissible; research on three essential aspects would then remain possible:

- The materials composing the fuel.
- The fuel itself, including the rod.
- The assemblies.

To this list should be added a fourth element, that of the core, together with its management strategy (or strategies) and associated drive system.

In this paper we shall review these three classifications. The core aspect will be addressed later, when the first three aspects will have been optimised with respect to a core-fuel strategy.

Fuel materials

The idea directing research on materials is that, in order to reduce, or even balance the plutonium stream, while increasing its consumption, $^{238}\text{U} \rightarrow ^{239}\text{Pu}$ conversion has to be reduced, hence the notion of U-free Pu. To achieve this, a matrix has therefore to be defined which will contain the fissile material and which should have at least the physico-chemical characteristics of UO_2 , or better if possible. A non-negligible aspect for the physical behaviour of the core is that, for a constant geometry, the moderation ratio would be modified towards better control. The physical moderation ratio being defined by the ratio of the number of moderator atoms to the number of “heavy” atoms of the fuel, it is obvious that in the case of a 50% matrix, 50% fuel composite, the moderation ratio is practically doubled (to within a density correction factor), which theoretically brings the moderation ratio of a standard 17×17 assembly to 4. This is not quite exact insofar as the decrease in capture (less ^{238}U) cannot be directly transposed to an increase in moderation capacity (more water), but does, nonetheless, very significantly increase the effective moderation ratio.

Three elements have been selected for the materials:

- *Ceramic-Ceramic “CERCER”*. The fuel in the form of spheres of ceramic is closely bonded to a ceramic which permits filling the fuel rod. The matrix meets very precise “a minima” specifications, those of UO_2 .
- *Ceramic-Metal “CERMET”*. The ceramic fuel is bonded to a metallic matrix.
- *Solid Solution (Solid Solution Pellet, SSP)*. The fuel and the matrix form a solid solution in order to obtain very precise characteristics for highly specific objectives (storage, for example).

CERCER

Besides work on actinide incineration, targets and fuel for CAPRA-type reactors, which is being performed by the CEA in the framework of other programmes, CERCER composites have been developed for pressurised water reactors.

A preliminary analysis resulted in the elimination of MgO , which disintegrates under PWR conditions in the event of cladding failure. A 40% volume spinel (MgAl_2O_4) matrix was finally selected using fuel spheres some $120 \mu\text{m}$ in diameter. The spheres are pre-cracked for better resistance under irradiation and a specific patented technique involves introducing a gap between the matrix and the sphere during sintering so as to accommodate fission gases without damaging the matrix. Studies show that viscoplasticity is significantly improved, thus reducing the risk of pellet-cladding interaction (PCI) [3,4]; in addition, thermal conductivity is slightly but nonetheless improved.

CERMET

Neutron analyses [5,6,7] resulted in the definition of three classes of metals: absorbent (stainless steel, Mo, etc.), intermediary (Inconel, etc.) and transparent (Zr, silumin, etc.). It is obvious that using metals which are transparent with respect to neutrons leads to greater savings in fissile material, but that absorbent metals permit maximising fissile material loading, and therefore greater in-vessel plutonium storage (+20% for an equal amount of produced energy compared to a transparent metal), which is an interesting factor from the standpoint of strategies aiming to reduce surface stocks or of resistance to proliferation.

The advantages [8,9] of this type of fuel lie in the fact that the metallic matrix is a potential supplementary barrier to fission gas releases and ensures excellent thermal conductivity, as well as forming a metal to metal contact for the cladding and matrix, which makes PCI highly improbable.

SSP

Various lines of research are oriented towards a yttrium-aluminum-garnet (YAG) configuration with a view to obtaining a “rock-like” matrix, which would be optimal for ultimate deep geological formation storage. In order to be economically advantageous, this solution must enable high burn-up so that the final fissile nuclei inventory is as low as possible.

However, the poor thermal conductivity of these materials is liable to create heat removal difficulties. This particular field of study is being investigated by a number of our partners, such as the JAERI, PSI, ENEA, etc. At the CEA, the technological development of these concepts is actively monitored, together with the understanding of the physics and neutronics of these materials.

Fuels and rods

The materials being generally defined, the fuel has to meet neutronic as well as in-core behaviour criteria. The interest of a composite from the neutronic standpoint lies in the fact that the balance is favourable insofar as capture of ^{238}U is considerably reduced. This is offset by the fact that the Doppler coefficient is much lower and that the effective beta decreases to a degree that could make the consequences of a rod drop accident very damaging. In addition, the decrease, or even the disappearance of conversion, means increasing the quantity of fissile material in order to preserve the same cycle duration, which results in ^{235}U over-enrichment or in an increase in the Pu content, which could lead to positive draining coefficients and thus restrict cycle durations [9]. It should also be considered that this need for over-reactivity would have to be offset by additional means of control, particularly burnable absorbers, whose use could complicate core management.

We shall address here only CERCER and CERMET aspects.

CERCER

The interest of this compound lies in its transparency to neutrons, which favours the balance, as well as in the fact that the ceramic has a neutral chemical behaviour with respect to the primary system. From the neutronics point of view, the behaviour of CERCER is very similar to that of CERMET with a neutronically transparent matrix.

CERMET

Figure 1 shows the distinct difference between the various classes of metals. It is also clear (Table 1) that the kinetic coefficients as a whole are only slightly affected by the nature of the metal and that, in fact, it is the plutonium vector that determines these coefficients. Finally (Table 2), it is to be observed [10] that thorium, although not really inert from the neutronic standpoint, moderates plutonium tendencies.

The studies performed [9,10,11] have demonstrated that two candidates were potentially of interest among the “transparent metals”: Zr, as it is the metal used for the cladding, and silumin (88% Al, 12% Si), which has particularly interesting thermal conductivity and whose advantages have been presented in an accompanying paper [9].

However, for reasons of experience, know-how and material availability, an Mo CERMET (64% Mo, 36% UO₂, 19.6% ²³⁵U enriched) was inserted into the TANOX installation in the SILOE reactor (1994) (Figure 2). Observation of the first experiments (Figures 3-6) showed that, after burn-ups of about 55.4 GWj/t for the CERMET (T2-2 rod) and 40.3 GWj/t for the CERCER (T2-3 rod), the behaviour of the CERCER rod was disappointing: the pellets bonded to each other and adhered to the cladding to such a degree that bits of pellet were torn away upon opening. The CERMET, however, remained absolutely intact; the integrity of the pellets was so well preserved that they were still mobile in the cladding upon opening. The release of ⁸⁵Kr was about three times greater for the CERCER than for the CERMET, whereas the burn-up obtained for the latter was higher. These results led to further our understanding of spinels and an interpretation of the results was performed [12,13]. The swelling phenomenon that had not been observed in the SPIN and CAPRA studies was explained [14,15] by the difference in operating temperature between fast neutron reactors and pressurised water reactors; the lower temperature in PWRs does not allow self-repair (vitrification) of faults in the spinel network under irradiation.

The TANOX-CCE experiment was thus performed in 1997 in order to achieve a burn-up of about 130 GWj/t with CERMET fuel (80% Mo, 20% UO₂, 40% enriched ²³⁵U; the ceramic contained a 2% mass of Er₂O₃ intended to simulate Pu fuels which have to contain poisons for control purposes). In order to take into account the data acquired from the first series of tests, the external faces of the pellets were covered with a thin film (5 µm) so as to prevent gas diffusion through the surface of the ground ceramic spheres tangent to the external surface of the pellet. Analyses are still currently underway to measure all the releases, examine the behaviour of the matrix and the ceramic at very high burn-ups and to measure erbium consumption kinetics.

Assemblies

Studies on conventional 17 × 17 assemblies [9,16] show that the possibility of using a core loaded 100% with standard assemblies and “inert matrix” rods is limited either to highly enriched UO₂ fuels, and is therefore unfavourable from the economic point of view, or to MOX fuels for cycles of 12 to 18 months (Figure 7).

For Pu fuels in inert matrices, the limitations linked to the Doppler coefficient, the draining coefficient and the low value of the delayed neutron fraction lead to non-controllable solutions, or solutions that are potentially sensitive to rod drop accidents or cold shocks. To avoid these situations, two alternatives have been proposed. One is currently being developed by our partners (PSI and ENEA-POLIMI) [17,18], and consists of envisaging a standard 17 × 17 assembly in which the inert

matrix is replaced by thorium and the assembly comprises the number of UO_2 rods needed to achieve acceptable kinetic coefficients. The other solution, developed by the CEA/DRN and known as the Advanced Plutonium Assembly (APA), represents a doubly heterogeneous solution (Het/Het), in which the fuel associates standard UO_2 rods (120) with APA rods (36), which are a solid solution of plutonium and cerium, $(\text{Pu,Ce})\text{O}_2$. The geometry of a conventional 17×17 assembly frame is in fact heterogeneous as it associates large annular rods (diameter: ~ 26 mm, thickness: ~ 1.3 mm) (Figure 8) and UO_2 . The rods are clad internally and externally so as to permit inside circulation of water to enhance Pu fuel cooling on the one hand, and on the other hand, to locally increase the moderation ratio, which can reach a value of 6, thus increasing the efficiency of control methods and rendering the moderation coefficient less negative. The fabrication of APA pellets has been achieved and their optimisation is underway [19-23].

Conclusions and prospects

In view of the history and the choices made in France, inert matrices appear to be an excellent way of providing solutions to the various problems of plutonium stock management. R&D must continue in the domain of understanding and improvement of CERCERs with, as objectives, resistance to irradiation and an increase in thermal conductivity and viscoplasticity. With respect to CERMETS, the notion of cold fuel offers enormous potential for investigation with a view to achieving very high burn-ups, as well as opening up new perspectives with regard to severe accidents [12]. A well-defined research programme is currently being set up to attempt to provide answers to these questions.

Finally, with respect to the core, studies are to be initiated, covering neutronics, thermal hydraulics and management strategies. These studies, associated with fabrication research (APA, CERMET), and accident studies connected to the specificity of these new concepts will, by the end of 1999, result in core images that could open onto innovating projects at the very beginning of the year 2000.

As shown previously [13,14,19-24], the use of APA assemblies in one-third of French nuclear power plants would lead to a stabilisation of the Pu stream. There remains the question of how to use the remaining stocks that have accumulated since the beginning of nuclear power in France up to an equilibrium point 0. These stocks could be advantageously used in CAPRA-type reactors. However, when fossil and fissile resources eventually become scarce and as fusion power will not be able to meet an increasing demand for energy for some time, it will perhaps be the moment to give fast neutron reactors back their real significance and essential usefulness for nuclear power plants as breeder reactors.

Acknowledgements

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Figure 1. PuO_2 CERMET reactivity swing

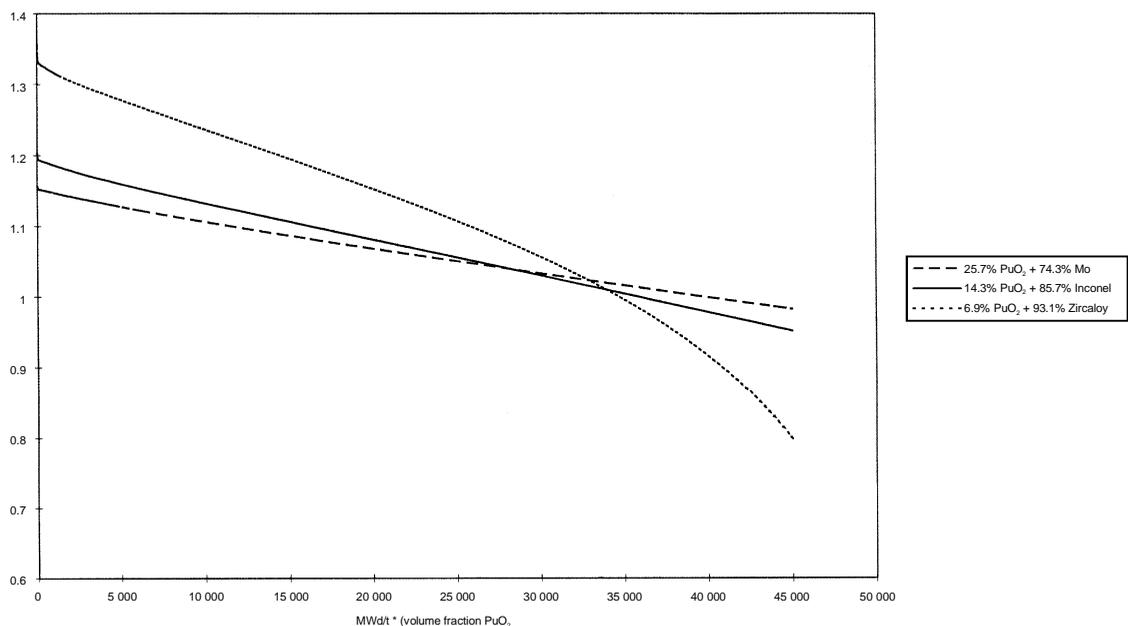
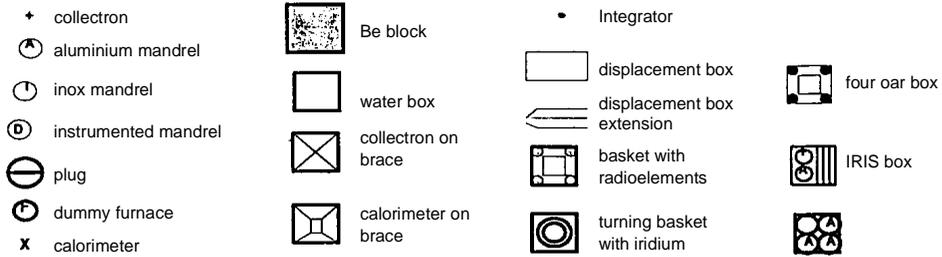
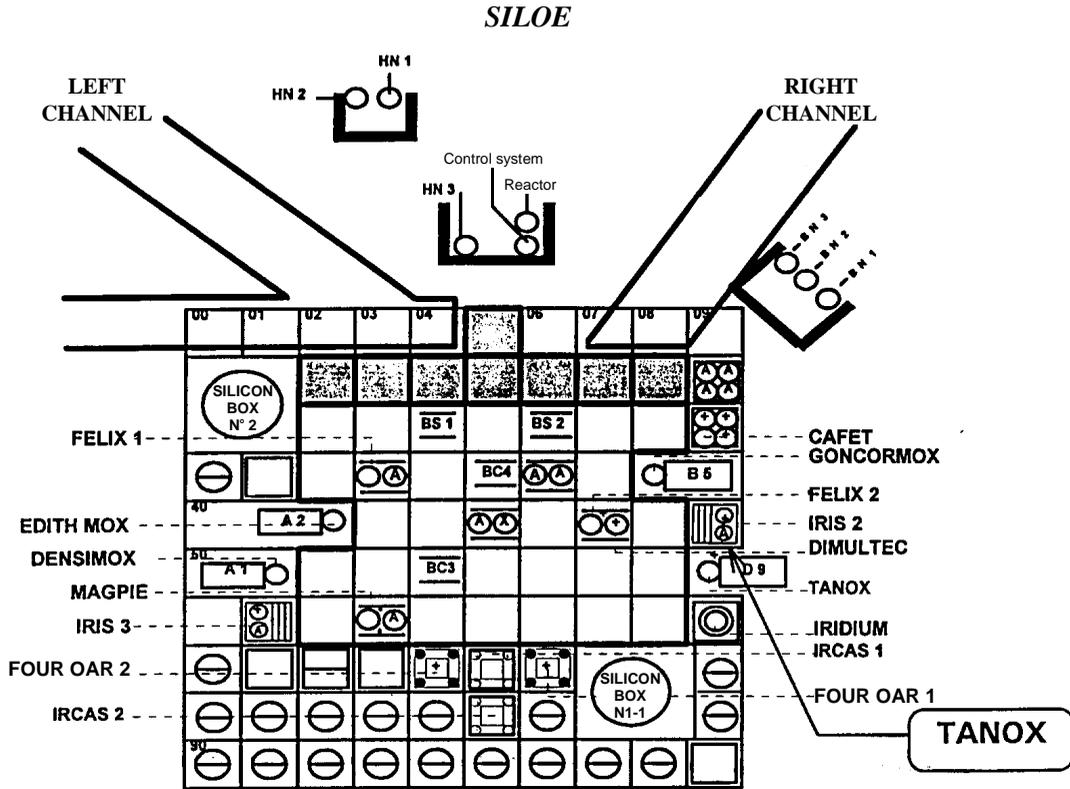


Figure 2. SILOE core and TANOX location



Pin fuel location

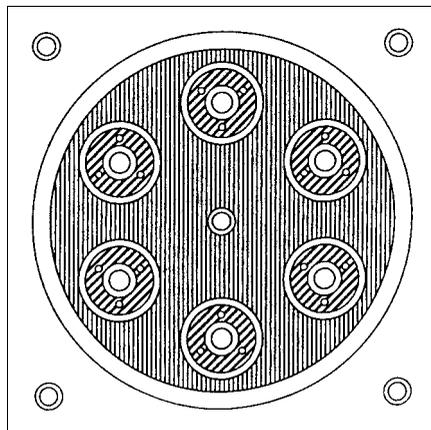


Figure 3. CERMET fuel after irradiation

- 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

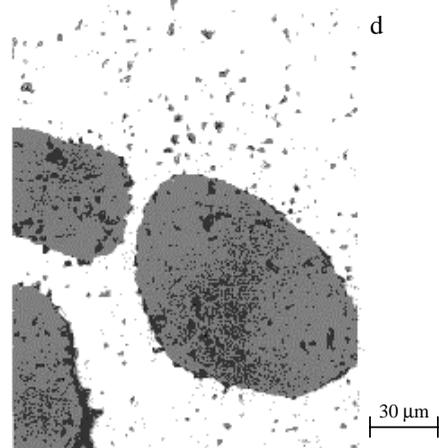
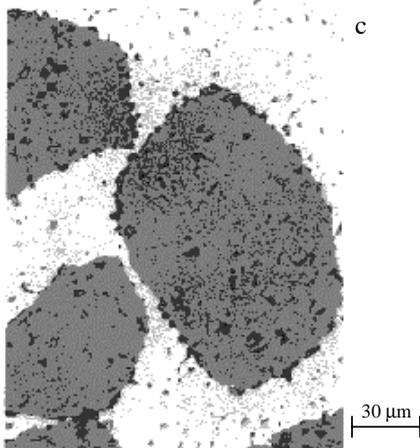
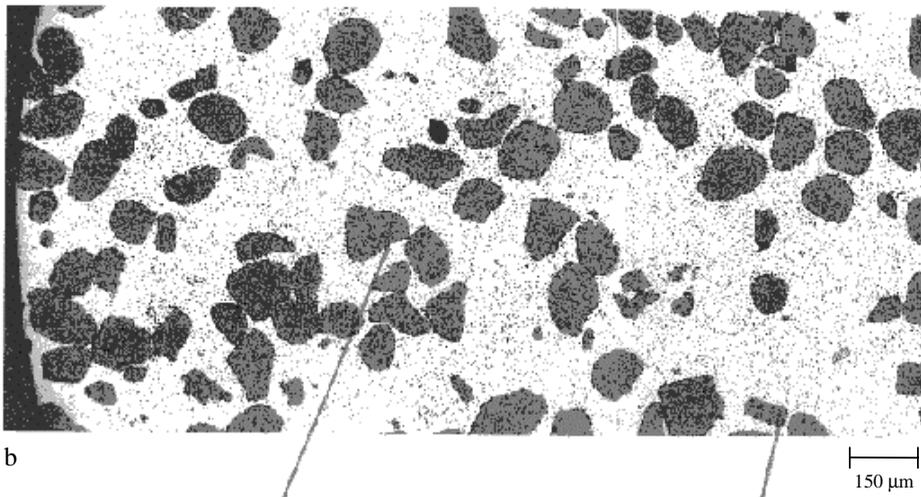
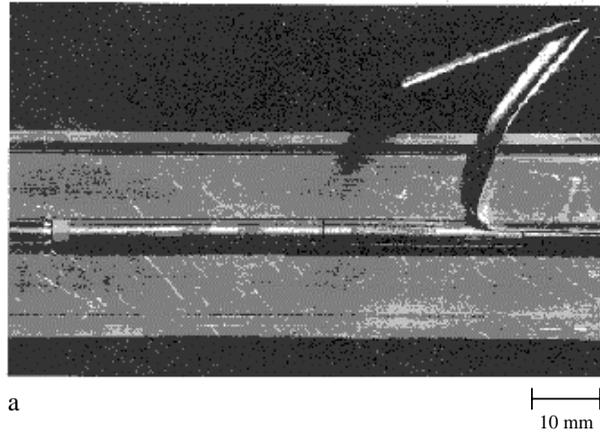


Figure 4. CERCER fuel after irradiation

- 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 centre (after chemical attack)

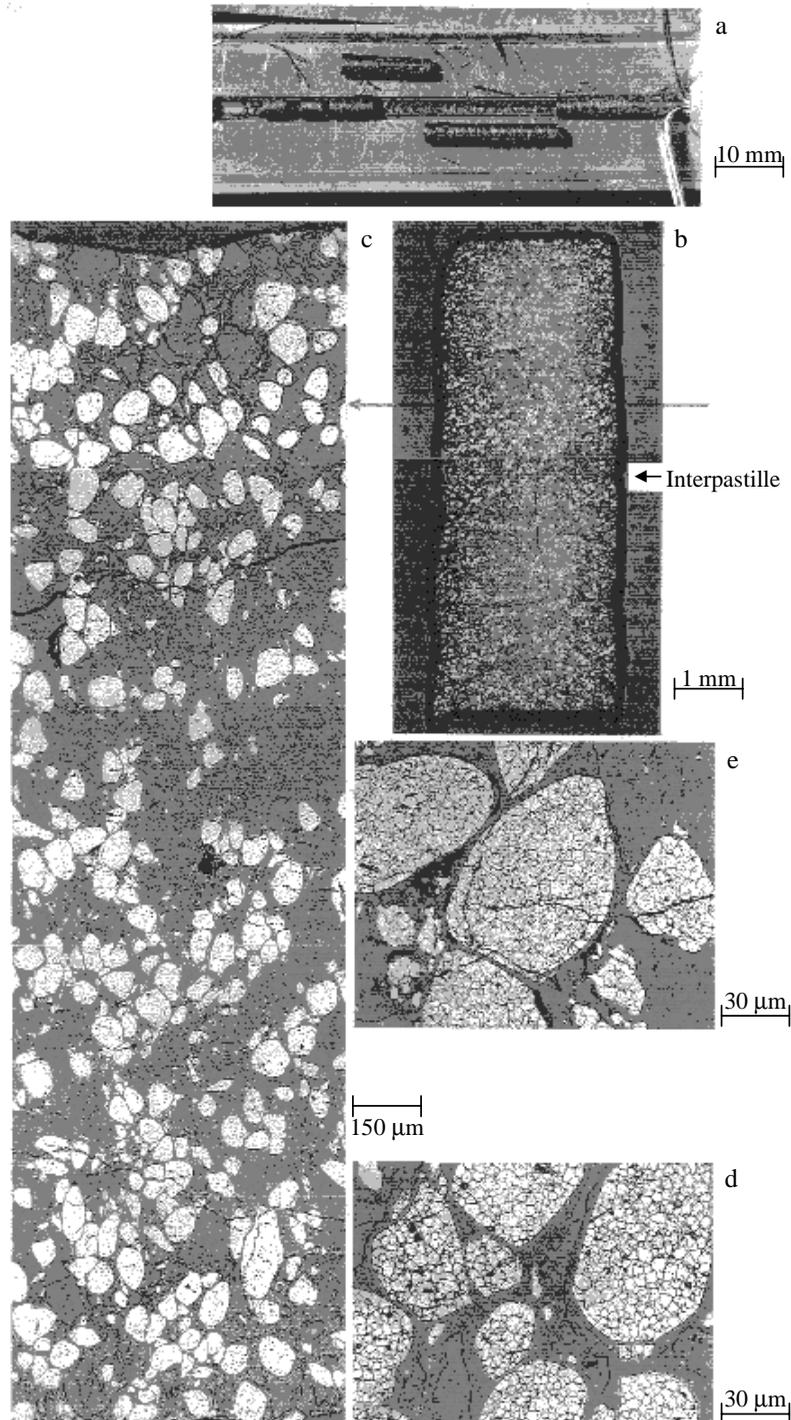


Figure 5. CERMET fuel pellet after irradiation and thermal treatment at 1 580°C for 30 min.

a. Macrography of a pellet

b. Fuel microstructure along a radius

c, d. Gas bubbles precipitation to the joints of grains in a UO_2 nodule

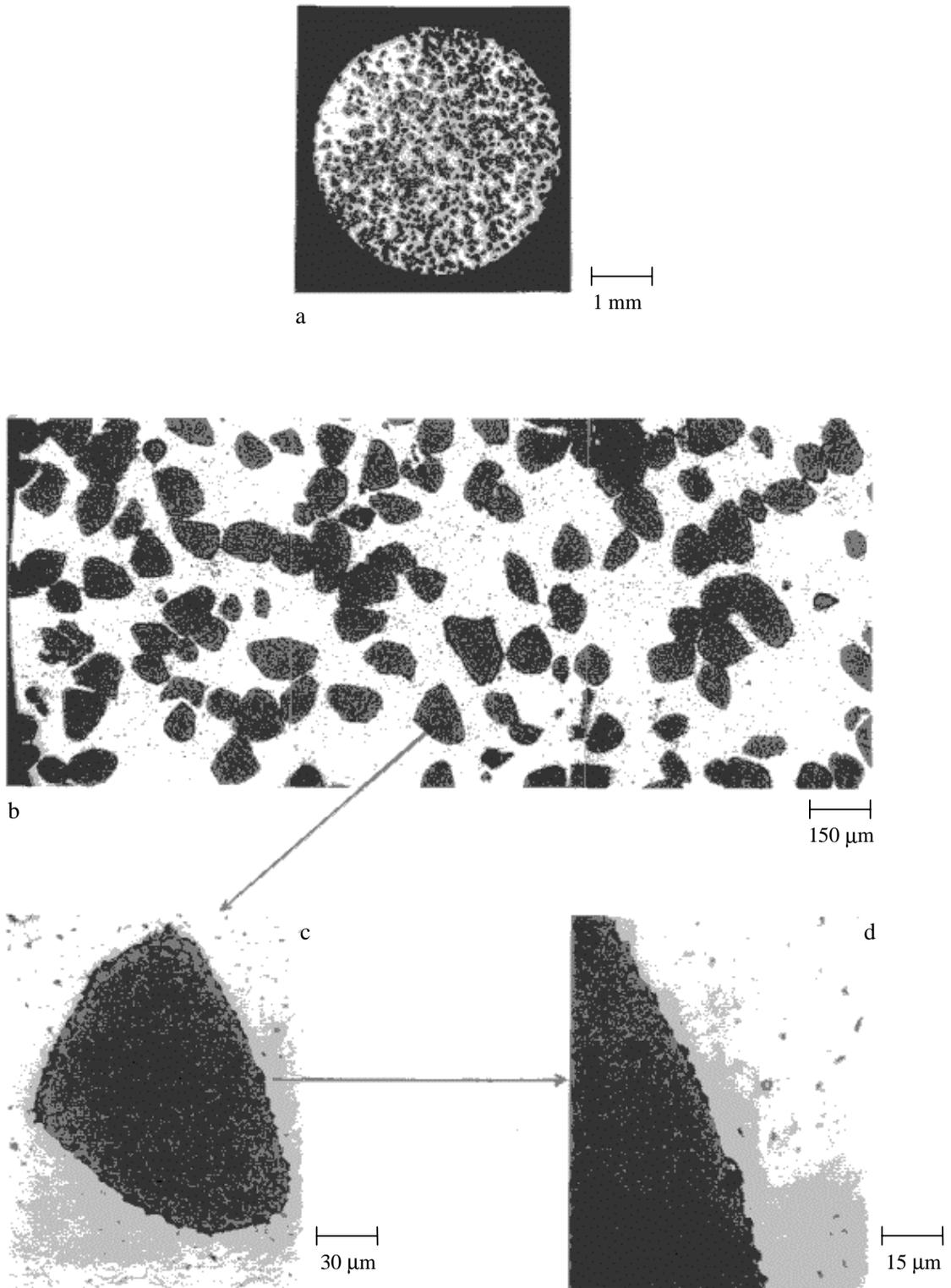
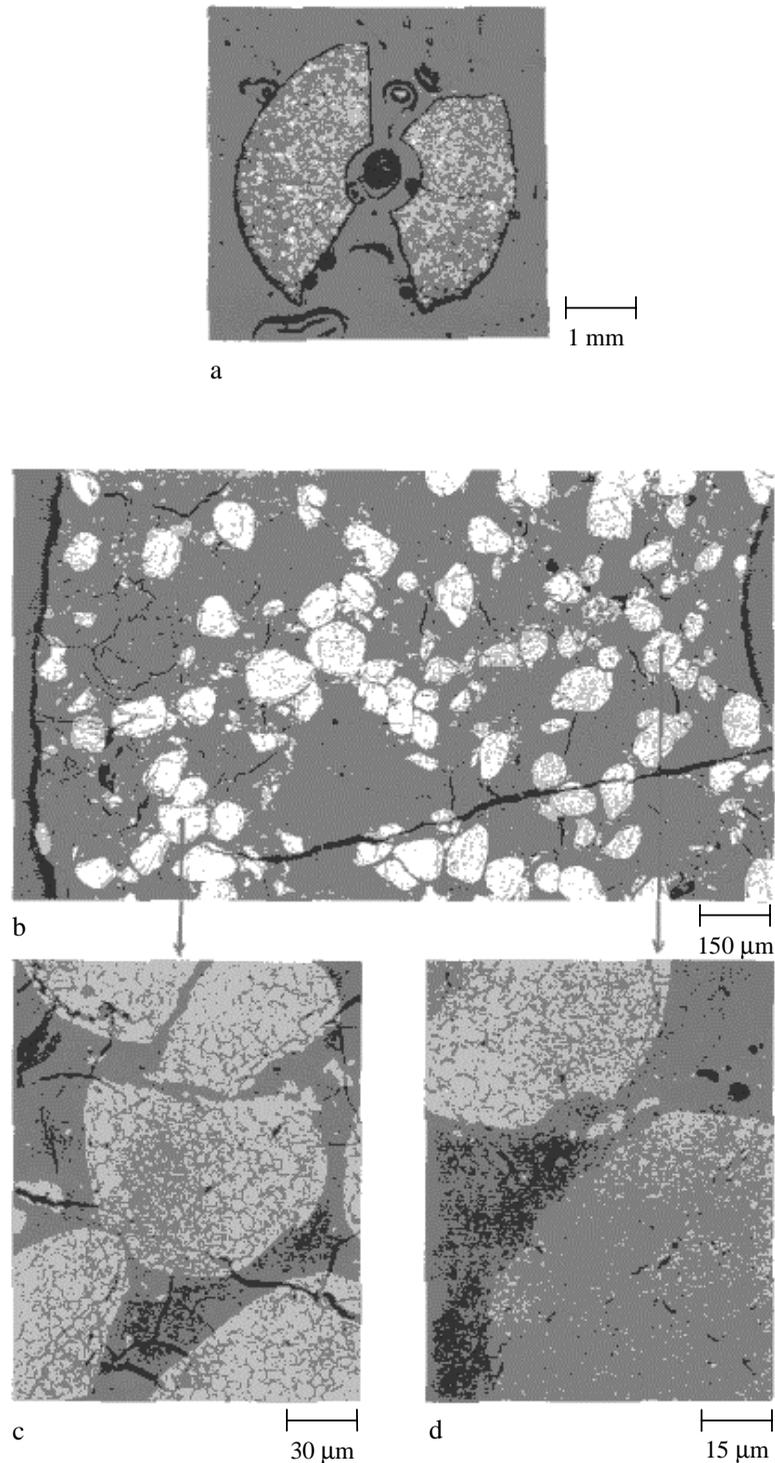


Figure 6. CERCER fuel pellet after irradiation and thermal treatment at 1 580°C for 30 min.

- a. Macrography of a pellet*
- b. Fuel microstructure along a radius*
- c. Gas bubbles precipitation to the joints of grains in a UO_2 nodule and disappearance of the edging ribbon around the UO_2 nodules*
- d. Gas bubbles precipitation to the joints of grains in a UO_2 nodule*



**Figure 7. Cycle length versus Pu content and draining coefficient
CERMET MOX (50% Zr₄, 50% MOX)**

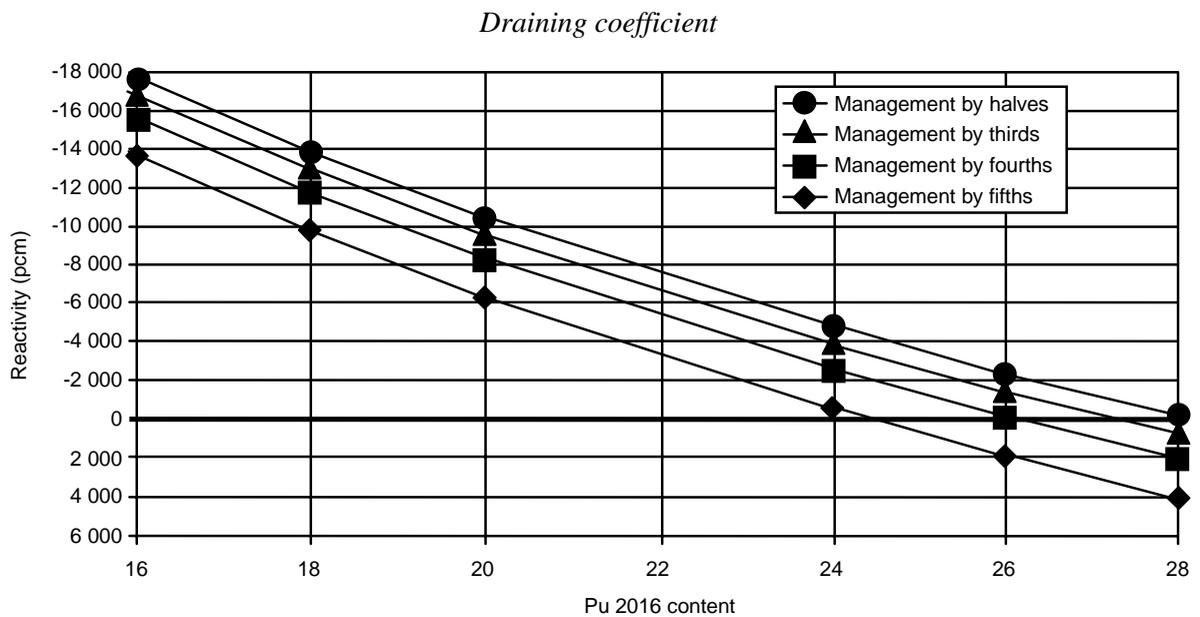
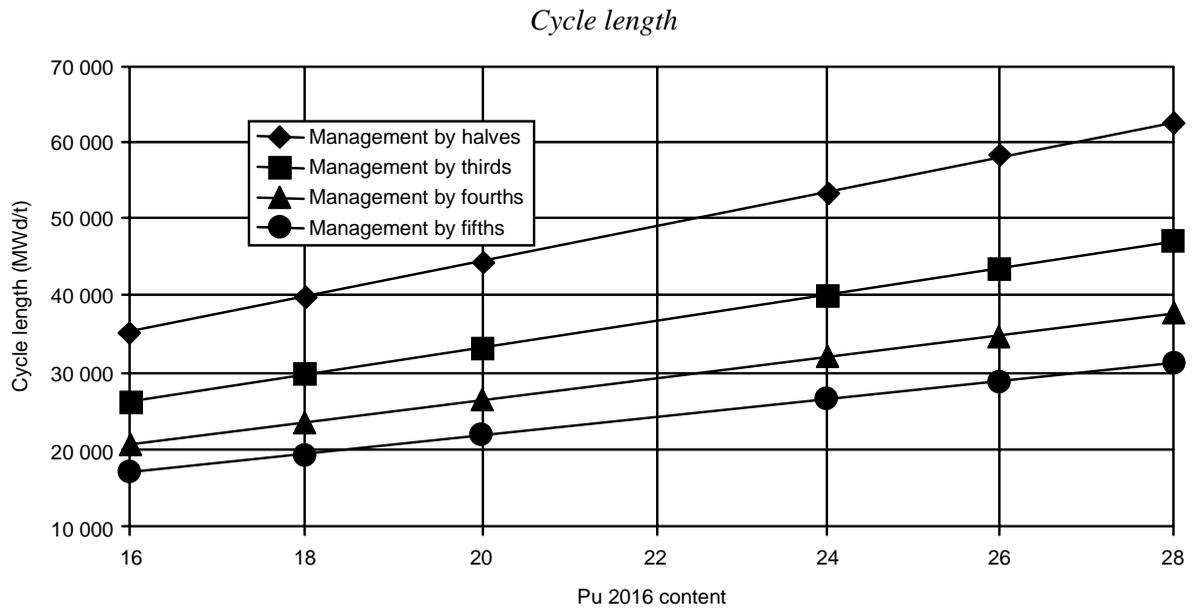
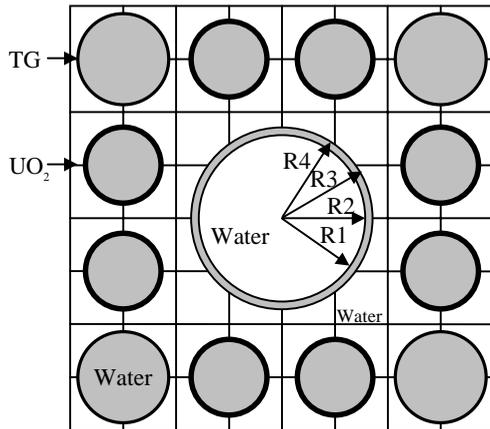


Figure 8. APA: Advanced Pu assembly

Geometric parameters



R4 (mm) = 11.5
 R3 (mm) = 11.0
 R2 (mm) = 9.739 } 1.261 mm
 R1 (mm) = 9.239
 V_{PuO₂}/V_{comb} = 0.23

Local moderation ratio = 5.96 (annular pin)
 Global moderation ratio = 3.47

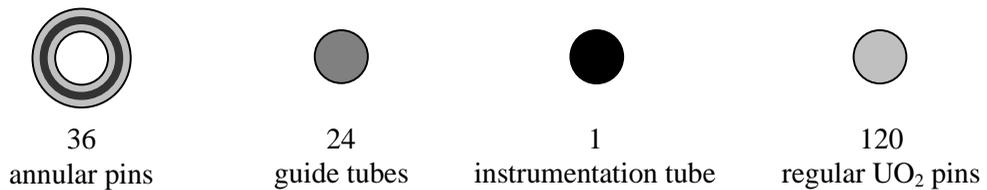
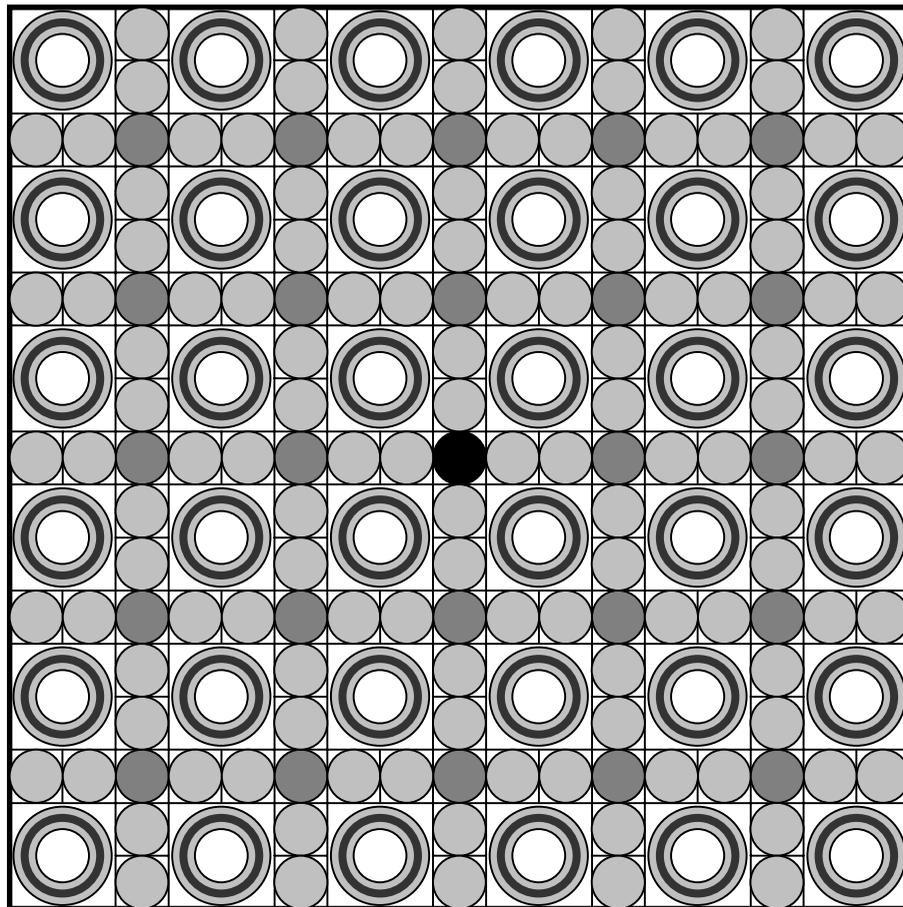
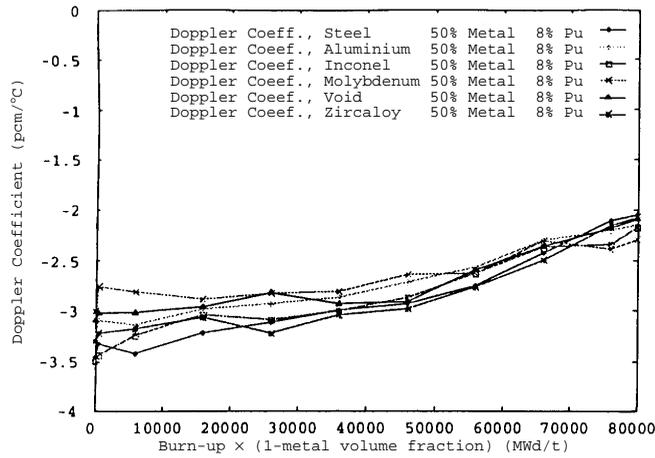
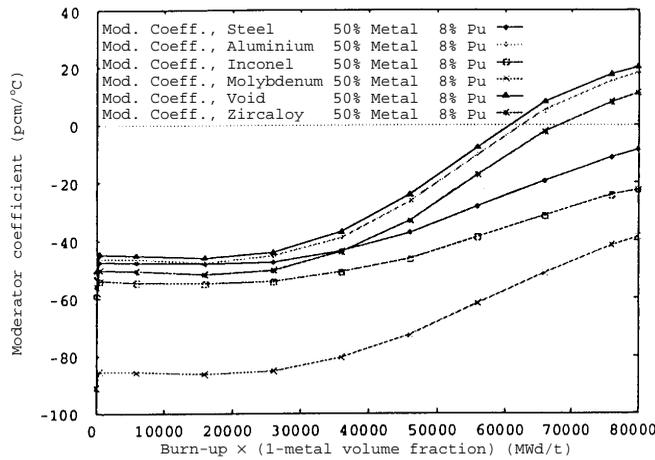


Table 1. Kinetic coefficients for a 50% MET, 50% MOX

Doppler coefficient



Moderator coefficient



Differential boron worth (around 600 ppm)

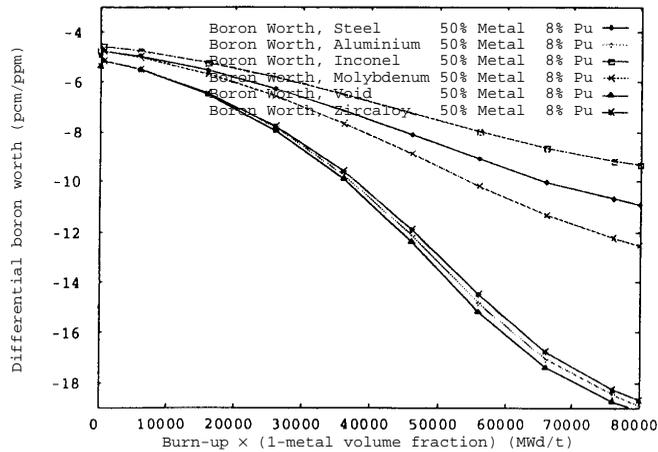


Table 2. Kinetic coefficients for several fuels

Doppler coefficient			Draining coefficient (1 200 boron ppm)		
	BOL	EOL		BOL	EOL
UO ₂	-2.00	-3.39	UO ₂	-61 429	-94 613
UO ₂ + Inconel	-1.99	-3.29	UO ₂ + Inconel	-33 029	-63 650
UO ₂ + Molybdenum	-1.68	-2.59	UO ₂ + Molybdenum	-45 168	-75 969
UO ₂ + Zircaloy	-1.72	-3.55	UO ₂ + Zircaloy	-53 536	-181 167
MOX + Inconel	-2.79	-2.98	MOX + Inconel	+4 953	+3 281
MOX + Molybdenum	-1.93	-1.99	MOX + Molybdenum	+11 618	+13 434
MOX + Zircaloy	-2.97	-3.42	MOX + Zircaloy	-13 471	-28 877
PuO ₂ + Inconel	-1.04	-1.10	PuO ₂ + Inconel	+4 183	-3 841
PuO ₂ + Molybdenum	-0.71	-0.57	PuO ₂ + Molybdenum	+9 481	-8 922
PuO ₂ + Zircaloy	-1.09	-1.80	PuO ₂ + Zircaloy	-9 873	-142 821
PuO ₂ + ThO ₂	-3.30	-3.47	PuO ₂ + ThO ₂	-16 690	-31 704

Cell Calculation

Cycle equivalent = 18 months

1. UO₂

- ²³⁵U enrichment = **3.7%**

2. CERMET UO₂ – Volume fraction of metal = 0.5

- **Inconel:** ²³⁵U enrichment = **14.1%**
- **Molybdenum:** ²³⁵U enrichment = **18.7%**
- **Zircaloy:** ²³⁵U enrichment = **7.5%**

3. CERMET MOX – Volume fraction of metal = 0.5

- **Inconel:** ²³⁵U enrichment = **32.5%**
- **Molybdenum:** ²³⁵U enrichment = **50.0%**
- **Zircaloy:** ²³⁵U enrichment = **18.0%**

4. CERMET PuO₂

- **Inconel:** Volume fraction of Pu = **14.3%**
- **Molybdenum:** Volume fraction of Pu = **25.7%**
- **Zircaloy:** Volume fraction of Pu = **6.9%**

5. CERCER PuTh

- Volume fraction of Pu = **14.0%**

RADIATION STABILITY OF INERT MATRIX FUELS

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Abstract

Inert matrix fuels for transmutation of minor actinides or for burning excess Pu are subjected to four different damage sources:

- i) Thermal or fast neutrons.
- ii) α -decay (5-6 MeV He-ions, ~100 keV daughter recoil atoms).
- iii) Fission fragment impact (70-100 MeV heavy ions of elements between Ga and Dy).
- iv) Intense β - and γ -radiation.

The response of the matrices differs significantly for these damage sources. As an example, spinel MgAl_2O_4 is very stable against neutron irradiation up to high fluences whereas it can be rather easily turned into an amorphous state with properties different from those of the crystalline state (e.g. much lower density) through the impact of fission products. The physics of energy loss of energetic primary ions and of secondary ions in the collision cascades produced by the primary ions and by neutrons is discussed. There is a large database on α -decay damage in many ceramics. In addition, a vast experience exists in simulating the recoil atoms of α -decay through ion implantation in accelerators. Recently irradiations with fission products of fission energy, and also of still higher energies, in large accelerators have yielded extensive information on structural stability, swelling, etc. of many possible inert matrix materials, including Al_2O_3 , CeO_2 , spinel MgAl_2O_4 , zircon ZrSiO_4 , monazite CePO_4 , stabilised cubic ZrO_2 , SiC, etc. These results are summarised and discussed, and confronted with the available but much smaller experience on actual reactor irradiations. The main gaps in the knowledge existing today are summed up and suggestions for the future are made.

Introduction

Reprocessing of spent nuclear reactor fuel produces liquid high-level waste (HLW) containing minor actinides (Np, Am, Cm). If these are chemically separated (partitioned) from the HLW, their radiotoxicity is reduced. The quantity of the minor actinides should then also be significantly reduced by transmutation, i.e. by further irradiation in existing or new nuclear reactors. To avoid the formation of new actinides, the separated Am, Np (or Cm) are best introduced into U-free, innovative fuels. The same is true for excess civilian or military Pu. This means that the Pu to be burnt and the partitioned actinides to be transmuted should be incorporated into a matrix which is “inert” against the formation of new actinides, replacing the UO_2 of today’s fuels or, alternatively, an actinide compound (not containing U) should be used without any support. The possibilities for such innovative fuels have been summarised before [1]. In the present paper, the strategy of using an inert matrix to obtain structural and mechanical stability as well as acceptable thermal properties is adopted. In this case, a material with low atomic number and with properties similar or preferably even better than those of today’s fuel UO_2 should be selected. The U-free fuels with a support can be either a homogeneous material, i.e. a solid solution (e.g. $(\text{Ce,Pu})\text{O}_2$), or a heterogeneous material, i.e. a two-phase material (e.g. AmO_2 in spinel MgAl_2O_4). Because of the low melting points and the existence of different crystallographic phases of actinide metals (e.g. six different phases for Pu with a melting point of only 908 K), the actinide will be used as a ceramic in this latter case. The inert matrix can be a metal with a high melting point (case of CERMET, e.g. PuO_2 in W) or also a ceramic (case of CERCER, e.g. AmO_2 in spinel or in ZrO_2).

The selection of suitable and promising candidate materials as support for the actinide to be transmuted or the Pu to be burnt, must be based on the neutronic, physical and chemical properties in relation to fuel fabrication, fuel performance and, if wanted or necessary, reprocessing. Therefore, the neutron capture cross-sections of the elements constituting the matrix should be small to save as many neutrons as possible for transmutation or burning, to keep neutron activation low and to avoid changes in materials properties. Other criteria to be considered are ease of fabrication, availability and low costs of the starting materials; good thermal properties (no phase transformation, high melting point, good thermal conductivity); good mechanical properties; good compatibility with clad (zircaloy, stainless steel) and with the coolant (H_2O , Na) [2].

A particular concern for extended reactor irradiation and/or high actinide content of the fuel is radiation stability. Stability against radiation of inert matrices has to be considered for the impact of four damage sources:

- Neutrons of the reactor (thermal, epithermal or fast).
- α -decay, i.e. impact of an He-ion of typically 5-6 MeV energy and the heavy recoil atom, i.e. the daughter atom of the α -decay event, e.g. ^{238}Pu in the decay of ^{242}Cm , with an energy of ~ 100 keV.
- The impact of fission fragments, i.e. swift heavy ions (fission products of elements between Ga and Dy) with energies between 70-100 MeV.
- Ionisation by the intense β -, γ -radiation in the reactor.

Note that neutron and fission damage occur only in the reactor (neglecting spontaneous fission) where the fuel is at elevated temperatures. In contrast, α -decay starts from the moment of fabrication and is thus also accumulated before and after reactor irradiation, when the fuel is cold. β -, γ -irradiation is largest during irradiation, but exists during storage as well. These complex processes of damage formation are treated in the present paper for the case of ceramic inert matrices. These damage processes should not render the matrix amorphous, since the properties of an amorphous phase are usually very different from those of the starting crystalline phase. A good example is the much lower density, an example being Al_2O_3 which swells by up to ~30% when rendered amorphous by ion impact. During recrystallisation, e.g. at ~970 K for amorphous Al_2O_3 , high gas release occurs [3], another unwanted process. In addition, no excessive swelling due to other mechanisms and no excessive degradation of the thermal conductivity, λ , due to damage accumulation should occur. A good knowledge of radiation damage effects as a function of irradiation (and storage) time and temperature is thus a vital prerequisite to select a suitable matrix. The present state of knowledge is described in the following. Earlier reviews containing general information on radiation damage in ceramics can be found in Refs. [4-7].

Damage production due to different damage sources

In this section, a brief description of the processes of formation of atomic displacements (lattice defects, Frenkel pairs) and of structural changes produced by the different damage sources is given.

Displacements produced by neutrons

A neutron of mass $M_1 = 1$ and of energy E_n , while passing through a medium of mass M_2 , will occasionally collide (mean free path ~1 cm) with a lattice atom, imparting to it an energy (depending on the impact parameters) up to a maximum energy given by:

$$E_{\max} = \frac{4M_1M_2}{(M_1 + M_2)^2} E_n \quad \text{or} \quad \sim \frac{4E_n}{M_2} \quad \text{for } M_2 \gg M_1$$

The maximum primary knock-on atom (PKA) energies for a neutron of energy $E_n = 1$ MeV are thus for Al_2O_3 : 150 keV for Al, 250 keV for O; and for ZrO_2 : 45 keV for Zr, 250 keV for O, but most interactions with neutrons will lead to a smaller energy transfer.

The minimum neutron energy to produce one displacement is given by $E_{\max} = E_d$, where E_d is the displacement energy (typically 20 to 40 eV). Hence:

$$E_n^{\min} \sim 0.1 \text{ keV}$$

Thermal (low energy) neutrons do not produce direct displacements.

Irradiation with fast neutrons will thus cause localised collision cascades, similar to those produced by the recoil-daughter atoms of the α -decay (see below), or to those produced by ion implantation with ions in the energy range of a few to some 10^2 keV energy. Neutron irradiation at low T and high fluence can also produce metamictisation (amorphisation), e.g. in quartz.

Displacements produced by β -, γ -decay events

Electrons can also produce isolated displaced atoms if their energy is high enough. The minimum energy, E_n^{min} , to displace a lattice atom is given by:

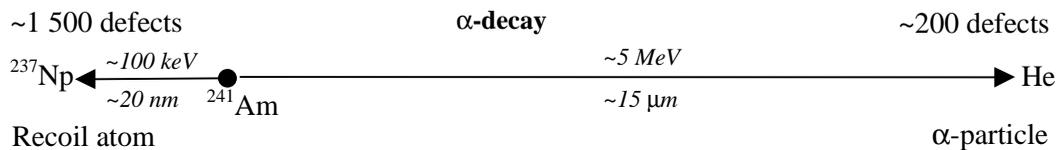
$$E_d = 2 \frac{m_0}{M} \frac{E_e^{min}}{m_0 c^2} (E_n^{min} + 2m_0 c^2)$$

where m_0 is the electron mass, M the mass of the displaced atom and c the velocity of light. β -decay causes thus very few isolated point defects. In-cascade recovery due to thermal spike effects is absent and a given damage level (dpa level), e.g. expressed as increase in the lattice parameter, will cause a larger effect than the same dpa level produced by heavier ions of lower energy. However, ionisation processes may also cause atomic defects, in particular in materials not resistant against radiolysis. At high doses and low temperatures, electrons can cause amorphisation, e.g. in α -SiC below 220 K. At higher temperatures, a large fraction of the defects (isolated Frenkel pairs) caused by electrons are recoverable, compared with the high density of inter-cascade defects caused by ion impact. Thus no amorphisation occurs in structurally unstable substances under electron impact at higher temperatures, whereas it still occurs under the impact of heavy ions (see below).

α -decay damage

The α -decay has two damaging sources:

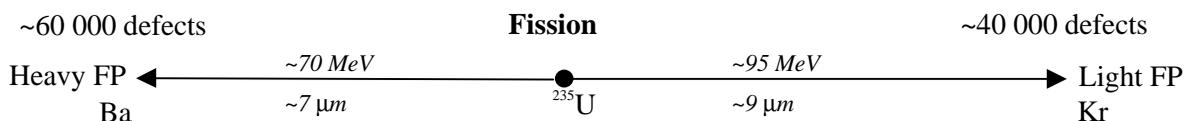
- A He-ion (α -particle) of ~ 5.5 MeV, with mainly electronic stopping, producing about 200 displacements, most of them as largely isolated defects at the end of its range of some 10-20 μm .
- A heavy recoil atom, e.g. ^{237}Np in the decay of ^{241}Am which receives a recoil energy E due to conservation of momentum, $ME = mE_\alpha$, hence typically ~ 100 keV (or 91 keV in the decay of ^{241}Am). These recoil atoms show predominantly nuclear stopping and produce a dense collision cascade with typically $\sim 1\,500$ displacements on a short distance of ~ 20 nm. Defect clustering can occur stabilising the damage.



Impact of fission fragments

Fission produces two fission products (FP) (plus 2-3 neutrons). These FP fall into two groups, the light ones (LFP, typically Mo or Kr with ~ 95 MeV energy) and the heavy ones (HFP, typically I or Ba with ~ 70 MeV energy). Further typical values (for the case of UO_2) are given in the illustration below: range, number of defects produced, etc. The exact values depend on the substance used (e.g. E_d , atomic number, density, etc.). Because of the high energy deposition rate, typically

20-30 keV/nm, a locally (over-) heated track (fission spike or thermal spike) may be formed. Such fission tracks are visible in transmission electron microscopy in many materials, an example being spinel MgAl_2O_4 . Most of the energy deposition is by electronic energy loss, in particular for the more energetic light FP. This, in addition to causing local heating up to or above the melting point, can cause the formation of additional defects and/or rearrangement of existing defects. An extreme case is the destruction of pre-existing fission gas bubbles by the passage of a fission spike, known to occur in the conventional nuclear fuel, UO_2 [8].



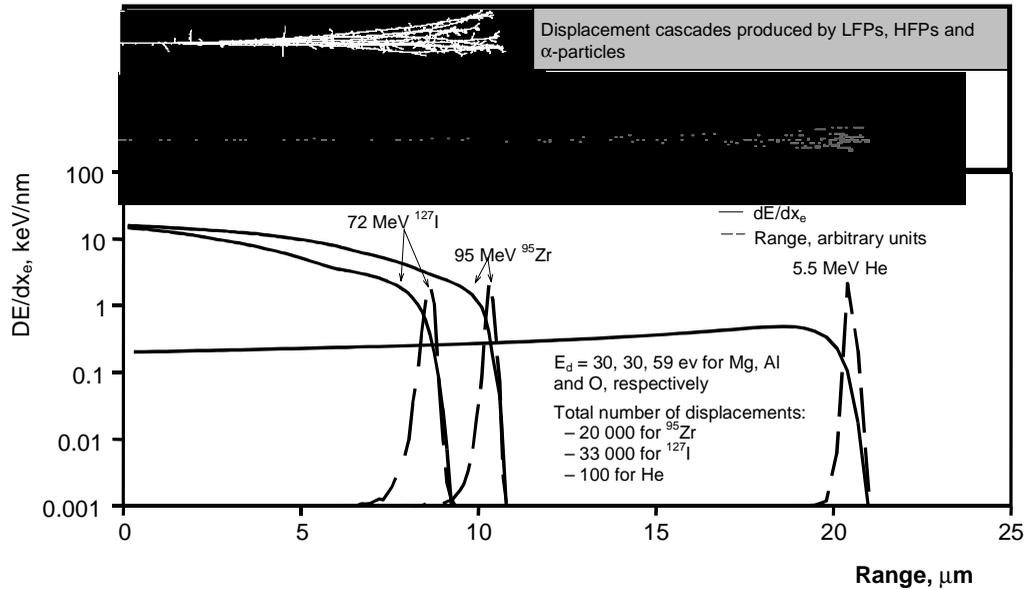
A short description of the sequence of events in the fission spike is the following:

- *Primary phase or ballistic phase* – the passage of the fission fragment – is very short but it defines the initial size and shape of the spike. Most Frenkel defects are produced by secondary collision cascades. The deposited Coulomb energy is dissipated into local heating through electronic interactions with recoiling ions to produce a thermal spike.
- *Second or quenching phase* – recombination of vacancies and interstitials occurs when the spike comes to thermal equilibrium. An interstitial-rich outer zone and a vacancy-rich inner zone form. The hydrostatic pressure field originally created by the molten core of the spike – contributing to the separation of interstitials from the vacancies of the Frenkel pairs formed in the primary phase – is replaced by compressive stresses in the outer zone and dilatational stresses in the core.
- *Third or track annealing phase* – more recombination occurs, some vacancy clusters are stabilised by fission gases forming embryos for later bubbles.

The processes in these three phases are repeated many times all through the fuel volume in a homogeneous fuel. All of the fuel is affected after a rather short time, the level of one displacement per atom (dpa) being typically reached within less than one day. The consequences are a significant fission-enhanced diffusion [9], fission-enhanced creep, re-solution of fission gas from bubbles, etc.

Figure 1 shows in its upper part the large difference between the high defect density in a fission spike and the largely isolated defects in α -particle damage, and in its lower part the very large (by up to a factor of $\sim 10^2$) difference in electronic energy loss, $(dE/dx)_e$, between these two damage sources. (Note the logarithmic scale.) The energy loss curves show the electronic stopping only, i.e. the ionisation part, with high values of 18 keV/nm at the point of fission (full ion energy) for the case shown, i.e. spinel MgAl_2O_4 . The nuclear, i.e. the displacement damage part, peaks at the end of the range ($\sim 1 \text{ keV/nm}$) and is very small (but still present) at the point of fission ($\sim 0.1 \text{ keV/nm}$). The ratio of nuclear to electronic stopping is always low for those high energies, but it varies between 1:180 at the fission site and some 1:3 toward the end of the range, or even $< 1:1$ at the very end. The recoil atoms of the α -decay and the high defect density produced by their slowing down are not included in the sketch below because of the large difference in range: about 1 500 displacements are formed along a very short track of only $\sim 20 \text{ nm}$, or $0.002 \mu\text{m}$, as shown in the sketch for α -decay on a previous or $0.002 \mu\text{m}$, as shown in the sketch for α -decay on a previous page.

Figure 1. Upper part: displacement cascades of a Zr and an iodine fission product as well of a 5.5 MeV α -particle in spinel $MgAl_2O_4$. The individual dots correspond to displacement events. The lateral straggling at the end of the range is indicated with a scale in μm ($\pm\mu m$). Lower part: electronic energy loss (full lines) and ranges of the same three ions (dashed lines). Material: spinel $MgAl_2O_4$; calculations with the TRIM96 code [10].



Experimental techniques, simulation experiments and results

Many potential candidates for inert matrices have been produced with incorporated short-lived actinides (e.g. ^{238}Pu , ^{241}Am , ^{242}Cm) [7,11] and many candidate substances have been irradiated in nuclear reactors [6]. Also, so-called “natural analogues”, i.e. natural minerals containing some U or Th have been studied to get helpful information on α -decay damage. These minerals have accumulated large amounts of damage at very low rates over geological time scales. These investigations have provided an extensive database for neutron and α -decay damage. Additional results have been obtained by simulating the two contributions to α -decay damage by using He-beams of different energy [12] to study damage due to the α -particle and heavy ions of about 10^2 keV energy to investigate in a second single effect study the interaction of recoil atoms with the matrices.

Figure 2 shows as an example the swelling of zircon, a material with a large solubility for actinides, as a function of α -decay dose [7,13]. The crystalline phase undergoes a unit cell expansion ΔV_{uc} but its fraction f_c decreases due to an increased fraction f_a of the amorphous phase. Similarly well understood results exist for a large number of ceramics [7,11], mainly studied in the context of radioactive waste solidification and burial. This database and the knowledge that emerged from these studies provides a very valuable starting point for selecting and testing inert matrix fuels.

Figure 3 gives a typical example of neutron damage effects, again shown for volumetric swelling. Al_2O_3 is seen to be unstable against fast neutrons. In an early study [3], Al_2O_3 was shown to be amorphised by the impact of heavy ions (Xe, 40 keV), to recrystallise at ~ 970 K, the moving boundary crystalline/amorphous sweeping the rare gas out of the crystal. Al_2O_3 is also unstable against fission product impact (see below) with swelling of up to 30%. It is thus discarded as inert matrix.

Figure 2. Experimental data for macroscopic swelling V_m in Pu-doped zircon $ZrSiO_4$, with the calculated fraction of crystalline (f_c) and amorphous (f_a) components of total swelling [7,13]

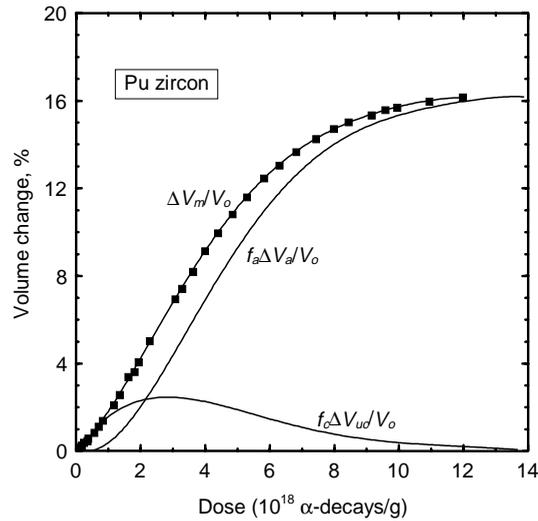
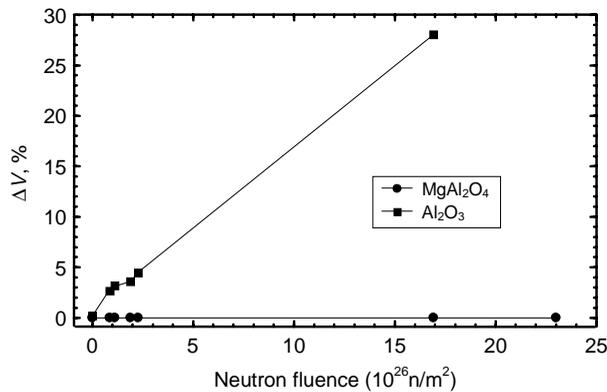


Figure 3. Swelling of spinel $MgAl_2O_4$ and of Al_2O_3 during neutron irradiation (925 K, $E_n > 0.1$ MeV). At the highest n-fluence (Phenix reactor, Santenay experiment), 28% swelling and powder formation of Al_2O_3 were observed. $f_a \Delta V_d/V_0$



A number of criteria exist to predict structural instability and formation of a metamict, amorphous state. These include:

- i) *A structural criterion*: cubic materials tend to be stable, anisotropic materials (like hexagonal Al_2O_3) tend to become amorphous [3].
- ii) *A bonding criterion*: materials with high ionicity (Pauling ionicity > 0.6) tend to be stable, whereas less ionic materials and semiconductors tend to become amorphous [4]. This empirical criterion is a valuable refinement in adding the aspects of co-ordination and the argument of variable bond-angles versus fixed bond-angles in strongly ionic materials. This criterion has subsequently been refined by a number of authors and is now a valuable means of predicting structural stability.

iii) A thermal spike model stating that a substance remains crystalline if its amorphous phase has a low crystallisation temperature, T_c : if T_c/T_m (melting point) is < 0.27 the material remains crystalline, whereas materials with $T_c/T_m > 0.27$ amorphise under heavy ion impact. For instance, $T_c/T_m = 970/2345 = 0.41$ for Al_2O_3 . Amorphisation is not the only ion impact induced phase change. Ion impact can also recrystallise previously amorphous ceramics, and can cause polygonisation, i.e. the formation of very small, but still crystalline grains. This phenomenon is also called grain-subdivision and is of large interest in high burn-up UO_2 fuel where it occurs in the rim of UO_2 -pellets (rim effect). It was also found in other materials which are otherwise structurally stable against ion impact in the 10^2 keV to 10^2 MeV regime, e.g. in MgO and ZrO_2 [3,14,15].

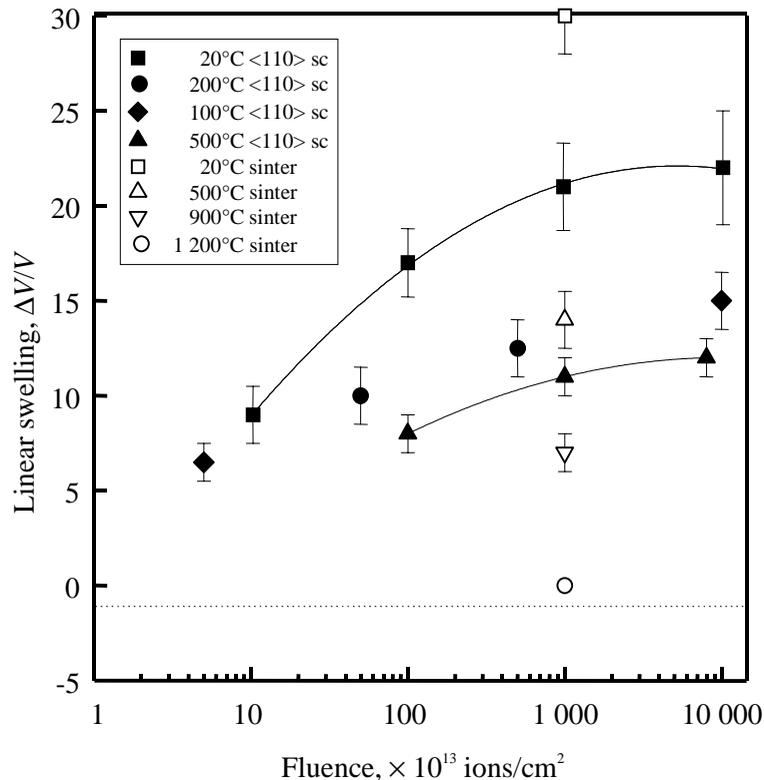
In contrast to the large swelling of Al_2O_3 , the spinel MgAl_2O_4 is seen (Figure 3) to be dimensionally extremely stable during neutron irradiation. This radiation resistance has been attributed to several factors, in addition to crystal structure and ionicity: efficient point defect recombination, large size of the smallest stoichiometric cluster (a void of one Mg, two Al and four O vacancies), high stability of faulted loops (impeding loop unfauling and evolution to a network dislocation structure), efficient site exchange of different cations and low interstitial migration energies [16]. MgAl_2O_4 was therefore first considered to be a good inert matrix. The impact of heavy ions in the 10^2 keV energy regime caused only partial amorphisation in the Al_2O_3 sublattice whereas the MgO sublattice remained stable [17]. Continued work described below showed, however, a severe instability of spinel towards fission damage.

The damage produced by the stopping of fission product can also be realistically simulated in single effect studies by using beams of fission products of fission energy, e.g. iodine ions of 72 MeV. Both irradiation rate and dose (fluence) can be varied, as well as the specimen temperature whenever a small furnace is installed, as in the experiments performed at the TASCC accelerator in co-operation with Drs. P.G. Lucuta and R.A. Verrall in Chalk River, AECL, Canada, which are summarised below.

Figure 4 shows the response of spinel to the impact of fission products at fission energy. This material, which is very stable during neutron irradiation, shows very significant swelling due to fission damage, as evidenced by a “pop-out” of the irradiated area (see right side of Figure 4). Since the irradiated region is constrained in the lateral direction by the surrounding unirradiated MgAl_2O_4 material, all of the volumetric swelling occurs in the direction normal to the surface. In the left part of Figure 4, the measured “pop-out” or “step-height” is related to the total range of the irradiating beam (see Figure 1) to calculate swelling. Recent cross-sectional electron microscopy studies of spinel have shown [16] that amorphisation occurs in the electronic stopping region of the fission product but only down to a threshold dE/dx_e value of 7 keV/nm, hence to a depth of 4.3 μm , the range being $\sim 7 \mu\text{m}$. If swelling is solely due to amorphisation, the resultant swelling values will be even larger than those shown in Figure 4.

Amorphisation and large swelling (up to 30%) have also been seen in Al_2O_3 irradiated in the same way as spinel in Figure 4, as mentioned above. It occurred already at low fluences and provided an additional argument to discard Al_2O_3 as inert matrix. Poor radiation stability during fission product impact (again 72 MeV iodine) was also seen with zircon and with monazite CePO_4 . CeO_2 showed swelling and indicated polygonisation, whereas no observable swelling up to high fluences was found for UO_2 , SiC and Y_2O_3 -stabilised ZrO_2 .

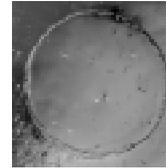
Figure 4. Swelling of single crystalline and of sintered spinel $MgAl_2O_4$ as a function of fluence of a fission product of fission energy (72 MeV iodine) with the irradiation temperature as parameter



Profile of an implanted area



Optical micrograph of an implanted area



This latter material did not become amorphous, as expected based on the above criteria, but developed a heavily disordered – though still crystalline – structure, as evidenced by Rutherford backscattering/channelling experiments and transmission electron microscopy. $(Zr,Y)O_{2-x}$ proved also to be stable during impact with heavy ions in the 10^2 keV region [19] and is thus a stable matrix, though its resistance against radiation is not as good as that of UO_2 . Though amorphisation does not occur in UO_2 and has not been observed so far in ZrO_2 , polygonisation has been found at high fluences of non-soluble ions (e.g. Xe and I). Polygonisation is the term used to describe the rearrangement of those dislocations formed during irradiation which do not annihilate one another, into walls of dislocations, thus forming low energy “subboundaries” and rather perfect but slightly disoriented subgrains between those new boundaries. The resulting structure is characterised by new small grains; the technological terms generally used are grain subdivision, and “rim-structure” for the specific case of high burn-up UO_2 [20].

The present results can favourably be confronted with results of reactor irradiations. In the early days of development of peaceful nuclear energy, dispersion-type fuels with enriched UO_2 in matrices of Al_2O_3 and ZrO_2 were irradiation-tested at the Bettis Laboratories [21]. The Al_2O_3 matrix swelled by ~20%, whereas a fuel based on UO_2 and CaO-stabilised ZrO_2 showed good structural and dimensional stability, in agreement with the above results. Very recently, spinel with very small inclusions of AmO_{2-x} (11 wt.% total Am-content) was irradiated at HFR Petten for 360 days, with a (calculated) transmutation of the Am of ~99%, thereof 35% fission [22]. The spinel matrix swelled into the available free space of the irradiation device, i.e. ~18 vol.%, again in agreement with the present

results [23]. Since Am is largely transmuted to other actinides, the effective decay chain being $^{241}\text{Am} \rightarrow ^{242}\text{Cm} \rightarrow ^{238}\text{Pu} \rightarrow ^{239}\text{Pu}$ with a large amount of α -decays of the short-lived nuclides ^{242}Cm and ^{238}Pu , about ten times more He was calculated to be formed than fission gases. The contribution of this large amount of He to the observed swelling, as well as the behaviour of He (bubble formation, solubility, release) in inert matrices have still to be quantitatively investigated. Again, simulation studies with accelerated He-beams are a good means to obtain the missing knowledge.

Conclusions, gaps in our knowledge and suggestions for future work

The complex phenomenon of bulk radiation damage in inert matrices can only be understood by proper consideration of the damage source: α -particle, recoil atom, fission event, neutrons, β - and γ -decay. Single effect studies, e.g. accelerator experiments at controlled temperature are a good way to understand these different damage effects.

Caution should be used when one single effect study using one damage source only promises radiation stability. The matrix may yet be susceptible to one of the other damage sources or to the complex interplay of all damage sources and effects during its use for transmutation. The final proof of stability will have to be made with a reactor irradiation, preferably in an instrumented capsule.

Even if damage effects are important, as when spinel is subjected to fission, an innovative, optimised fuel microstructure can reduce these effects, e.g. by placing the actinide to be incinerated or transmuted into small spheres (~200 μm diameter) of a substance resistant against α -decay and fission damage such as $(\text{Zr},\text{Y})\text{O}_2$ which then is incorporated into a matrix like spinel offering good radiation resistance against neutrons and good thermal properties [24].

While the databases for neutron damage and for α -decay damage are rather comprehensive for a number of possible inert matrices, this is not the case for fission damage. In particular, the effects of temperature during irradiation should be studied. Very little is known on radiation resistance of nitrides such as ZrN in solid solution with AmN or PuN, or candidates for cermets like TiN or CeN. In addition to investigating their resistance against the four damage sources discussed, their other properties (thermal, mechanical, interaction with coolant and clad) should be carefully studied. The same is true for the corresponding actinide compounds, i.e. PuN, NpN and AmN.

Finally, the chemical effects of α -decay and fission should be well understood before deciding on a specific matrix. The in-growth of daughter products of the α -decay and of fission products proceed in parallel to the in-growth of damage and both phenomena are known to affect the fuel behaviour, a good example being the thermal conductivity of today's fuel, UO_2 [25]. Studies similar to those performed on UO_2 and SIMFUEL [25] are needed to obtain the specific conditions of inert matrices as well.

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DISPOSITION OF EXCESS PLUTONIUM BY THE ROX-LWR SYSTEM

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Abstract

As an option for the disposition of excess plutonium both from nuclear power plants spent fuels and from dismantled warheads, a system of plutonium rock-like oxide (ROX) fuels and their once-through burning in LWR (ROX-LWR system) has been investigated. The features of the system are remarkable both from the proliferation resistance and environmental safety points of view. As the ROX fuel is a kind of inert matrix fuel with ^{238}U -free matrices, it has a high plutonium transmutation capability. Chemical stability of ROX in nitric acid provides a “chemical barrier” against reprocessing. The ROX fuel consists of stable natural-analogous mineral-like phases, which have geological stability, and the leach rate of ROX in water is remarkably low. Consequently, committed individual dose equivalents from disposed spent ROX fuels are lower than those from MOX and UO_2 fuels by more than two orders of magnitude. Though the spent fuel disposal cost in the ROX-LWR system is high, total fuel cycle cost seems to be competitive with conventional UO_2 and MOX fuel cycles.

Introduction

Because of delays in fast reactor programmes, the stockpile of plutonium from nuclear power plants, and in addition military-grade plutonium recovered from dismantled warheads, will amount to some 100 tonnes. The utilisation and disposition of this surplus plutonium is the key issue from the nuclear proliferation and the nuclear fuel cycle points of view. The most realistic way to utilise the excess plutonium is to burn it in existing LWRs in the form of MOX fuel. In the MOX-LWR system, however, the spent MOX fuel still contains large amounts of plutonium, and has a much higher plutonium transmutation rate. As a result, U-free inert-matrix fuel (IMF) is not studied in many countries [1-6].

A kind of IMF, plutonium rock-like oxide (ROX) fuels and their once-through burning in LWRs (ROX-LWR system) have been investigated by the Japan Atomic Energy Research Institute (JAERI) [7]. The ROX fuel consists of chemically stable phases of fluorite (stabilised ZrO_2 (SZR) or ThO_2) and spinel ($MgAl_2O_4$). In this fuel, PuO_2 is solidified in a fluorite phase. With ^{238}U -free matrices, a large part of the plutonium can be burned after irradiation in conventional LWRs. The spent ROX fuel also consists of natural analogous geologically stable phases, and is disposed directly as high level wastes (HLWs) after about 50 years cooling. From the high plutonium burn-up rate and the high stability of the fuel, the ROX-LWR system has proliferation resistance and environmental safety. In all the stages of fuel fabrication, irradiation and disposal, the ROX-LWR system is basically built on conventional technologies and facilities. The economic competitiveness of the ROX-LWR system as compared to UO_2 and MOX fuel cycles is thus anticipated.

Several disadvantages of ROX for use in conventional LWRs as compared with UO_2 and MOX fuels have been discovered [8]. Particularly, ZrO_2 based ROX (Zr-ROX) does not contain a fertile nuclide such as ^{238}U , and the negative Doppler reactivity coefficient becomes very small. The effect of the Doppler coefficient on the behaviour in reactivity insertion accident (RIA) is very important. The excess reactivity decreases considerably due the lack of any fertile nuclide, and this makes power distribution flattening difficult. In addition, the melting temperature of ROX is much smaller than that of UO_2 . Loss of coolant accidents (LOCA) and other thermal-hydraulic behaviours are sensitive to the power peaking factor. Several improvements were studied to solve these difficulties of the ROX fuel. As a fuel material, spinel may have unfavourable irradiation effects such as swelling [9]. To avoid the irradiation effects, a particle dispersed type fuel of fluorite particles in spinel matrix is currently being examined, together with the fuel of fluorite-spinel two phases homogeneous mixture.

In this paper, some results of recent ROX-LWR system R&D activities are presented.

Advantages of ROX fuel

Plutonium transmutation in inert-matrix fuel LWR

The input and transmuted (difference between input and output) amount of plutonium per 1 GWe reactor power per 300 EFPD is shown in Table 1, together with the transmutation percentage (ratio of transmuted/input amount) for a 17×17 type 1100 MWe class PWR core with IMF (Zr-ROX) in comparison with the MOX fuel cases. It seems difficult to realise a full IMF loading core because of severe behaviours in accidents due to small reactivity coefficients [8]. Table 1 therefore considers a partial Zr-ROX loading core and full Zr-ROX cores with UO_2 or ThO_2 additives. Effects of these

Table 1. Input and transmuted amount (tonne/GWe/300 EFPD) of plutonium in ROX and MOX fuelled cores (1 170 EFPD burn-up, corresponding to 45 GWd/t in MOX core)

		W-Pu		R-Pu	
		Input	Transmuted	Input	Transmuted
1/3 Zr-ROX + 2/3 UO ₂ (in 1/3 ROX only)	²³⁹ Pu	0.34	0.25 (75%)	0.27	0.18 (67%)
	Total Pu	0.36	0.16 (43%)	0.46	0.19 (41%)
	²³⁹ Pu		0.33 (99%)		0.26 (98%)
	Total Pu		0.31 (86%)		0.34 (74%)
Zr-ROX (Er) - UO ₂	²³⁹ Pu	0.93	0.85 (92%)	0.80	0.70 (88%)
	Total Pu	0.99	0.69 (69%)	1.36	0.81 (60%)
Zr-ROX-ThO ₂	²³⁹ Pu	0.97	0.94 (97%)	0.80	0.75 (93%)
	Total Pu	1.03	0.82 (79%)	1.37	0.90 (66%)
MOX (once-through)	²³⁹ Pu	0.88	0.56 (63%)	0.99	0.44 (45%)
	Total Pu	0.94	0.30 (32%)	1.69	0.41 (25%)
(recycled once)	²³⁹ Pu	0.76	0.54 (71%)	0.76	0.44 (58%)
	Total Pu	0.81	0.35 (43%)	1.30	0.48 (37%)
(recycled three times)	²³⁹ Pu	0.66	0.52 (79%)	0.61	0.42 (63%)
	Total Pu	0.70	0.40 (57%)	1.05	0.53 (50%)
(recycled five times)	²³⁹ Pu	0.61	0.51 (83%)	0.56	0.42 (75%)
	Total Pu	0.65	0.42 (65%)	0.95	0.56 (59%)

Shaded area: cell calculation

modifications of the Zr-ROX core are discussed later. In all cases, assumed discharge burn-up is about 1 170 EFPD with 18 kW/m average linear heat rate. This corresponds to about 45 GWd/t in the MOX case. In this table, ROX fuel data were obtained with two-dimensional core burn-up calculations, while MOX data was obtained with cell burn-up calculations. For MOX fuel burn-up, plutonium recycle is considered. In the recycling calculation, all the plutonium in the spent fuels of the preceding cycle and an additional necessary amount of “fresh” plutonium to maintain a 1 170 EFPD discharge burn-up are loaded in the core. For plutonium recycled MOX cases, Table 1 shows the total input amount of “fresh” plutonium of all cycles as well as the discharged plutonium amount in spent fuels of the final cycle, as averaged values over 300 EFPD.

In Zr-ROX fuel assemblies in 1/3 Zr-ROX + 2/3 UO₂ core, the plutonium transmutation percentage is extremely high. The advantage of IMF is this high plutonium transmutation rate. The plutonium transmutation rate in full core Zr-ROX cases are also larger than in the full-MOX core, but the percentage decreases with ThO₂ or UO₂ additives. As a result with UO₂ additive, the difference in the plutonium transmutation percentage from plutonium recycled MOX cases becomes small. The full IMF loaded core, however, has an advantage of annual plutonium transmutation amount even with UO₂ or ThO₂ additives, i.e. 1.5-2.0 times as much as MOX core, and about three times larger than in a 1/3 Zr-ROX core.

Chemical stability of ROX

Phase relation studies have been made to clarify phase equilibria and element distributions over phases of simulated fuel and spent fuel systems of SZR (or ThO₂)-corundum (Al₂O₃) [10,11], SZR (or ThO₂)-spinel [12] and SZR (or ThO₂)-spinel-corundum [13]. Table 2 summarises phase relations

Table 2. Phases and elements distribution in simulated ROX and spent ROX fuels

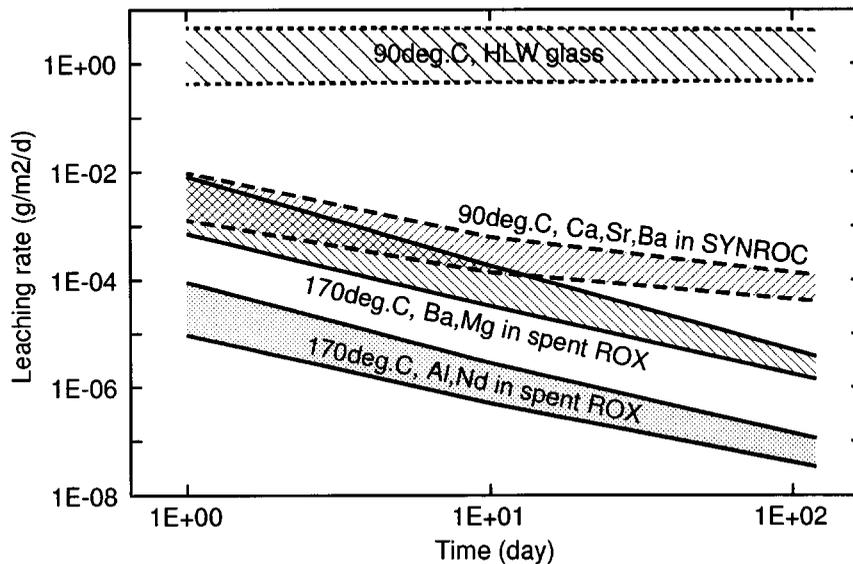
	Fluorite ZrO ₂ (Y,Gd) ThO ₂	Spinel MgAl ₂ O ₄	Corundum Al ₂ O ₃	Hibonite M ²⁺ Al ₁₂ O ₁₉ M ³⁺ M ²⁺ Al ₁₁ O ₁₉	Alloy
Actinides	XX	–	–	–	–
Lanthanides	X	–	–	XX	–
Alkaline-earth	–	X	–	XX	–
Mo, Tc	–	–	–	–	XX
Pt group	–	–	–	–	XX

XX: main distribution, X: secondary distribution, –: not detected

and elements distribution of the main elements in simulated fuels and spent fuels. Plutonium, actinides and lanthanide FPs are distributed in fluorite phases (SZR or ThO₂) by making solid solutions. Noble metals are precipitated as alloys with molybdenum in matrix. With the presence of excess Al₂O₃, alkali (Cs, Rb) and alkaline earth (Mg, Sr, Ba) elements and some rare earth elements form a hibonite-type phase (M⁺Al₁₂O₁₉ and M³⁺M²⁺Al₁₁O₁₉).

The chemical stability of ROX was examined by using simulated spent ROX fuels, which consist of five phases of fluorite, spinel, corundum, hibonite and alloy. Dissolution behaviour of ROX in hot nitric acid solution, and ROX leachability in hot water were observed [14]. In the test in 3 mol% HNO₃ at 420 K for three days, dissolution amounts were 2% for Zr, 30-50% for Al and MG and 60-89% for Ba. The fluorite phase, which is a host phase of plutonium, is remarkably stable against hot nitric acid. It may be said that the ROX fuel provides a “chemical barrier” against reprocessing and plutonium proliferation. The leaching rates of elements in simulated spent ROX in hot water are shown in Figure 1, together with the rates of HLW glass [15] and SYNROC [16]. The leaching rates of the elements in simulated spent ROX are far lower than those of HLW glass, presumably because of the low solubilities of matrix materials in hot water.

Figure 1. Leaching rates of elements in simulated spent ROX in hot water, in comparison with those in HLW glass and SYNROC

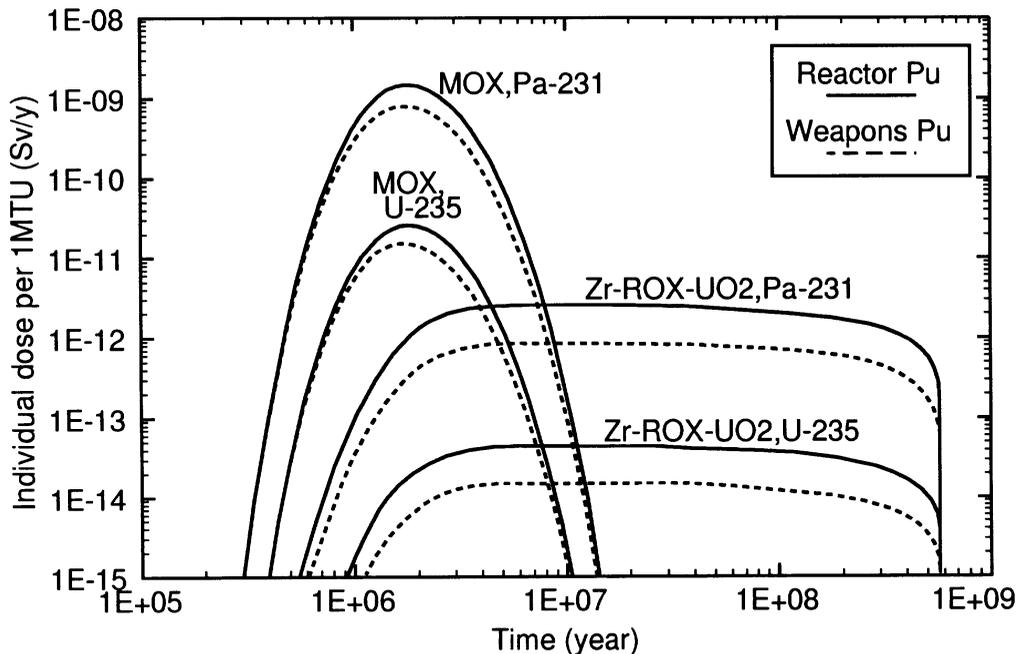


Environmental safety

An environmental safety study was carried out for the direct disposal of spent ROX and MOX fuels [17]. Safety analyses were performed on 1 tonne U equivalent amount of spent ROX and MOX fuels, which in the conventional PWR generated the same amount of energy as 1 tonne of U fuel in the same irradiation time of 1 000 EFPD. The spent fuels are assumed to be contained in a metallic canister, which is placed in a disposal borehole covered with a buffer material. The repository of these radio active wastes is in stable granite rock mass at depths ranging from 500-1 000 m, surrounded by fracture zones connected with biosphere. In this analysis, it was assumed that the lifetime of the canister is 1 000 years, and the release of radioactive nuclides starts 1 000 years after the closure of the repository. The radionuclides then diffuse through the buffer zone, and are transported by the groundwater flow in the granite rock mass and in the fractured zone to the biosphere. The individual dose equivalent due to the exposure pathway ingesting drinking water was calculated. Only the alpha-emitting nuclides were considered here.

Individual dose equivalents due to the geologic disposal of the spent ROX and MOX fuels are shown in Figure 2 for a $4n + 3$ decay chain, which gives the peak dose equivalent both for ROX and MOX spent fuels. In this figure, only important nuclides are shown from a radiological point of view. The peak individual dose due to spent MOX fuels is contributed from ^{231}Pa and about 1.4×10^{-9} Sv/y. The peak dose due to spent ROX fuels, however, is only about 2.4×10^{-12} Sv/y, also from ^{231}Pa . The dose equivalent from spent ROX fuels is less than that from spent MOX fuels by more than two orders of magnitude. The same tendency can be seen in the other radionuclides. This is mainly because the leaching of radionuclides in spent ROX fuels is determined by the dissolution of fuel matrix, SZR, while the leaching in spent MOX fuels is controlled by the solubility limits of radionuclides.

Figure 2. Individual dose from $4n + 3$ decay chain of disposed Zr-ROX and MOX spent fuels



Modifications in ROX fuel and core design

Accident behaviours

For the improvement of Doppler coefficient of ROX, two approaches were considered. One is including additives such as UO_2 or ThO_2 , and the other is a heterogeneous core with ROX and UO_2 fuel assemblies. The additives UO_2 and ThO_2 are also effective for power distribution flattening. In addition, Er_2O_3 burnable poison was examined for power peaking reduction. Original and improved core characteristics of Zr-ROX fuelled PWRs are shown in Table 3.

Table 3. Doppler reactivity ($\% \Delta k/k$) and power peaking factor in Zr-ROX PWR (BOC)

	Doppler reac. (900 \rightarrow 1 200 K)	Peaking factor
Weapons-Pu		
Zr-ROX	-0.098	2.7
Zr-ROX (Er) - 15 UO_2	-0.61	2.1
Zr-ROX - 24 ThO_2	-0.56	2.4
1/3 Zr-ROX + 2/3 UO_2	-0.48	2.8
Reactor-Pu		
Zr-ROX (Er) - 8 UO_2	-0.59	2.2
Zr-ROX (Er) - 18 ThO_2	-0.64	–
UO_2	-0.75	2.0

The core characteristics in Table 3 were used in the analyses of RIA and LOCA. The results of accidents analyses are shown in Tables 4 and 5. In Table 4 it can be seen that in both the UO_2 + Er_2O_3 and ThO_2 added cases, the maximum fuel enthalpy can be less than 230 kcal/kg, the limitation value in the conventional PWR, and the maximum fuel temperature less than the melting point of 2 200 K. The fuel enthalpy is still larger than in a UO_2 PWR. Considering the density of Zr-ROX, i.e. less than 6 g/cm^3 , the energy release of 230 kcal/kg in Zr-ROX might be equivalent to about 120 kcal/kg in UO_2 . The actual enthalpy condition for Zr-ROX pin failure is currently being investigated [18]. In the 1/3 ROX, further improvement seems necessary.

Table 4. Maximum fuel enthalpy (E) and fuel temperature (FCT) in Zr-ROX PWR RIA event

	E (kcal/kg)	E (kcal/cm ³)	FCT (K)
Weapons-Pu			
Zr-ROX	>> 230		>> 2 200
Zr-ROX (Er) - 15 UO_2	193	1.08	1 700
Zr-ROX - 24 ThO_2	224	1.25	1 950
1/3 Zr-ROX + 2/3 UO_2	244	1.36	2 100
Reactor-Pu			
Zr-ROX (Er) - 8 UO_2	192	1.08	1 700
UO_2	93	1.02	2 080

Table 5. Peak cladding temperature (PCT) during LOCA of Zr-ROX PWR

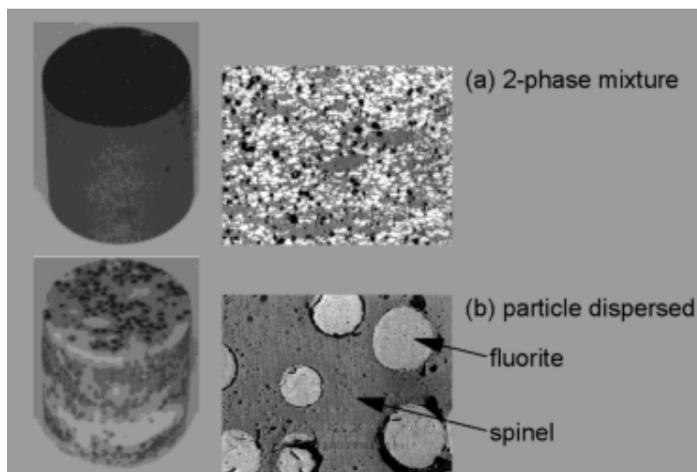
	PCT (K)
Weapons-Pu	
Zr-ROX	> 1 500
Zr-ROX (Er) - 15 UO ₂	1 090
Zr-ROX - 24 ThO ₂	1 240
1/3 Zr-ROX + 2/3 UO ₂	1 240
Reactor-Pu	
Zr-ROX (Er) - 8 UO ₂	1 090
UO ₂	1 080

The LOCA analysis results for modified Zr-ROX fuel cores are shown in Table 5. In all modified Zr-ROX fuel cores, the peak cladding temperature is successfully reduced to less than the limit of 1 470 K (1 200°C). In the Zr-ROX (Er)-UO₂ fuel core, the cladding temperature is lower than the other cores mainly because of the smaller power peaking factor. In the other two modified cores, the peak cladding temperatures are not as low as that in the conventional UO₂ core. Improved LOCA behaviour may be preferable in these cores.

Irradiation behaviour

The spinel phase in the ROX fuel matrix may have unfavourable irradiation effects such as swelling, and to avoid these effects, a particle dispersed type fuel of fluorite particles in spinel matrix is being examined [19], together with the fuel of fluorite-spinel two phases homogeneous mixture. In this particle dispersed type fuel, fluorite phase particles of a few μm diameter, which contain fissile material, are distributed in the spinel matrix. The matrix damaged by fission fragments can be limited to the area within the range of fission fragments (about 8 μm) around particles. Irradiation of test samples of the particle dispersed type fuel has just started to determine the effect on irradiation behaviours.

Figure 3. Fabricated test samples of Zr-ROX pellets of (a) uniform two-phase mixture of fluorite and spinel, and (b) fluorite particles dispersed in spinel matrix



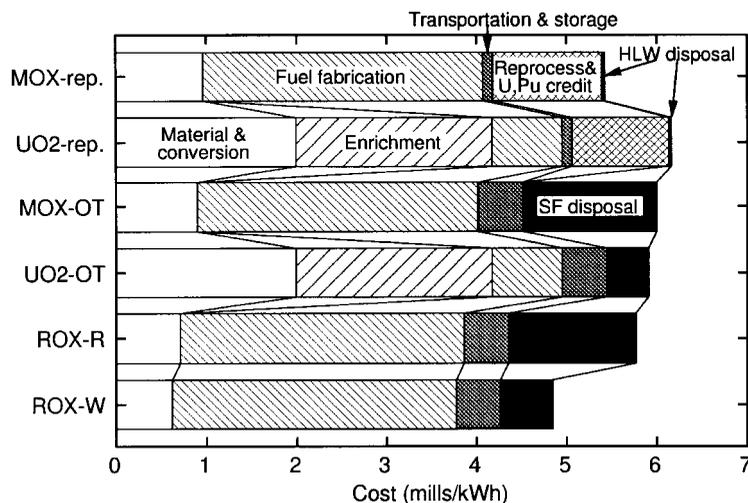
Cost estimation of ROX-LWR system

The ROX-LWR system is based on a conventional technology background. The economic competitiveness of the ROX-LWR system as compared to UO_2 and MOX fuel cycles is anticipated. Fuel cycle cost estimations of the ROX-LWR system have been performed, and the results were compared with UO_2 and MOX cases [20], based on guidelines developed by OECD NEA [21]. In this estimation, the average cost of one 1 GWe class PWR of 45 GWd/t average burn-up was calculated with operation starting in the year 2000 for a 30-year period. Compared cases include:

- MOX LWR + reprocessing (MOX-rep.).
- UO_2 LWR + reprocessing (UO_2 -rep.).
- MOX LWR once-through (MOX-OT).
- UO_2 LWR once-through (UO_2 -OT).
- ROX LWR once-through with reactor-grade Pu (ROX-R).
- ROX LWR once-through with weapons-grade Pu (ROX-W).

It was assumed the price of fissile plutonium is the same for reactor-Pu and weapons-Pu. It was also assumed for ROX calculations that the price of additives such as ZrO_2 , Al_2O_3 , MgO , etc. is 10% of the cost of natural U, and the fuel fabrication cost is equal to that of MOX. For the six cases, estimated fuel cycle costs with the discount rate of 5% are compared in Figure 4. In ROX cases and the other once through systems, the spent fuel disposal cost becomes the lowest in UO_2 -OT, and it is very high in MOX-OT and ROX-R systems. This is because, in the current estimation, the disposal cost is considered proportional to the decay heat of the spent fuels. Thus the decay heat of spent ROX-R is very large, and is almost the same as the MOX-OT system, which also uses reactor-grade Pu. In general, the total fuel cycle cost of the ROX-LWR system seems to be competitive with conventional UO_2 and MOX fuel cycles. With reactor-grade Pu, the total cost and its breakdown become similar to the MOX once-through system.

Figure 4. Cost estimation for various fuel cycle systems (discount rate: 5%)



Conclusion

As an option for the disposition of excess plutonium both from nuclear power plants spent fuels and from dismantled warheads, the feasibility of a system of plutonium rock-like oxide (ROX) fuels and their once-through burning in LWRs (ROX-LWR system) has been investigated. The features of the system can remarkably enhance both the proliferation resistance and the environmental safety:

- *Proliferation resistance*: As the ROX fuel is a kind of inert matrix fuel with ^{238}U -free matrices, it has a high plutonium transmutation capability. As a result, the spent fuels do not contain valuable plutonium nuclides. Chemical stability of ROX fuels is excellent even in nitric acid solution, and they become themselves “chemical barriers” against reprocessing, and hence proliferation.
- *Environmental safety*: The ROX fuel consists of stable natural-analogous minerals, which have geological stability. The leach rates of spent ROX fuels in hot water is remarkably low. In consequence, individual dose equivalents from disposed spent ROX fuels are lower than those from MOX and UO_2 fuels by more than two orders of magnitude.

The ROX-LWR system consists of conventional facilities and equipments based on the present technological background. From the fuel cycle cost estimation of the ROX-LWR system, total fuel cycle cost of the system seems comparable with conventional UO_2 and MOX fuel cycles.

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REACTORS WITH Th/Pu BASED FUELS

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Abstract

The use of (Th,Pu)O₂ fuels for plutonium consumption is reviewed here, both in thermal and fast reactors, for a first recycling of reactor grade or weapons grade plutonium. Mass consumption rates (kg/TWhe), relative consumption rates (% initial loading) and isotopic degradation are assessed. Changes in operation and safety parameters are investigated, as well as the production of fissile uranium bred from thorium. Radiotoxicities vs. time after storage have also been checked.

The overall potential interest of thorium for plutonium burning is established. Plutonium consumption is higher in PWRs but still fairly important in sodium cooled fast reactors. Operation and safety parameters remain within an acceptable range.

Introduction

The revival of interest for thorium fuel cycles is motivated by preoccupations linked to the management of long-lived nuclear waste or otherwise strategic materials, and to the availability of large low-cost energetic resources alternative to uranium. We focus here on plutonium disposition in (Th,Pu)O₂ fuelled reactors, both thermal and fast ones.

In order to limit the growth of plutonium stockpiles, MOX (U,Pu)O₂ fuels are already used in thermal reactors. Up to now, only partial loadings of MOX fuels have been performed (30% MOX loading in 15 French 900 MWe PWRs); then the plutonium build-up in the UOX part of the core is compensated by the consumption in the MOX part. Full MOX PWRs would consume approximately 60 to 70 kg/TWhe plutonium. On the other hand, fast reactor studies (in the frame of the CAPRA programme) have provided a sound design for fast, MOX fuelled, sodium cooled, plutonium burners with a plutonium consumption of approximately 75 kg/TWhe.

In any case, the plutonium consumption in MOX (U,Pu)O₂ reactors is limited by its conversion from ²³⁸U. Hence some uranium-free designs have been examined, with or without inert matrix, and among them, the use of Th/Pu fuels. Thorium could be a good matrix to support plutonium, with a high melting point allowing for large burn-ups and providing some extra Doppler effect due to the resonances of ²³²Th. However, an output for the highly fissile ²³³U bred from thorium has then to be found. For example, it could be used later on to start a Th/U fuel cycle. Moreover, the plutonium consumption in a Th/Pu fuel would be somewhat lesser than in a (Pu + inert matrix) fuel, as the ²³³U bred from ²³²Th will contribute to the fissions.

The most recent calculation tools and databases at the CEA have been used to assess the neutronic performance of (Th,Pu)O₂ fuelled reactors and the associated mass balances. A reactor grade and a weapons grade plutonium are loaded in a (Th,Pu)O₂ fuelled 900 MWe PWR, while two variants have been investigated for fast reactors aiming at either a maximum or minimum ²³³U production besides the Pu consumption. The first one involves an EFR-like core design, the second one a CAPRA-like design. All parameters are compared to similar parameters in standard reactors, i.e. a conventional 900 MWe UOX PWR for PWR studies, and an EFR-like large (1 450 MWe) fast reactor for fast reactor studies.

Calculation tools and methods

The tools used for PWR calculations are the APOLLO-2 [1] cell code for fuel assembly calculation (transport) and burn-up, run on a 2-D model of a 900 MWe PWR standard 17 × 17 assembly, and the CRONOS-2 [2] code for whole core calculations. Nuclear data come from an extended version of the CEA93 library, with an energetic mesh of 172 energy groups covering the 0-20 MeV range. This library is based on the JEF-2.2 datafile, and the extension was to include all the nuclides relevant for thorium fuel cycles in the library. The decay chain contains 28 heavy nuclei from A = 228 (²²⁸Th) to A = 245 (²⁴⁵Cm), 12 being self-shielded (²³²Th, ²³³U, ²³⁵U, ²³⁸U, ²³⁷Np, ²³⁹Pu, ²⁴⁰Pu, ²⁴¹Pu, ²⁴²Pu, ²⁴¹Am, natural Zr). Fission yields for ²³²Th and ²³³U have been taken into account. Radiotoxicities have been computed using the DARWIN/PEPIN [3] code, with dose conversion factors taken from the ICRP-68 recommendation. This code solves the Bateman equations by analytical or numerical methods.

Neutronic calculations for FR cores are performed with the ERANOS [4] code system: cell calculations with the ECCO code, using a 1-D cylindrical model, and core calculations with

appropriate other modules within ERANOS (diffusion approximation, RZ geometry). Nuclear data come from the ERALIB1 file containing data based on the JEF-2.2 file and adjusted over a wide set of experimental data (the main uranium, plutonium and structure nuclides are adjusted). Cell calculations involve a step using the 1 968 energy groups ERALIB1 file and a condensation to the 33-group structure used for core calculations. The decay chain involves 27 heavy nuclides from $A = 231$ (^{231}Pa) to $A = 248$ (^{248}Cm). Sodium void and Doppler calculations involve the fuel zones only.

Core designs

A conventional homogeneous (no zoning) 900 MWe PWR has been used, with an average burn-up at unloading of 33 GWd/t. The fuel is either enriched UO_2 , with 3.25% $^{235}\text{U}/\text{U}$, or $(\text{Th,Pu})\text{O}_2$. The plutonium isotopic compositions used, i.e. spent UO_2 fuel Pu and weapons grade Pu, are shown in Table 1. In all cases, the fissile content is adjusted to ensure a 33 GWd/t burn-up for the spent fuel, by setting $k_\infty = 1.03$ at 22 GWd/t with no boron.

Table 1. Pu isotopic compositions used (w%)

	PWR 900 MWe		FR 1 450 MWe	
	Reactor grade	Weapons grade	EFR	CAPRA
^{238}Pu	1.34	–	1.96	5.55
^{239}Pu	57.38	94.04	52.94	38.95
^{240}Pu	22.67	5.37	25.49	26.71
^{241}Pu	12.61	0.59	9.80	13.06
^{242}Pu	4.73	–	7.84	14.42
^{241}Am	1.27	–	2.00	1.31

Two fast reactor variants with $(\text{Th,Pu})\text{O}_2$ fuel have been used. The first one is close to the European Fast Reactor (EFR) project core design, the second one to the CAPRA [5] design. The main characteristics of these cores, together with an EFR-like core with $(\text{U,Pu})\text{O}_2$ fuel, are shown in Table 2, while the Pu isotopic compositions used are given in Table 1. The CAPRA design involves a dilution of the fuel in the core in order to increase the plutonium content in the fuel. This dilution is performed at three levels : pin (by using small diameter hollow pellets), SA (with a bundle of 336 fuelled pins + 133 unfuelled pins), and core (with 52 unfuelled “diluent” SAs). This allows for a greatly reduced fuel volume fraction in the core, as shown in table 2. A degraded plutonium is used for the CAPRA-like core in order to increase even more the $\text{Pu}/(\text{Th} + \text{Pu})$ content in this core. Both variants have a power output of 1 450 MWe (3 600 Mwth).

Results for Th/Pu PWR cores

The main results are gathered in Tables 3 to 6 below.

The presence of plutonium in the fuel reduces the control rod worth, the soluble boron efficiency and the delayed neutron fraction, and makes the moderator coefficient more negative. A full core calculation with the CRONOS-2 code in the case of reactor grade Pu has shown that the initial amount of soluble boron, 2 080 ppm at 0 GWd/t with no xenon, is acceptable, as well as the calculated peaking factor (1.27). Anyway, core operation and safety studies will have to assess the impact of all these changes, and some modification of the classical control devices may prove necessary.

Table 2. Main characteristics of fast reactor cores

	FR EFR	FR CAPRA
Core height/diameter (cm)	100/405	100/402
SAs in enrichment zones	217/108/72	168/198
Fissile content (w%) (U,Pu)O ₂ cores: Pu/(U + Pu) (Th,Pu)O ₂ cores: Pu/(Th + Pu)	18.3/22.4/26.9 20.5/25.0/30.0	– 43.2/45.0
Core volume fractions (%) Fuel Steel Sodium	37.51 23.52 32.94	21.40 24.79 36.82
Fertile blankets Lower/upper axial (cm) Radial (number of SAs)	25/15 78	– –
Fuel residence time (EFPD)	1 700	990

Table 3. Main neutronic parameters for PWRs (1 pcm = 10⁻⁵ Δk/k')

Fuel type		UO ₂	(Th,Pu)O ₂
Pu grade		–	reactor
Boron efficiency	pcm/ppm	8.9	3.6
Doppler	pcm/°C	-2.6	-3.3
Moderator coefficient	pcm/°C	-15.4	-22.2
100% coolant void	pcm	-70 000	-68 600
β _{eff} 0 GWd/t	pcm	595	279
33 GWd/t	pcm	522	357
Control rod worth	pcm	34 000	23 800

Table 4. Mass balances for PWRs (0 → 33 GWd/t)

Fuel type Pu composition	(Th,Pu)O ₂	
	Reactor grade	Weapons grade
Fissile content (%)	6.5 (Pu)	5.0 (Pu)
0 GWd/t (²³⁹ Pu + ²⁴¹ Pu)/Pu (%)	70.0	94.6
33 GWd/t (²³⁹ Pu + ²⁴¹ Pu)/Pu (%)	47.8	53.8
Mass balances 0 → 33 GWd/t (kg/TWhe)		
Th	-73.36	-67.93
²³¹ Pa	+0.44	+0.45
U + ²³³ Pa	+52.21	+61.37
²³⁷ Np	+0.01	+0.00
Pu + ²³⁸ Np + ²³⁹ Np	-117.63	-128.02
Am	+3.94	+1.94
Cm	+2.99	+0.69
MA (²³¹ Pa + ²³⁷ Np + Am + Cm)	+7.38	3.08
Conversion factor	0.631	0.497
Pu consumption (% initial loading)	47.1	65.8

Table 5. U and Pu isotopic compositions at 33 GWd/t

Fuel type Pu composition	(Th,Pu)O ₂	
	Reactor grade	Weapons grade
²³² U	0.35	0.42
²³³ U	90.96	88.38
²³⁴ U	7.51	9.55
²³⁵ U	1.11	1.66
²³⁶ U	0.07	0.00
²³⁸ Pu	3.21	0.43
²³⁹ Pu	25.06	30.02
²⁴⁰ Pu	35.52	37.60
²⁴¹ Pu	22.73	23.76
²⁴² Pu	13.49	8.19

Table 6. (Th + reactor grade Pu) spent fuel radiotoxicity, compared to standard UO₂ spent fuel

10 ² years	10 ³ years	10 ⁴ years	10 ⁵ years	10 ⁶ years
× 3.2	× 2.7	× 2.2	× 10	× 3.6

Capture cross-sections are higher for ²³²Th than for ²³⁸U. As a consequence, the initial fissile content is higher with ²³²Th based fuels than with ²³⁸U based fuels, and the conversion factor is higher also. It is reminded, to allow comparisons, that the conversion factor for a standard 900 MWe PWR with enriched UO₂ (3.25% ²³⁵U/U) is 0.475 and that the minor actinide production in a standard 900 MWe PWR is 3.34 kg/TWhe.

The Pu relative consumption is rather high, and would be increased with higher fuel burn-ups; one could expect a Pu consumption of ≈73% (reactor grade Pu) or ≈84% (weapons grade Pu) after a 60 GWd/t burn-up.

Similarly, the Pu isotopic degradation is very effective, and will be even more important with higher burn-ups; one could expect some 30-35% (²³⁹Pu + ²⁴¹Pu)/Pu after a 60 GWd/t burn-up. The question of further recyclings of such a degraded plutonium in reactors, mixed with some “fresh” UO₂ spent fuel Pu topping, has not been examined.

The uranium produced is a high grade reactor fuel, and enough uranium would be produced to start a (Th,U)O₂ 900 MWe PWR core after ≈9 years of operation of a (Th,Pu)O₂ 900 MWe PWR core. Some of the daughter products of ²³²U have very hard γ-ray emissions, such as ²¹²Bi (1.8 MeV) and ²⁰⁸Tl (2.6 MeV). This means that the significant amounts of ²³²U in the spent fuel uranium would make its use as a weapons material more difficult than for ²³⁵U or weapons grade Pu, despite a rather low critical mass (≈15 kg). It is worth noting that the presence of ²²⁸Th, belonging to the same decay family, makes the spent thorium highly active for ≈10 years after irradiation.

Radiotoxicities with respect to UO₂ spent fuel are increased in a similar way as for (U,Pu)O₂ MOX fuel. Radiotoxicity reductions could be expected only with the use of a (Th, ²³³U)O₂ fuel.

Results for Th/Pu FR cores

The main results are gathered in Tables 7 to 12 below.

Table 7. Main neutronic parameters for FRs (1 pcm = 10⁻⁵ Δk/k')

Core type Fuel		EFR (U,Pu)O ₂	EFR (Th,Pu)O ₂	CAPRA (Th,Pu)O ₂
Δk/kk' (BU)	pcm/EFPD	-7.98	-7.09	-25.26
Δk/kk' (Na void)	pcm	+2 351	+1 508	+1 504
Doppler constant	pcm	-695	-616	-673
Max. linear rating	W/cm	425	485	430
Max. BU	at%	19.5	21.1	21.6
Max. damage	dpa	189	189	111

Table 8. Sodium void perturbation splittings (pcm)

Core type Fuel	EFR (U,Pu)O ₂	EFR (Th,Pu)O ₂	CAPRA (Th,Pu)O ₂
Component-wise			
Non-leakage	+3 112	+2 368	+2 521
Leakage	-761	-860	-1 017
Region-wise			
Inner core	+1 748	+1 169	+1 032
Middle core	+540	+352	-
Outer core	+63	-13	+472

Table 9. Mass balances (kg/TWhe) from BOL to EOL + 5 years

	EFR (U,Pu)O ₂			EFR (Th,Pu)O ₂			CAPRA (Th,Pu)O ₂
	core	ax. bl.	rad. bl.	core	ax.bl	rad. bl.	core
Th	-	-	-	-68.37	-17.37	-11.34	-37.88
Pa	-	-	-	+0.22	+0.03	+0.02	+0.11
U	-79.41	-18.71	-12.27	+38.42	+13.08	+8.73	+25.20
Np	+0.31	+0.05	+0.04	+0.05	≈0	≈0	+0.06
Pu	-23.04	+14.28	-9.51	-75.29	≈0	≈0	-100.42
Am	+6.00	+0.02	+0.01	+6.54	≈0	≈0	+12.70
Cm	+0.54	≈0	≈0	+0.52	≈0	≈0	+1.28
Total	-95.60	-4.36	-2.71	-97.91	-4.26	-2.59	-98.93

Table 10. Relative Pu consumptions (% initial loading) and isotopic degradations between BOL and EOL + 5 years

Core type	EFR	CAPRA
BOL (²³⁹ Pu + ²⁴¹ Pu)/Pu (%)	62.7	52.0
EOL (²³⁹ Pu + ²⁴¹ Pu)/Pu (%)	43.1	36.5
Pu consumption (% initial loading)	44.1	36.3

Table 11. Pu isotopic compositions at EOL + 5 years (w%)

Core Fuel	EFR U/Pu	EFR Th/Pu	CAPRA Th/Pu
²³⁸ Pu	1.70	2.91	5.46
²³⁹ Pu	59.14	36.04	27.65
²⁴⁰ Pu	27.76	41.12	37.82
²⁴¹ Pu	4.23	7.04	8.88
²⁴² Pu	7.18	12.88	20.18
²⁴¹ Am	—	—	—

Table 12. U isotopic compositions at EOL + 5 years (w%)

Core Fuel	EFR Th/Pu	CAPRA Th/Pu
²³² U	0.23	0.22
²³³ U	92.93	90.28
²³⁴ U	6.22	8.60
²³⁵ U	0.51	0.65
²³⁶ U	0.10	0.25

The large reactivity loss in the CAPRA-like core is the main reason for its lower fuel residence time. The Doppler feedback is of similar amplitude in the three cores. This is due to the good resonance properties of ²³²Th and to the slightly softer spectrum in the CAPRA-like core.

The sodium void value is reduced by 1/3 in the (Th,U)O₂ cores. This is mainly due to the neutronic properties of ²³²Th and ²³³U.

The fission threshold for ²³²Th is located at a higher energy than for ²³⁸U and, at a given energy, the fission cross-section of ²³²Th is lesser than that of ²³⁸U, so that the spectrum hardening associated with sodium voiding introduces less positive reactivity.

The $v\sigma_f/\sigma_a$ ratio for ²³³U increases much less with energy than for ²³⁹Pu. Once again, the sodium voiding brings about less positive reactivity.

In the case of the CAPRA-like core, the degradation of the isotopic composition of plutonium is compensated by the increase in leakage due to the fuel dilution.

All these trends are confirmed by the splitting of sodium void reactivity by means of a perturbation theory analysis: decrease of the non-leakage component when thorium is used, increase of the absolute value of the leakage component in the CAPRA-like core.

The protactinium effect (decay of ²³³Pa with a 27 day half-life on ²³³U) would increase the core reactivity by ≈1 300 pcm, which is roughly 10 times the standard neptunium effect of (U,Pu) cores (decay of ²³⁹Np with a 2.36 day half-life on ²³⁹Pu). The effective delayed neutron fraction β_{eff} is close to that of a (U,Pu) core, but as ²³²Th is less fissile than ²³⁸U the relative contribution of ²³²Th to β_{eff} in (Th,Pu) cores (15%) is much lesser than that of ²³⁸U in a standard (U,Pu) core (40%).

The EFR-like (Th,Pu)O₂ core, with blankets, produces large amounts of uranium, enough in fact to be able to start an EFR-like (Th,U)O₂ core after 15 years of operation. A self-sustaining (Th,U)O₂ large fast reactor core can then be designed, relying only on thorium feeds.

The plutonium consumption, 75 kg/TWhe, is similar to that of a reference (U,Pu)O₂ CAPRA core [5], but with a lesser Pu content in the fuel (30% maximum instead of 45%). This plutonium consumption is increased by 1/3 in the CAPRA-like core, and simultaneously the uranium production in the core is reduced by 1/3 with respect to the EFR-like (Th,Pu)O₂ core (blankets not included). The relative consumptions and isotopic degradations are given in Table 10. Further recyclings of this degraded Pu with PWR plutonium toppings in a (Th,Pu)O₂ cores have not been investigated.

Tables 11 and 12 detail the isotopic compositions of uranium and plutonium in the spent fuel. The uranium produced has a higher fissile content in the EFR-like core because the lower grade uranium coming from the core is assumed to be blended with the higher grade uranium coming from the blankets. The global fraction of ²³²U/U is of the same order of magnitude as that for PWR spent fuel. However, it can be seen that the uranium from the upper axial blanket (i.e. with the lesser burn-up), produced at the rate of 3.65 kg/TWhe (average over 1 700 EFPD) has a ²³³U content of 98% and a ²³²U content of only 0.07%. With a smaller irradiation time, the ²³²U content would be much lesser; this can be better achieved with radial blanket SAs or even fertile ThO₂ SAs placed within the core.

The use of (Th,Pu)O₂ fuels with weapons grade plutonium has not been investigated in these large fast reactors; however, calculations were performed on a smaller reactor design, similar to the Phénix reactor (250 MWe), with a fuel residence time of 540 EFPD, i.e. a maximum burn-up of roughly 8%. As for plutonium consumption, the following results were obtained. The plutonium consumption was 82 kg/TWhe, representing 23% of the initial loading. The (²³⁹Pu + ²⁴¹Pu)/Pu content was lowered from 95% to 88% only.

Conclusions

The use of a (Th,Pu)O₂ fuel in PWRs allow a high plutonium consumption, either for mass consumption (i.e. kg/TWhe) or relative consumption (i.e. % of the initial loading), and with an important degradation of the Pu isotopic compositions. The relative consumption and isotopic degradation could be easily improved by increasing the spent fuel burn-up. The values obtained here for a 33 GWd/t burn-up are:

	kg/TWhe	% initial loading	% (²³⁹ Pu + ²⁴¹ Pu)/Pu
Reactor grade Pu	-118	-47	70 → 48
Weapons grade Pu	-128	-66	95 → 54

The neutronic characteristics remain in the usual range, although there is a worsening of the delayed neutron fraction, the boron efficiency and the control rod worth that may need some core adjustment.

The large amount of uranium produced: ≈50-60 kg/TWhe, mostly ²³³U, could be used to feed (Th,U)O₂ reactors, either thermal or fast ones. For example, enough uranium to start a 900 MWe (Th,U)O₂ PWR (≈960 kg) can be built up by operating a 900 MWe (Th,Pu)O₂ PWR for 9 years. The significant proportion of ²³²U would make the use of this uranium as a weapons material more difficult than for ²³⁵U or Pu.

In fast reactors, (Th,Pu)O₂ fuels have been investigated along two main lines: Pu consumption together with a high U production to start a (Th,U) cycle, or increased Pu consumption and reduced U production. The main results for a ≈20 at% maximum burn-up are:

	kg/TWhe U	kg/TWhe Pu	% initial loading	% (²³⁹ Pu+ ²⁴¹ Pu)/Pu
High U production	+60	-75	44	63 → 43
Low U production	+25	-100	36	52 → 37

²³²Th and ²³³U allow to reduce the sodium void reactivity by 1/3 with respect to a conventional (U,Pu)O₂ core design, while the Doppler feedback is almost kept constant. Delayed neutron fractions similar to a standard (U,Pu)O₂ core have been found, but there is a large “protactinium effect” of 10 times the well-known neptunium effect in (U,Pu)O₂ cores.

With the high U production option, the large amount of uranium produced (≈60 kg/TWhe), mostly ²³³U, could be used to feed (Th,U)O₂ reactors. For example, enough uranium to start a large (Th,U)O₂ fast reactor of 1 450 MWe power (≈8.5 t) can be built up by operating a 1 450 MWe (Th,Pu)O₂ fast reactor for 15 years. The significant proportion of ²³²U would again make the use of this uranium as a weapons material more difficult than for ²³⁵U or Pu, except that with fast reactors the blanket SAs offer more flexibility to breed high grade weapons material at low burn-ups.

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PRELIMINARY EVALUATION OF A BWR WITH CERMET FUEL CORE LOADING

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Abstract

Although there are no boiling water reactors (BWRs) in France, evaluation studies of this type of reactor are underway at the CEA in the framework of the Innovation Programme. These studies, whose purpose is to better define the safety, operational behaviour and fuel cycle capacity of the system, and to generally improve understanding of its physics, inevitably entailed adapting the CEA's calculation tools, qualified for PWRs, to BWRs. With respect to the cycle aspect, studies have been initiated to evaluate the capability of this system to use MOX fuel. A very preliminary outline of the study extended to composite fuels is presented here.

Introduction

Electricity production in France is ensured by 58 pressurised water reactors (PWRs); no boiling water reactor has been built on French territory. A number of our European neighbours, however, possess reactors of this type, which represent one-third of world-wide nuclear-produced electricity. This type of reactor has certain features and assets that have to be better defined to enable highlighting its potential advantages with respect to given aspects of the cycle.

In particular, the capability of BWRs of using MOX fuel represents a potential advantage for solving the problem of balancing the stocks of Pu and irradiated fuels awaiting reprocessing. A PWR-BWR comparison phase and refining of the calculation scheme are currently under development. This work, using conventional CEA computer calculation codes qualified for PWRs, includes an evaluation of BWR potential for using MOX fuels.

In view of the fact that studies are underway on the evaluation of the behaviour of composite fuels with Pu in PWRs [1-6], we shall merely make a preliminary examination of the behaviour of a Pu CERMET fuel in a BWR, using a very simplified calculation scheme.

The study

The study was performed using the APOLLO2 computer code for a cell in an infinite environment.

The reference fuel used in the studies focused on PWRs is a CERMET (PuO₂,Zry) containing a volume fraction of 8% PuO₂ CERamic, which itself has a mass fraction of 2.3% Er₂O₃, needed to control the potential overpower permitting a cycle duration of 18 months.

We used the same fuel for the BWR, although more refined calculations tend to show that a 7.5% PuO₂ content would be enough to last through the 18 months and ensure a discharge burn-up of 600 GWd/t (48 GWd/T equivalent UO₂).

If the PWR assembly is homogeneous from the spatial and spectral points of view, the same is not true of BWRs (Figure 1). To take this heterogeneity into account in cell calculations, we had to define a specific procedure, assuming that the equivalent moderation ratio (MReq) is equal to the geometric moderation ration (MRgeo), corrected by a factor that takes the “quality” of the moderation into account, together with the two-phase aspect of the moderator.

That is to say:

$$MReq = MRgeo \times (1 - \gamma\alpha)$$

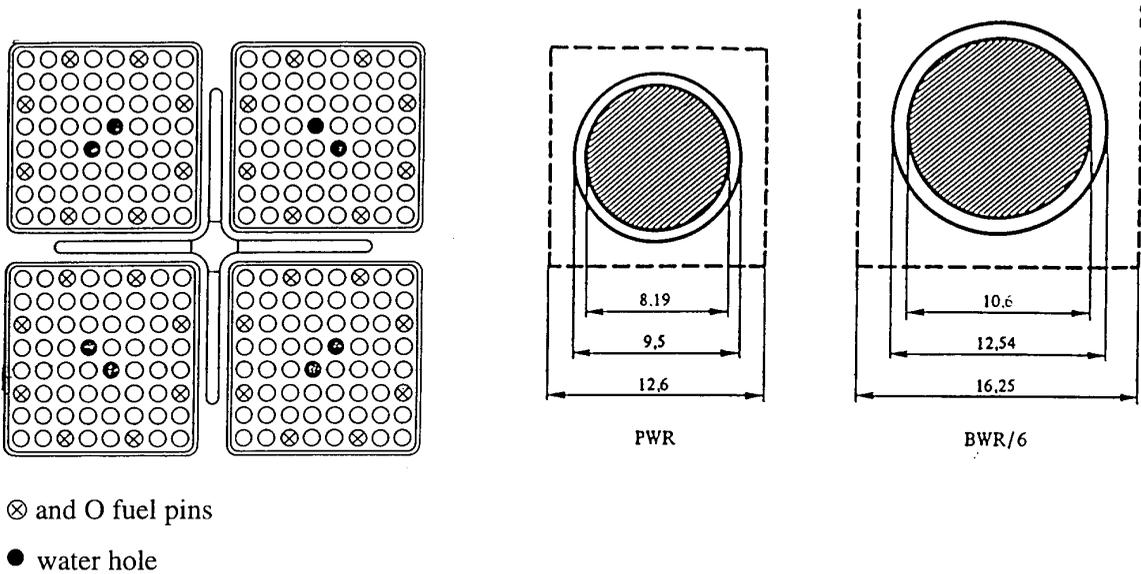
where: γ = volume of two-phase water/total volume of water (= 1 in the cell, and $\neq 1$ in the assembly)
 α = average void fraction (40% here)

The following relationship is established:

$$(MRgeo)_{cel} = (MRgeo)_{ass} \times (1 - \gamma\alpha)/(1 - \alpha)$$

The geometric correction being made, the APOLLO2 calculations were performed.

Figure 1. Cell and assembly of a BWR/6



Results

Table 1 compares the Doppler and draining coefficients for the academic situation (for a PWR) of a 40% void fraction. The values in parentheses in the table come from a calculation for a BWR assembly performed using less simplistic assumptions and a more sophisticated calculation scheme for a MOX situation, showing the validity of the simplified calculations, which prove sufficient for providing realistic tendencies.

Table 1. Doppler and draining coefficient

Boron worth: PWR = -2.36 pcm/ppm, BWR = -1.48 pcm/ppm

Doppler coefficient (pcm/°C)

	BOL	EOL
PWR	-1.10	-1.52
BWR	-1.27	-2.44 (-2.76)

Draining coefficient (pcm/%)

	BOL	EOL
PWR ($\alpha = 40\%$)	-89	-368
BWR ($\alpha = 40\%$)	-117 (-110)	-449

It should be noted that the Doppler coefficient is better (BOL + 15%, EOL + 65%) in the case of the BWR for this type of fuel, which is an important safety factor able to partially compensate the low effective beta of the Pu CERMET. The higher draining coefficient (in absolute values) for the BWR shows that the drained situation would be easier to control than in a PWR with this type of fuel. The low boron efficiency value shows that greater boration will have to be envisaged for the safety injection. It should be noted, however, that this implies only minor changes in operation since soluble

boron is not used in normal operation. Nevertheless, an analysis using a suitable calculation scheme will have to be made with respect to the efficiency of the rod cluster control assemblies since these do play a determining role in operation.

Table 2 presents Pu consumption together with the production of minor actinides.

Table 2. Pu consumption and MA production

Pu consumption (% of initial Pu)

	EOL PWR	EOL BWR
²³⁸ Pu (2.74)	2.09	1.85
²³⁹ Pu (56.54)	4.80	2.84
²⁴⁰ Pu (26.04)	14.44	13.90
²⁴¹ Pu (7.41)	8.87	7.13
²⁴² Pu (7.28)	9.23	10.45
Tot Pu (100)	39.43	36.16

MA production (% of initial Pu)

	BOL	EOL PWR	EOL BWR
²⁴¹ Am	0.7	0.58	0.46
²⁴³ Am	0.	2.35	2.35
²⁴² Cm	0.	0.31	0.32
²⁴⁴ Cm	0.	1.78	1.59
Tot AM	0.7	5.04	4.73

The more efficient consumption of ²³⁹Pu and ²⁴¹Pu for the BWR, accompanied by a slight increase in the ²⁴²Pu content, should be noted. The BWR is a ²⁴¹Pu consumer compared to the initial content, whereas the PWR is a producer. The difference in total consumption is not very great, but notable (~ -7%).

Extrapolated to all of a country's NPPs, this difference results in a significant reduction in the quantities of stored Pu.

The same tendency can be noted with respect to the production of actinides, with an overall lower production (-6%) for the BWR, which, in addition, consumes ²⁴¹Am with greater efficiency than the PWR, and produces less ²⁴⁴Cm than the PWR.

Conclusion

There are no fundamental differences for this type of fuel between a PWR and a BWR with respect to Pu consumption as a result of the absence of ²³⁸U, which cannot play a role in the spectral modification of the two reactors as it does with UOX or MOX fuel loading. However, a lesser consumption of Pu can be observed for the BWR (-7%), accompanied by a more favourable minor actinide production, and a higher Pu fissile isotopes consumption than that of the PWR (-6%). Tendencies for the Doppler and void coefficients and soluble boron efficiency show that this fuel proves to be no "worse" than in a PWR and is perhaps better. However, a calculation using a more accurate scheme should provide elements which will enable to appreciate the efficiency of the control rod clusters.

In a more heterogeneous view of assemblies, the APA [7-11] concept has shown that loading a PWR core with Pu-containing composites was possible and contributed to balancing the Pu stream while assuring acceptable kinetic coefficients. In view of the very similar behaviour of this fuel in a PWR and in a BWR, a transposition of the APA concept for BWRs should permit Pu CERMET loading in these cores.

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REACTOR PHYSICS ANALYSIS OF PLUTONIUM ANNIHILATION AND ACTINIDE BURNING IN CANDU[®] REACTORS

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Abstract

High neutron economy allows CANDU reactors to operate with minimum fissile content. On-power refuelling and simple fuel bundle design enable existing CANDU reactors to use advanced plutonium-based fuel cycles in the whole core, without requiring major modifications. These unique capabilities distinguish CANDU reactors as the most efficient thermal reactors for plutonium annihilation and actinide burning, using an inert-matrix, non-fertile material such as silicon carbide (SiC) as the carrier. Both weapons-derived excess plutonium and plutonium mixed with minor actinides derived from the reprocessing of light-water reactor (LWR) spent fuel can be disposed of safely and efficiently in existing CANDU reactors. The absence of fertile material in the inert-matrix fuel prevents the formation of any additional plutonium. Over 97% of the initial fissile plutonium would be destroyed, compared with about 50% if conventional mixed oxide (MOX) fuel was used. Detailed full-core fuel-management simulations demonstrate that the PuSiC fuel will operate within existing safety limits established for the natural-uranium fuel.

CANDU[®] is a registered trademark of Atomic Energy of Canada Limited (AECL).

Introduction

CANDU reactors have unsurpassed flexibility to accommodate different fuels and fuel cycles. This flexibility is a result of the following key design features:

- High neutron economy, which is due to the use of heavy water as coolant and moderator, the use of low neutron absorbing structural materials, and on-power refuelling permits the initial fissile content in the fuel to be burned down to low levels.
- On-power refuelling provides flexibility in fuel management. Both axial and radial power distributions can be shaped and controlled, as can the amount of reactivity added to the reactor during refuelling.
- The simple fuel bundle design facilitates optimisation of the fuel composition. Fuel material and fissile content can be varied from ring to ring to achieve design objectives, such as tailoring reactivity coefficients, or minimising linear element ratings. The simple, compact fuel bundle also results in extremely low fuel cycle costs with natural uranium fuel, and should reduce the costs for advanced fuels, including those involving remote fabrication.

A conventional approach for the disposition of excess weapons-derived plutonium would be to introduce it into existing CANDU reactors as MOX fuel. This option is readily available and would enable a quick start to the disposition of weapons-derived plutonium. However, the flexibility of CANDU reactors enables the consideration of more advanced options that result in near-complete destruction of the fissile plutonium, if that is the strategic objective. One of these options is the use of an inert-matrix, non-fertile material as the carrier for the weapons-derived plutonium. The absence of ^{238}U prevents the formation of ^{239}Pu , and over 97% of the initial ^{239}Pu is destroyed. A variant of this option is the annihilation of the plutonium and other minor actinides derived from the reprocessing of spent LWR fuel, which is another embodiment of the synergism between CANDU reactors and LWRs.

Assessment of potential candidates for inert-matrix material

All potential candidates for inert-matrix material must have a very low neutron absorption cross-section. They should also have high thermal conductivity, high thermal capacity and high melting temperature. The list of candidates under consideration in Europe, the United States and Canada includes SiC, MgAl_2O_4 (spinel), ZrSiO_4 (zircon), ZrO_2 , CeO_2 , and BeO. Table 1 shows some of the relevant thermal properties of these candidates.

AECL has performed extensive studies to assess the relative merits of these potential inert-matrix material candidates. On the basis of these assessments, AECL is focusing its efforts on SiC as the most promising candidate for inert-matrix applications. Its very high thermal conductivity will result in very low temperatures, both in normal operation and in postulated accidents, with the expected benefit of low fission-gas release. Its high melting temperature is also a benefit. There do not appear to be any long-lived activation products resulting from its irradiation, and it would appear to be a stable waste form. Simulated irradiation performance, and other tests, are positive to date. A great deal of further work must be done to assess SiC and other candidates. If other materials are shown to be superior, they too can be used for actinide burning or plutonium annihilation in CANDU. Another paper in this workshop describes AECL assessments of the fuel aspects of inert matrix carriers [1].

Fuel-management flexibility in CANDU reactors

The on-power refuelling feature of CANDU reactors allows operation with 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 also be controlled by fuelling each region at a different rate. Local power peaking can be minimised by judiciously selecting individual channels to be refuelled and by limiting the number of fresh fuel bundles inserted into a channel at each refuelling operation.

Detailed full-core fuel-management studies were conducted using the RFSP [2] finite-reactor code to assess the power distributions and reactivity effects of PuSiC fuel in an existing CANDU 6 reactor. The lattice code WIMS-AECL [3] was used to determine the optimum amount of plutonium and actinides in the bundle using the CANFLEX[®] fuel bundle design.

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 optimise the reactor global power shape. Figure 2 shows the configuration of the fuel elements in a CANFLEX fuel bundle. Plutonium and minor actinides are restricted to the 35 elements in the two outer fuel rings. The inner eight elements contain only SiC and a small amount of gadolinium.

Gadolinium was used to suppress the excessively high reactivity of the fresh fuel. Coolant void reactivity is negative. There is a high degree of flexibility in the choice of the amount of plutonium and minor actinides in the outer elements and in the amount of gadolinium in the inner elements in order to achieve specific goals, such as reactor power distribution, coolant-void reactivity, and plutonium and actinide destruction efficiencies. The combination of on-power fuelling with a simple and flexible fuel bundle design offers many possibilities for plutonium annihilation and actinide burning in existing CANDU reactors.

Composition of the PuSiC fuel bundles

The weapons-derived PuSiC fuel bundle contains 210 g of plutonium and 60 g of gadolinium. The central element contains 20 g of gadolinium, and the remaining 40 g of gadolinium is distributed uniformly over the seven elements in the next ring. Each of the outer two fuel rings contains 105 g of plutonium mixed with SiC. The plutonium content per element in the outermost fuel ring is two-thirds of that in the third fuel ring. This differential enrichment scheme improves the element power distribution and reactivity effect over the lifetime of the fuel bundle. The initial material composition of the weapons-derived PuSiC fuel bundle is given in Table 2.

For burning of plutonium and minor actinides obtained from reprocessing of spent LWR fuel, the chosen composition consists of 356 g of plutonium plus 66 g of minor actinides in the two outer fuel rings and 20 g of gadolinium in the two inner rings. The central element contains 10 g of gadolinium and the remaining 10 g of gadolinium is distributed uniformly over the seven elements in the next ring. Each of the two outer fuel rings contains 178 g of plutonium and 33 g of minor actinides mixed with SiC. The plutonium and minor actinide content per element in the outermost fuel ring is two-thirds of that in the third fuel ring. The ratio of minor actinides to plutonium is about 50% greater

than that in the unadjusted spent PWR fuel. The composition corresponds approximately to spent PWR fuel having an initial enrichment of 3.5%, and a burn-up of ~35 000 MWd/te. The initial material composition of the civilian PuSiC actinide fuel bundle is given in Table 3.

Lattice reactivity of the PuSiC fuel

Figure 3 shows the variation of the lattice k_{∞} as a function of the total energy produced in the PuSiC bundle for both the weapons-derived plutonium bundle and the civilian-plutonium actinide bundle, with and without gadolinium. Although natural UO_2 fuel is not used in this study, its k_{∞} versus bundle burn-up characteristics are also shown in this figure for comparison purposes.

It is clear that the k_{∞} of the PuSiC fuel lattice begins at a much higher value than that for fresh natural UO_2 fuel and that it decreases very rapidly as the initial fissile material is depleted. This excessive rate of reactivity depletion, especially for the weapons-derived plutonium fuel, would have produced unacceptably high reactivity and power perturbations in the channels being refuelled. It is necessary to use a relatively large amount of gadolinium in the weapons-derived PuSiC bundles in order to minimise the refuelling perturbations and to achieve acceptable global and local power distributions. Fuel burn-up penalty is not a concern in the weapons-derived PuSiC option because the objective is to maximise the rate and degree of destruction of the plutonium.

In the civilian PuSiC fuel bundle, on the other hand, the energy output from the plutonium and the minor actinides is an important parameter. Only the minimum amount of gadolinium is used. The reactivity of the fuel is held down by adding a relatively large portion of minor actinides to the plutonium-actinide fuel mix.

Coolant-void reactivity is -6 mk for the weapons PuSiC option and -4 mk for the civilian PuSiC option. The power coefficient would also be negative for both options. There would be no power pulse in a postulated loss-of-coolant accident (LOCA), and the safety and licensing analyses would be greatly simplified. The fuel-temperature coefficient is very slightly positive, about 12 $\mu k/^{\circ}C$. However, the sign of the fuel-temperature coefficient is not relevant with this fuel: an increase in fuel power would result in a very small increase in fuel temperature because any increase in heat in the fuel would immediately be transferred to the coolant because of the high thermal conductivity of the SiC. The increase in coolant temperature would reduce the coolant density and produce a negative reactivity feedback because of the negative coolant-void reactivity effect.

Full core fuel management simulations

Detailed, realistic fuel management simulations were performed using RFSP for a standard CANDU 6 reactor using either weapons-derived PuSiC or civilian PuSiC actinide fuel in the whole core. Acceptable bundle and channel powers were obtained for both cases using a bi-directional, two-bundle shift refuelling scheme. The refuelling rate per full-power day is 15 bundles for the weapons-derived PuSiC fuel and 9.5 bundles for the civilian PuSiC actinide fuel. These fuelling requirements are within the capability of the current fuel handling system.

Figure 4 shows that the time-average channel power distributions for both PuSiC options are very similar to that in the present natural UO_2 fuelled CANDU reactor. Figure 5 shows the time-average bundle power distributions in a central channel, normalised to a channel power of 6.5 MW.

Bundle powers are high at the inlet end for the PuSiC options because of the rapid depletion of the fissile plutonium. This axial power shape, which peaks towards the inlet (refuelling) end of the channel, will also result in improved thermalhydraulic performance (higher critical channel powers). The weapons-derived PuSiC option gives the highest time-average bundle power, 910 kW, compared with about 800 kW for the natural UO₂ case. In all cases, the maximum instantaneous channel power is less than 7.1 MW, and the maximum instantaneous bundle power is less than 990 kW. The fuel temperatures would be very low, even at these relatively high bundle powers, because the thermal conductivity of the SiC matrix is many times greater than that for UO₂.

Destruction efficiency of plutonium and minor actinides

Table 4 shows the plutonium and minor actinide content in fresh and in discharged PuSiC bundles for weapons-derived plutonium fuel and for civilian-plutonium actinide fuel. The discharge fuel burn-up is 145 MWd per bundle for the weapons-derived PuSiC fuel and 229 MWd per bundle for the civilian PuSiC fuel. In comparison, the fuel burn-up of a natural uranium CANDU fuel bundle, which contains 133 g of ²³⁵U is about 136 MWd per bundle.

The disposition rate of the weapons-derived PuSiC option is about 1.8 Mg of weapons-derived plutonium per GW(e)Y. About 75% of the total initial plutonium inventory and 97% of the initial ²³⁹Pu are destroyed. Two CANDU 6 reactors can therefore almost completely destroy 50 Mg of weapons-derived plutonium in less than 25 years.

The civilian PuSiC option gives a net destruction rate of 1.2 Mg of plutonium and 0.13 Mg of minor actinide per GW(e)Y. About 97% of the initial ²³⁹Pu and 88% of the initial ²⁴¹Am are destroyed; the net destruction efficiency of the minor actinides is 38%. Light-water reactors produce about 268 kg of plutonium and about 33 kg of minor actinides per GW(e)Y. One CANDU 6 reactor can effectively destroy much of the plutonium and minor actinides produced in three 1 GW(e)-size LWRs.

There is scope for further optimisation of the civilian-plutonium minor-actinide option. All or some of the minor actinides could be segregated into elements in the centre of the bundle. It is conceivable that after the first pass through the reactor, these separate elements are removed and irradiated in the moderator (for example, in the adjuster rod units) for an extended period, significantly increasing the net destruction efficiency of the minor actinides. These and other options will be the subject of further studies.

Conclusions

Good neutron economy and the use of on-power refuelling enable existing CANDU reactors to achieve near-complete annihilation of excess weapons-derived plutonium using an inert-matrix fuel. This approach can also be used to annihilate plutonium and destroy minor actinides derived from reprocessed spent LWR fuel. The flexibility of the CANDU design allows the plutonium inert-matrix fuel cycle to be optimised to meet specific goals. For example, the weapons-derived plutonium option can be designed to maximise plutonium destruction rate and the civilian-plutonium actinide option can be designed to extract maximum energy from the plutonium and the minor actinides. Detailed fuel-management simulations conclude that existing CANDU reactors can operate with a full-core of plutonium inert-matrix fuel.

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Table 1. Physical properties of inert-matrix materials

MATERIAL	MELTING TEMPERATURE	THERMAL CONDUCTIVITY		HEAT CAPACITY
		(°C)	100°C	
ZrO ₂	2 715	1.9	2.3	2.6
BeO	2 530	220	20	3.1
MgAl ₂ O ₄	2 135	10.7		
CeO ₂	2 600	10.9		2.6
SiC ¹	2 700	61.6	27	2.2
SiC ²	2 700	77.5	50	2.2
Si	1 410	108		1.7
UO ₂	2 878	8.8	3.2	2.6

¹ Sintered α -phase

² Sintered β -phase

Table 2. Initial compositions (g) of weapons plutonium inert-matrix fuel in a CANFLEX bundle

	Ring 1 (1 pin)	Ring 2 (7 pins)	Ring 3 (14 pins)	Ring 4 (21 pins)
²³⁹ Pu	–	–	98.72	98.72
²⁴⁰ Pu	–	–	6.1096	6.1096
²⁴¹ Pu	–	–	0.1435	0.1435
²⁴² Pu	–	–	0.0231	0.0231
Total Pu	–	–	105.00	105.00
Si (natural)	139.64	977.42	1399.01	2098.54
C (natural)	59.84	418.90	599.58	899.38
Gd (natural)	20.00	40.0	–	–

Table 3. Initial compositions (g) of civilian Pu-actinide inert-matrix fuel in a CANFLEX bundle

	Ring 1 (1 pin)	Ring 2 (7 pins)	Ring 3 (14 pins)	Ring 4 (21 pins)
²³⁷ Np	–	–	14.98	14.98
²³⁸ Pu	–	–	2.95	2.95
²³⁹ Pu	–	–	102.5	102.5
²⁴⁰ Pu	–	–	47.8	47.8
²⁴¹ Pu	–	–	15.5	15.5
²⁴² Pu	–	–	9.25	9.25
²⁴¹ Am	–	–	15.365	15.365
²⁴³ Am	–	–	2.649	2.649
Si (natural)	139.64	977.42	1399.01	2098.54
C (natural)	59.84	418.90	599.58	899.38
Gd (natural)	10.0	10.0	–	–
Total Pu	–	–	178.0	178.0
Total MA	–	–	33.0	33.0

Table 4. Plutonium and minor actinide content (g/bundle) in PuSiC bundles

Isotope	Weapons-derived PuSiC fuel			Civilian PuSiC actinide fuel		
	Fresh	Discharge (145 MWd)	Destruction (%)	Fresh	Discharge (229 MWd)	Destruction (%)
²³⁸ Pu	–	–	–	5.90	5.5	6.8
²³⁹ Pu	197.44	4.778	97.6	205.00	6.2	97.0
²⁴⁰ Pu	12.219	31.13	–	95.60	57.5	39.9
²⁴¹ Pu	0.2870	8.531	–	31.0	15.8	49.0
²⁴² Pu	0.046	8.384	–	18.50	41.53	–
<i>Total Pu</i>	<i>210.00</i>	<i>52.823</i>	<i>74.9</i>	<i>356.00</i>	<i>126.53</i>	<i>64.50</i>
²³⁷ Np	–	0.074	–	29.96	14.76	49.3
²⁴¹ Am	–	0.649	–	30.73	3.692	88.0
²⁴³ Am	–	0.068	–	5.298	9.937	–
²⁴² Cm	–	0.089	–	–	8.576	–
²⁴⁴ Cm	–	–	–	–	3.960	–
<i>Total MA</i>	–	–	–	<i>66.0</i>	<i>40.925</i>	<i>38.0</i>

Figure 1. Reactor core model of a CANDU 6

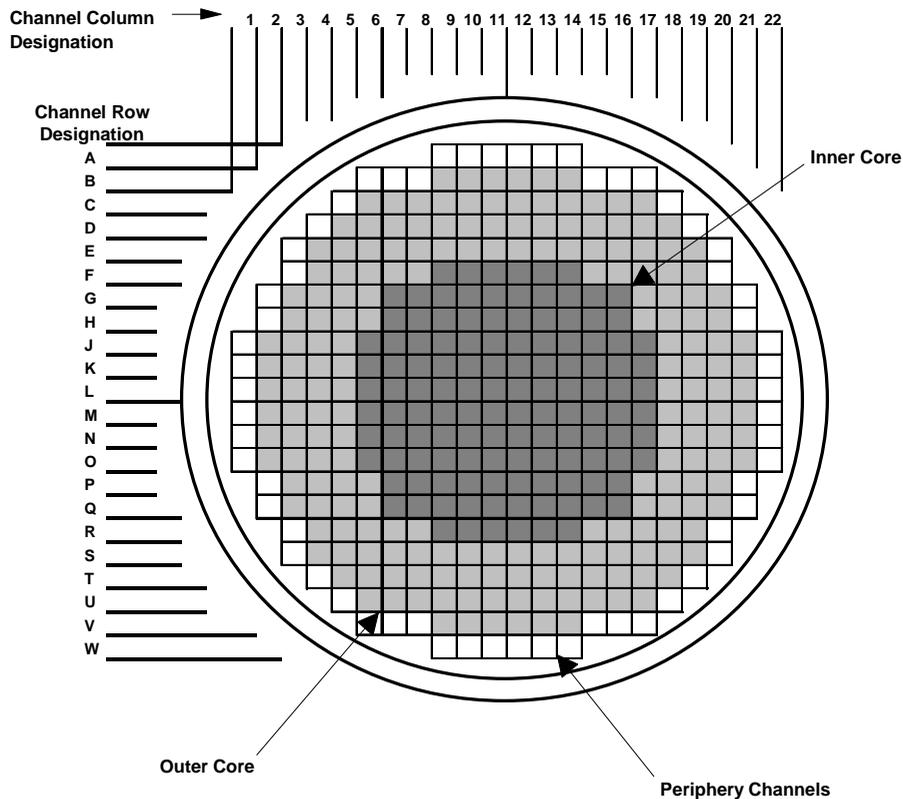


Figure 2. Configuration of fuel elements in a CANFLEX bundle

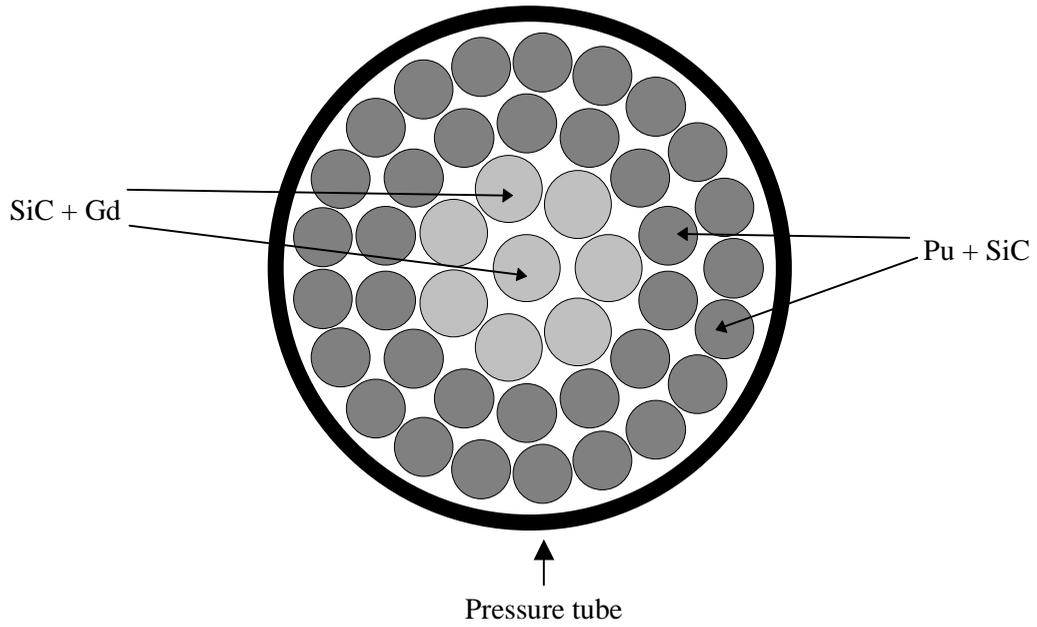


Figure 3. Lattice k-infinity vs. fuel burn-up

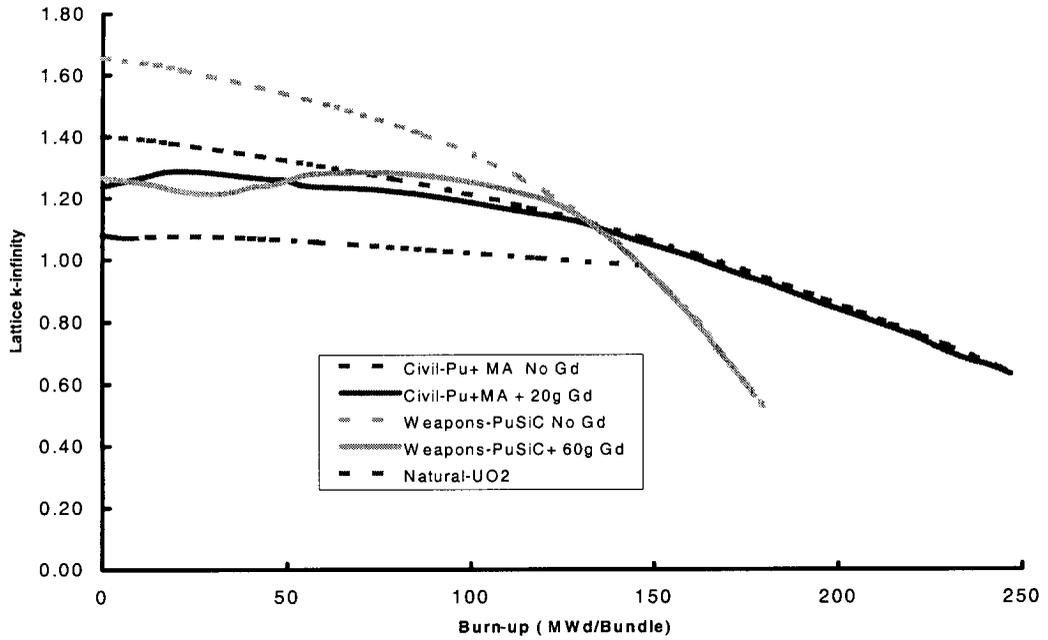


Figure 4. Radial power distribution

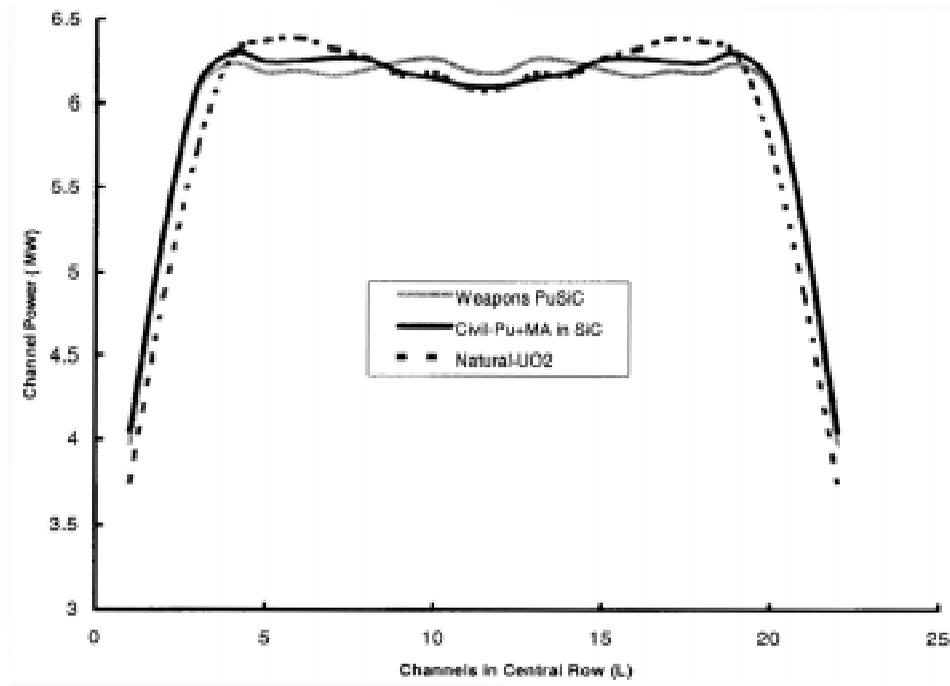
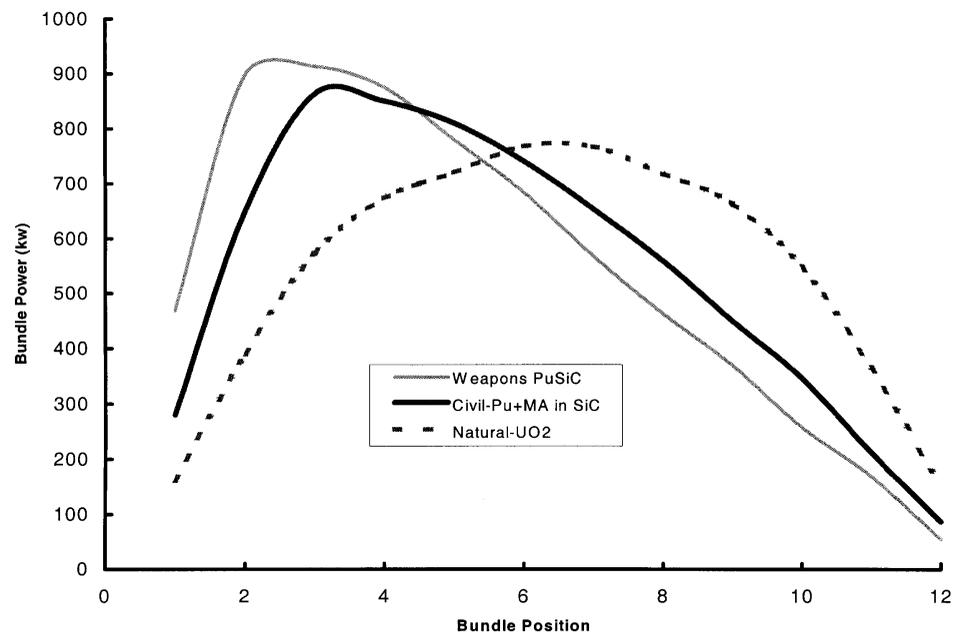


Figure 5. Axial power distribution in a central channel



NEUTRONICS OF INERT MATRIX Pu-FUEL RODS IN A UO₂ PWR ENVIRONMENT

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Abstract

Earlier studies have shown how inert matrix fuel (IMF) assemblies in a pressurised water reactor (PWR) could allow a much more effective reduction of plutonium than possible with MOX fuels. The proposed solid solution fuel consists of PuO₂ embedded in an inert matrix of ZrO₂, with Er₂O₃ as burnable poison being added to limit the initial reactivity excess. It was earlier demonstrated that, from the reactor physics viewpoint, one could establish an asymptotic fuel cycle for a current-day 1000 MW(e) PWR based on four IMF zones in the reactor core, one-fourth of the fuel assemblies being replaced each year. For practical reasons, it is necessary, however, to envisage the introduction of the IMF progressively into a PWR core. In this way, irradiation experience would be gained, allowing to test the neutronics and material technology properties of the proposed fuel, without undue influence on operational and safety related features of the core.

In the present investigation, the design of a special UO₂ assembly containing IMF rods (with reactor-grade plutonium) is considered, such that this could be irradiated during four cycles. A parameter study has been performed aimed at optimising the neutronics characteristics of such an assembly, which would effectively replace a standard uncontrolled UO₂ assembly. It is shown that the largest percentage reduction of the plutonium mass would be achieved with an assembly containing 49 IMF rods homogeneously distributed over the total of 205 fuel rod positions. The plutonium reduction, relative to the initial plutonium mass in this assembly, would amount to ~25-30%, corresponding approximately to that achievable with a 100%-MOX core loading. In the IMF rods themselves, ~75% of the initial plutonium mass would be consumed. These maximal relative plutonium reduction rates are obtained for a fuel without added burnable poison.

For the reference IMF composition with Er₂O₃, the relative plutonium consumption in the IMF rods is reduced by approximately 10%, but, at the same time, the maximal assembly power peaking factor (ppf) is improved significantly. In absolute terms, the plutonium reduction in the IMF rods (for the same maximal ppf) is considerably greater for the case with erbium due to the higher PuO₂ density.

The fact that the toxicity of the burnt IMF rods (which are intended to be disposed of without further reprocessing) would be significantly lower than that of a MOX assembly represents an additional incentive for the introduction, on a relatively short time scale, of a mixed UO₂/IMF assembly into a PWR.

Introduction

Due to economical reasons and a lack of public acceptance, the nuclear fuel cycle is not effectively closed today. Consequently, UO₂-fuelled light water reactors (LWRs) continue to dominate the nuclear energy scene, and a relatively novel situation is currently emerging with plutonium stockpiles increasing far beyond projected needs. The latter fact is of considerable concern not only from a resource viewpoint, but also from that of non-proliferation. The “self-generating” mode of plutonium recycle in LWRs, a strategy presently adopted by some utilities and involving the use of PuO₂/UO₂ (mixed oxide, or MOX) assemblies in about one-third of the core, does not offer a real solution in this context since nearly the same amount of plutonium is generated as is consumed. Designs for 100%-MOX cores have been investigated, with which reductions of approximately 30% of the plutonium mass would be achievable. The build-up of new plutonium from the ²³⁸U present in the MOX, however, remains disadvantageous from the viewpoint of inventory reduction and requires multiple recycling.

A much more efficient way to enhance the plutonium consumption in LWRs would be to eliminate plutonium production all together. This requirement leads to fuel concepts in which the uranium is replaced by an inert matrix. The inert matrix fuel (IMF) studied at PSI is based on ZrO₂ [1,2], which is a promising candidate material 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 ZrO₂ based inert matrix becomes a suitable replacement for UO₂ in MOX fuel, offering the sought after properties for hosting the plutonium, higher actinides and fission products. For reactivity control reasons, adding a burnable poison to this fuel proves to be necessary. The studies performed at PSI have identified Er₂O₃ as the most suitable candidate for this purpose.

It was earlier demonstrated that, from the reactor physics viewpoint, one could establish an asymptotic fuel cycle for a current-day 1 000 MW(e) PWR based on four IMF zones in the reactor core, one-fourth of the fuel assemblies being replaced each year [3]. In more recent studies [4,5], the influence of the introduction of a limited number of IMF assemblies on the performance of a standard UO₂-fuelled PWR (main characteristics given in Table 1) has been investigated. In these three-dimensional full core analyses, up to 12 uranium-free assemblies containing only IMF fuel rods were considered. The results obtained indicate that loading 4 to 12 IMF assemblies permits to envisage fuel management schemes compatible with current operational requirements (e.g. with respect to cycle length and boron concentration). Furthermore, with the proposed IMF assembly design, it is possible to obtain satisfactory power distributions, even for cases in which the IMF assemblies are loaded into the peak power positions.

The plutonium consumption capability of such mixed cores may be evaluated on the basis of the earlier investigated 100%-IMF core loadings. Caution is certainly due when drawing conclusions on the potential plutonium consumption rates of cores which go beyond the configurations with 4-12 IMF assemblies, because of the obvious need to perform additional fuel management studies. Nevertheless, based on the 100%-IMF loaded core results, it is possible to reliably assess at least the *relative* plutonium consumption capability of partial loadings with IMF as compared to those with MOX. Thus, one finds that the PWR reference core considered would consume the same amount of plutonium when loaded with 4 or 12 IMF assemblies as it would with 10 or 30 MOX assemblies, respectively. In other words, in terms of the “self-generating” mode mentioned earlier, it can be concluded that a 1/8 loading of IMF assemblies would permit the PWR to run in such a mode (i.e. with approximately 2.5 times less Pu-containing assemblies than in the corresponding MOX case).

As a first step towards PWR core loadings containing complete IMF assemblies, one could envisage the introduction of a limited number of IMF rods into a PWR. It goes without saying that these rods should not alter the operational and safety related characteristics of the hosting core. Furthermore, a high plutonium consumption rate in these rods should be aimed for, thus providing an immediate incentive to the plant's operators. In the present study, a parametrical approach has been adopted to assess the neutronics characteristics of a special UO₂ assembly containing a limited number of IMF rods. It is suggested that such an assembly could be irradiated in a commercial PWR for the usual full residence time by using it in place of a standard uncontrolled UO₂ assembly. As such, it would have the same geometrical specifications (see Table 1).

Table 1. Main characteristics of the PWR reference core

Parameter	Unit	Value
Thermal power	MW	3 150
Specific power density	MW/cm ³	108
Coolant temperature		
Inlet	°C	291.5
Outlet	°C	324.8
Coolant flow	kg/s	15 665
Pressure	bar	154
²³⁵ U enrichment of the UO ₂ fuel	wt%	4.3
Number of fuel rods per assembly	–	(15 × 15) - 20
Number of fuel assemblies in the core	–	177
Number of controlled fuel assemblies	–	48
Active fuel length	cm	355
Fuel assembly pitch	cm	21.56
Cell pitch	cm	1.43
Outer fuel pellet diameter	cm	0.913
Outer clad diameter	cm	1.075
Clad thickness	cm	0.0725

Description of the present study

The parameter study presented here addresses the main design features of the special assembly, i.e. the number and positions of the IMF rods, the ²³⁵U enrichment of the standard UO₂ fuel rods, and the densities of PuO₂ and the burnable poison in the IMF rods. The design criteria applied can be summarised as follows:

- The proposed assembly (consisting of a symmetric fuel arrangement with respect to the main lateral and diagonal axes) should have an average “reactivity” corresponding to that of the standard UO₂ assembly.
- The ²³⁵U enrichment should not significantly exceed 4.3 wt%, corresponding to the fuel presently in use (see Table 1).
- The power peaking factor (ppf, defined as the ratio between the maximum fuel rod power and the average assembly value) should not exceed, at any time during the irradiation period, the limit of 1.65.

- The plutonium consumption in the IMF rods should be at least as large as its build-up in the UO₂ fuel rods of the proposed assembly.
- The time-dependent toxicity of the burnt IMF rods (to be sent directly for final disposal) should be significantly lower than that of a representative present-day MOX fuel assembly, assumed to be employed in the same core and disposed of in a similar manner.

For several of the assembly designs obtained in the framework of the parameter study, average values have been computed for: (a) the reduction of the plutonium mass; (b) the relative plutonium consumption, with respect to both the whole fuel assembly and the IMF rods themselves; and (c) the thermal power generated in the IMF rods. Among the cases considered, the number and positions of the IMF rods leading to the maximum reduction of the plutonium mass (in relative terms) have been determined, having suitably fixed the other parameters. In addition, two representative sets of specifications are described, with which as much plutonium would be consumed in the assembly as is produced (see section entitled *Results*).

Calculational methodology

All the calculations have been performed at the single assembly level, using the two-dimensional code BOXER which forms part of the ELCOS system [6]. In the depletion calculations, effective full power days were chosen (instead of the specific burn-up) to quantify the irradiation time, the heavy metal content being different in the IMF and UO₂ fuel. Each of the four cycles was assumed to be 320 effective full power days in length, without any extension with power reduction. A depletion independent, time-averaged boron concentration of 500 ppm was used for the water coolant. The density of ZrO₂ in the IMF was 5.0 g/cm³, with Y₂O₃ being neglected from the neutronics viewpoint [4].

The standard assembly with UO₂ fuel rods, the ²³⁵U enrichment being 4.3 wt%, was analysed first to determine the k_{eff} -values¹ at the end of each cycle. Thus, an average k_{eff} value (of 1.045 without boron) was obtained. This reference value was taken as the target for each mixed UO₂/IMF assembly. In the calculations performed for a given number and arrangement of the IMF rods (at first without burnable poison), the appropriate PuO₂ density in the IMF was determined as a function of the ²³⁵U enrichment of the UO₂ fuel rods. Burnable poison was added to the IMF if the ppf exceeded, at any point during the irradiation period, the limiting value of 1.65.

The two-dimensional calculations were all based upon a well established broad-group structure for LWR applications (eight groups, of which three are thermal, i.e. below 1.3 eV). The cross-section library was based upon the Joint European File, Revision 1 (JEF-1.1) [7], except for the erbium isotopes data, which were taken from the Russian file BROND-2 [8], and ¹⁵⁵Gd, which was taken from JENDL-2 [9]. To simulate, as closely as possible, the spectral conditions seen by the IMF-containing assembly when loaded into a standard PWR, each case had to be considered with the assembly surrounded by a sufficiently thick “layer” of standard UO₂ fuel rods. It was found that even a single row of UO₂ fuel rods is sufficient for this purpose. As regards the cell calculations, a unit UO₂ cell (4.3 wt%) was first calculated in the fundamental mode spectrum for various burn-up states. The outgoing partial current from this cell served as boundary condition for the calculations for each cell type in the IMF containing assembly.

¹ The geometrical buckling assumed was $3.1 \cdot 10^{-4} \text{ cm}^{-2}$.

Results

The largest percentage reduction of plutonium is achieved by an assembly with 49 IMF rods distributed uniformly over the total of 205 fuel rod positions, i.e. with about one-fourth of the fuel rods consisting of IMF (see Figure 1). The results from the parametric calculations are summarised in Table 2.

Figure 1. Horizontal cross-sectional view of the south-east quarter of the optimised mixed UO₂/IMF assembly (49 IMF rods, maximal Pu mass reduction)

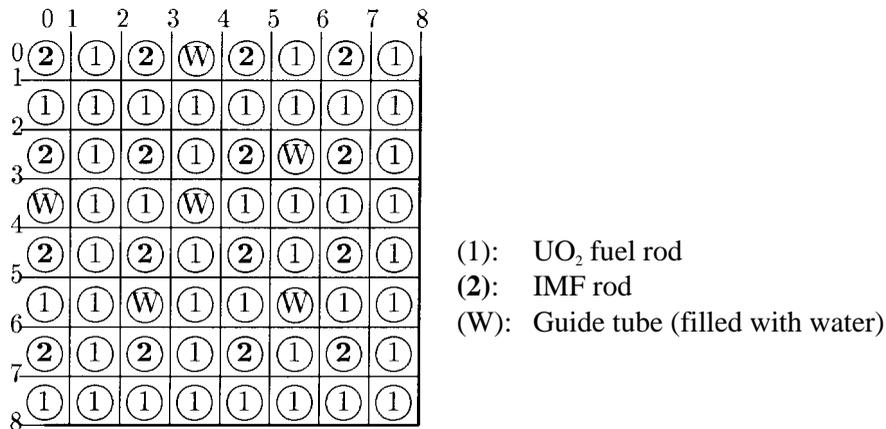


Table 2. Performance characteristics of assemblies with 49 IMF rods, in which a maximal %-reduction of plutonium is achieved

²³⁵ U enr.	PuO ₂ dens. in IMF	Er ₂ O ₃ dens. in IMF	Pu mass at EOL	Pu mass red.	Max. ppf	Pu cons. in the ass.	Pu cons. in the IMF rods	Power prod.
(wt%)	(g/cm ³)	(g/cm ³)	(kg)	(kg)		(%)	(%)	(%)
4.4	1.020	0.3432	7.3	2.8	1.54	28	69	35
4.3	0.738	–	5.5	1.8	1.50	25	78	23
4.2	0.788	–	5.7	2.1	1.54	27	77	25
4.1	0.839	–	5.9	2.4	1.58	29	76	26
4.0	0.889	–	6.2	2.6	1.62	30	75	27
3.9	0.939	–	6.4	2.9	1.65	31	74	28

From the tabulated values, it is seen that:

- For ²³⁵U enrichments between 3.9 and 4.3 wt%, the maximal ppf does not exceed 1.65 and burnable poisons have therefore not been considered. The following observations are made:
 - The initial plutonium mass ranges from ~7.3 to 9.3 kg. Correspondingly, the PuO₂ density in the IMF varies between ~0.74 and 0.94 g/cm³. Since similar densities are typical for conventional MOX fuels and the IMF rods are uniformly distributed among the UO₂ rods, it is not too surprising that the plutonium mass reduction in the assembly relative to the initial plutonium mass (~25-30%) is of the same order as that achievable with a 100%-MOX core loading.

- As much as ~5.7-6.9 kg plutonium are consumed in the IMF rods, whereas ~4 kg are built up in the UO₂ fuel rods. The plutonium consumption achieved in the IMF rods relative to the initial plutonium mass (~75%) is larger than that achievable with a 100%-IMF loading (~60%) [4]. In all cases, there remains less than 1% of the initial ²³⁹Pu mass in the IMF rods. The UO₂ fuel burns more slowly than the IMF, particularly in the later cycles. Neutrons produced by the UO₂ thus enhance the depletion of the IMF rods, as compared to the case of a 100%-IMF assembly.
- The IMF rods produce, globally, as much as ~4.1-5.1 MW(thermal), corresponding approximately to the power generated by the same number of UO₂ fuel rods (~4.3 MW(thermal)).
- The following trends are clearly observed:
The lower the ²³⁵U enrichment, the larger are: (a) the initial plutonium mass, and therefore also the density of PuO₂ in the IMF; (b) the plutonium mass reduction in the assembly; (c) the absolute plutonium mass consumption achieved in the IMF rods; (d) the thermal power produced by the IMF rods; but (e) also the maximal ppf during the irradiation period. However, in relative terms, the plutonium consumption rate in the IMF rods decreases with increasing plutonium density. All these variations are almost linear with respect to a change in the ²³⁵U enrichment.
- The IMF with erbium (as optimised in the previous studies [3,4,5], aiming at the introduction of 100%-IMF assemblies) requires an enrichment increase of the UO₂ fuel rods to 4.43 wt%. For the same maximal ppf, the plutonium consumption rate in the IMF rods, in relative terms, is ~10% lower in this case as compared to that without burnable poison. (The absolute plutonium mass reduction is, however, considerably higher due to the greater PuO₂ density). With respect to the relative plutonium consumption rate in the assembly as a whole, there is almost no difference between the two cases.

Two assembly designs yielding a “self-generating” mode have also been currently identified. Table 3 gives their specifications and main performance data.

Table 3. Description and performance characteristics of two assemblies, in which as much plutonium is consumed as is produced

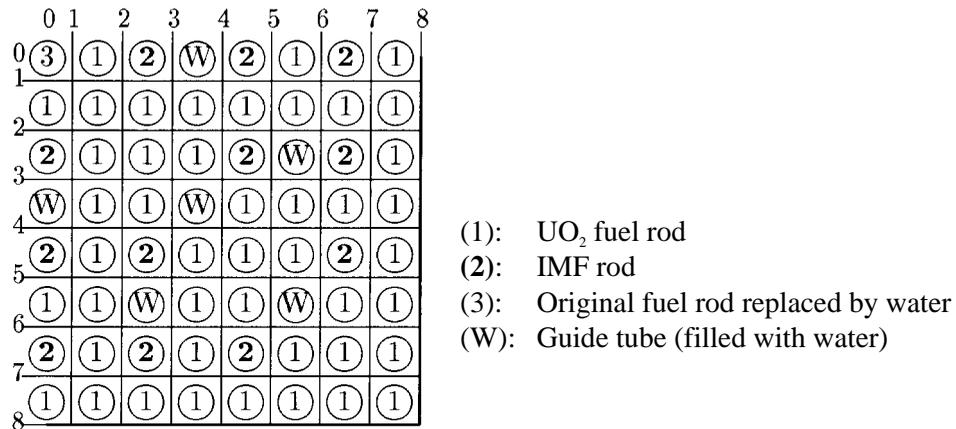
Assembly feature	²³⁵ U enrich. (wt%)	Number of IMF rods (%)	PuO ₂ density in the IMF (g/cm ³)	Pu mass EOL/BOL (kg)	Max. ppf	Pu cons. in the IMF rods	
						(%)	(%)
(a)	4.3	17.6	0.738	5.4	1.55	77.7	17.3
(b)	4.0	13.7	1.2	6.9	1.65	70.0	19.5

(a) Central fuel rod replaced by water

(b) IMF poisoned with 0.4 g/cm³ Gd₂O₃

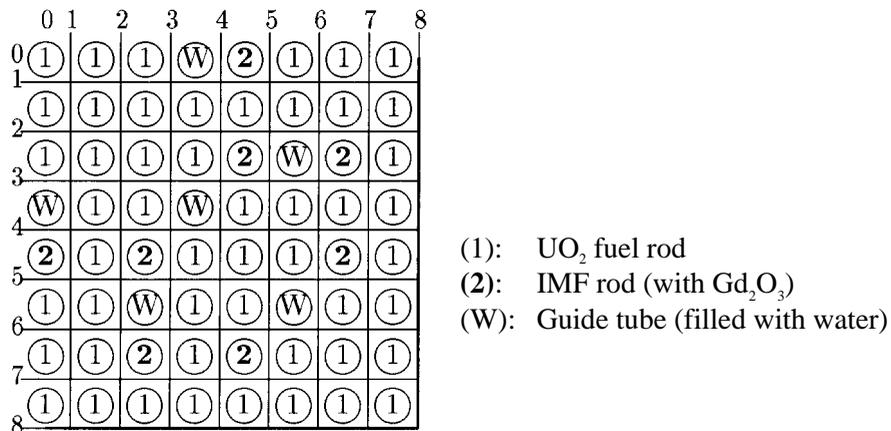
The first assembly was obtained by replacing by water the central fuel rod (of the previous design with 49 IMF rods), and by substituting UO₂ fuel rods for the IMF rods positioned along the main diagonal (see Figure 2). Thus, this assembly features 36 IMF rods, corresponding to about one-sixth of the total number of fuel rod positions available (204, in this case). The plutonium mass is 5.4 kg, and the plutonium consumption achieved in the IMF rods relative to the initial plutonium mass amounts once again to ~75%. The IMF rods, in this case, produce ~3.1 MW(thermal).

Figure 2. Horizontal cross-sectional view of the south-east quarter of a “self-generating” mixed UO₂/IMF assembly (36 IMF rods, Pu mass built-up = Pu mass consumed)



The specifications for the second assembly (see Figure 3) were determined so as to minimise the number of IMF rods. This assembly has 28 IMF rods, corresponding to less than one-seventh of the total number of fuel rods. In this case, poisoning is necessary. The calculations presented here considered Gd₂O₃ as a possible burnable poison. The Gd₂O₃ density employed is 0.4 g/cm³. The plutonium mass is 6.9 kg, and the plutonium consumption achieved in the IMF rods relative to the initial plutonium mass amounts to 70%. The IMF rods here produce ~2.4 MW(thermal).

Figure 3. Horizontal cross-sectional view of the south-east quarter of a “self-generating” mixed UO₂/IMF assembly (28 IMF rods, Pu mass built-up = Pu mass consumed)



Conclusions

The present study has investigated the performance characteristics of mixed UO₂/IMF assembly designs, which could represent a first step towards PWR cores containing 100%-IMF assemblies. The mixed UO₂/IMF assembly would be irradiated during four cycles and is intended to replace a standard uncontrolled UO₂ assembly.

It has been shown that the largest relative reduction of the plutonium mass is achieved by an assembly with 49 IMF rods uniformly distributed over the total of 205 fuel rod positions (see Figure 1). With ^{235}U enrichments of 3.9 to 4.3 wt%, the PuO_2 density in the IMF being smaller than $\sim 0.94 \text{ g/cm}^3$, the use of burnable poisons would not be required and one finds that:

- The plutonium mass reduction for the full assembly (relative to the initial plutonium mass) amounts to $\sim 25\text{-}30\%$, corresponding approximately to that achievable with a 100%-MOX loading.
- The plutonium mass reduction in the IMF rods, relative to the initial plutonium mass, amounts to $\sim 75\%$ and is larger than that achievable with a 100%-IMF loading ($\sim 60\%$). This is because the UO_2 fuel burns more slowly than the IMF, particularly in the later cycles. Neutrons produced by the UO_2 fuel rods enhance the depletion of the IMF, as compared to the 100%-IMF assembly case.
- The IMF rods generate approximately as much power as the same number of UO_2 fuel rods.

The fact that the toxicity of the burnt IMF rods would be significantly lower than that of a MOX assembly (assumed to be employed in the same core and disposed of later in a similar fashion) represents an additional incentive to envisage, on a relatively short time scale, the introduction of a mixed UO_2/IMF into a PWR.

The study has furthermore shown that a mixed UO_2/IMF assembly design based on the reference IMF with erbium (optimised for 100%-IMF assembly concepts) would yield somewhat lower relative, but considerably higher absolute, plutonium consumption rates in the IMF rods (for the same maximal ppf) than the above discussed designs without burnable poison. While the final decision as to which assembly should be considered for irradiation in a power reactor will have to take into account primarily the plant operator's requirements, it is clear that the most relevant physics and fuel performance data would be obtained from the irradiation of a test assembly based on the reference IMF design with erbium.

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**SOME NEUTRONIC PROPERTIES OF AN INERT
MATRIX FOR THE DEFINITION OF A 100% IMF CORE**

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Abstract

After having shown that in the current French and world-wide context, plutonium fuels in an inert matrix can provide a solution to the management of Pu stocks, an analysis of certain metals likely to be candidates for these matrices is presented. It is demonstrated that, although 100% core loading is possible for UO₂ and MOX CERMET composite fuels, albeit with some constraints, the same does not hold true for Pu CERMETS. A proposal can be made concerning the APA assembly, which allows efficient consumption of Pu, while retaining acceptable kinetics coefficients.

Introduction

If we consider the international context which tends to favour the most efficient use of natural resources, while minimising the volumes, quantities and radiotoxicity of resulting wastes, together with the desire to increase resistance to proliferation so as to reinforce the notion of public acceptance, a basis for serious thought is established. It should then be noted that the very early implementation by France of a reprocessing policy and the subsequent delay in the development of fast neutron reactors have resulted in an increase in Pu stocks. Finally, the policy of the utility, EdF, which only reprocesses the amount of irradiated fuel needed for the fabrication of the MOX fuels authorised to constitute up to 30% of core loads in 20 out of the 58 French reactors, further contributes to this increase in Pu stocks.

Certain solutions [1,2,3] can help to increase the fraction of loaded MOX fuel but the conversion of ^{238}U to ^{239}Pu will nevertheless continue to produce Pu. It is clear that, in this type of operational process, the Pu stream from French NPPs will always be positive. This is why, with a view to balancing the Pu stream in France, fuels in inert matrices [4,5,6] evidence particularly interesting advantages.

Neutronic studies

The ceramic matrices selected for pressurised water reactors, spinel (MgAl_2O_4) and CeO_2 , are transparent to neutrons; only metals (and therefore CERMETS) interest us here.

Figure 1 shows that three categories can be distinguished in the metals considered:

- Absorbent metals (steel, Mo, etc.).
- Intermediate metals (Inconel, etc.).
- Transparent metals (Zry, Al, etc.).

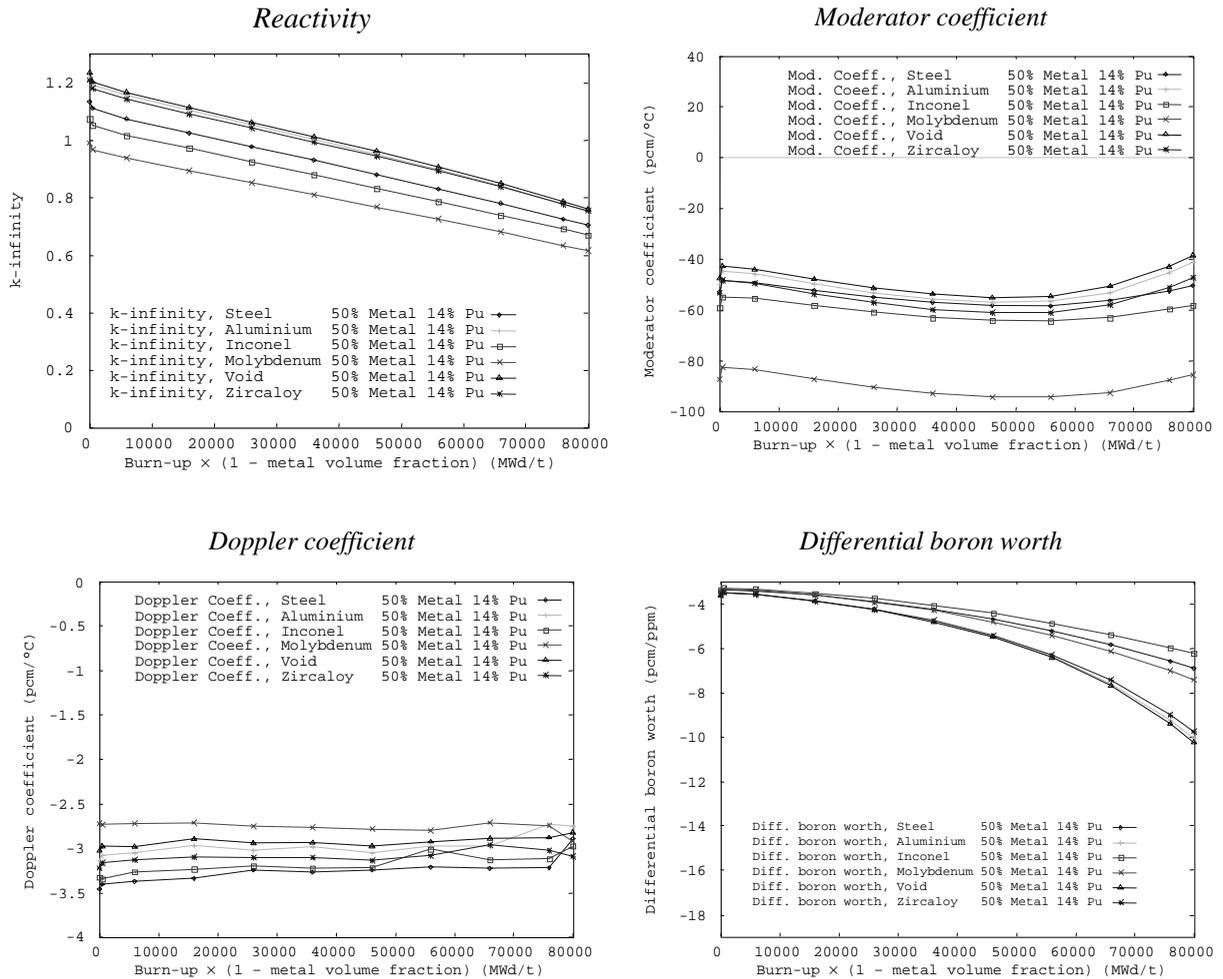
Their behaviour is clearly illustrated in Table 1, which presents a comparison of the contents required to ensure the same cycle duration (18 months) for different CERMETS (50% volume fraction), UO_2 and a Pu/Th CERCER.

**Table 1. Metal volume fraction 0.5, cycle length = 18 months
Fissile material content (%)**

	UO_2	CERMET UO_2	CERMET MOX	CERMET PuO_2	CERCER Pu/Th
^{235}U	3.7%				
Inconel		14.1%	32.5%	14.3%	
Mo		18.7%	50%	25.7%	
Zry		7.5%	18%	6.9%	
Th					14%

In particular, the very great absorption of Mo should be noted: it requires nearly four times more PuO_2 than Zry to sustain the same cycle duration. It should also be noted that ThO_2 CERCER behaves like an Inconel CERMET with respect to the Pu content needed to sustain the same cycle duration. Figure 1 shows, as regards assembly calculation, a certain number of tendencies, in particular:

Figure 1. Some characteristics of different metals as inert matrix



- The Doppler coefficients evidence a slight dependence in relation to the matrix (except in the case of a resonant metal, of course, W, for example, but rather more in relation to the Pu vector).
- The efficiency of soluble boron differs highly between elements transparent to neutrons (Zry, Al, void) and the others (absorbent).
- Reactivity depends considerably on the matrix.
- The moderator coefficient is very sensitive to the matrix.

With the aim of immobilising a large quantity of Pu in the core (resistance to proliferation) one could imagine using Mo to increase the Pu fraction; however, the very negative moderator coefficient would make cold hazards uncontrollable, and, in addition, in this case the efficiency of the soluble boron is low. Finally, it can be observed in Tables 2, 3 and 4, that the positive draining coefficient disqualifies this matrix (as well as Inconel), for both MOX and PuO₂.

Table 2. Boron worth (1 200 ppm)

	BOL pcm/ppm
UO₂	-5.73
UO₂ + Inconel	-3.37
UO₂ + Molybdenum	-2.91
UO₂ + Zircaloy	-5.73
MOX + Inconel	-1.29
MOX + Molybdenum	-0.93
MOX + Zircaloy	-2.07
PuO₂ + Inconel	-1.41
PuO₂ + Molybdenum	-1.17
PuO₂ + Zircaloy	-2.41
PuO₂ + ThO₂	-2.00

Table 3. Doppler coefficient (pcm/°C)

	BOL	EOL
UO₂	-2.00	-3.39
UO₂ + Inconel	-1.99	-3.29
UO₂ + Molybdenum	-1.68	-2.59
UO₂ + Zircaloy	-1.72	-3.55
MOX + Inconel	-2.79	-2.98
MOX + Molybdenum	-1.93	-1.99
MOX + Zircaloy	-2.97	-3.42
PuO₂ + Inconel	-1.04	-1.10
PuO₂ + Molybdenum	-0.71	-0.57
PuO₂ + Zircaloy	-1.09	-1.80
PuO₂ + ThO₂	-3.30	-3.47

Table 4. Draining coefficient (1 200 ppm)

	BOL	EOL
UO₂	-61 429	-94 613
UO₂ + Inconel	-33 029	-63 650
UO₂ + Molybdenum	-45 168	-75 969
UO₂ + Zircaloy	-53 536	-181 167
MOX + Inconel	+4 953	+3 281
MOX + Molybdenum	+11 618	+13 434
MOX + Zircaloy	-13 471	-28 877
PuO₂ + Inconel	+4 183	-3 841
PuO₂ + Molybdenum	+9 481	-8 922
PuO₂ + Zircaloy	-9 873	-142 821
PuO₂ + ThO₂	-16 690	-41 704

Cycle capacity of MOX and UO₂ CERMETS

Figure 2 and Table 5 show that in the case of UOX and MOX CERMETS, although there is no solution for a half-core management for MOX, obtaining a discharge burn-up of 120 GWd/t under other management conditions requires a 10% enrichment in ²³⁵U and a Pu 2016 content of about 20% [7,8] (Table 6).

Figure 2. Average burn-up at unloading

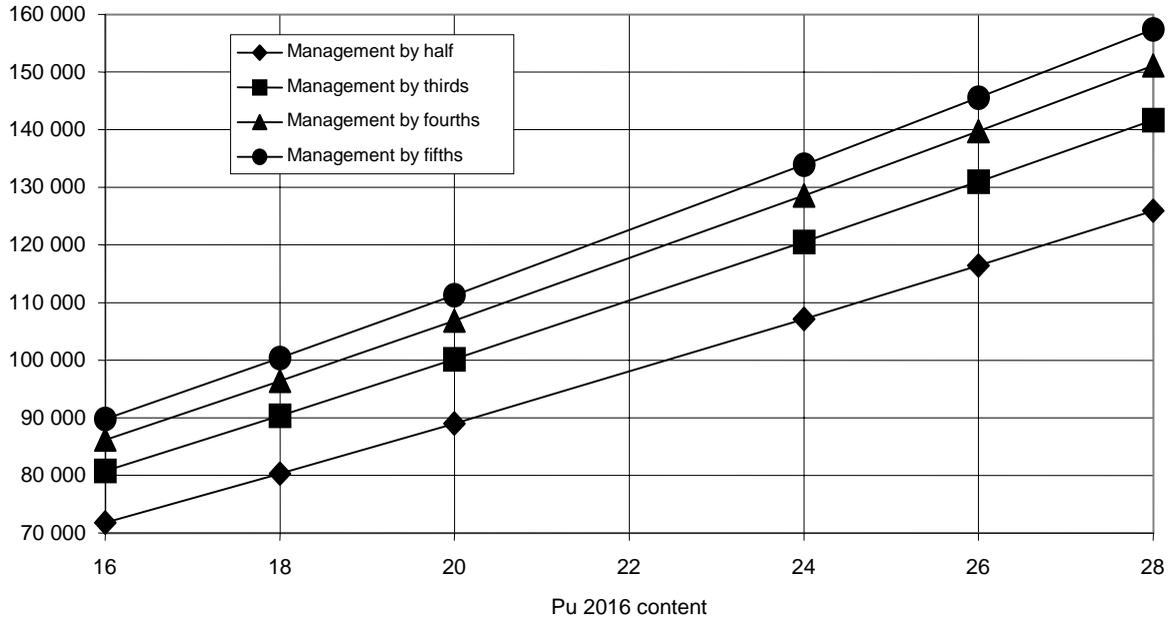


Table 5. Core management with two composite fuels

	CERMET/UOX ²³⁵ U%	CERMET/MOX Pu content %
1/2 core	11.2	???
1/3 core	10.4	23
1/4 core	9.9	20.8
1/5 core	9.6	20

Table 6. Pu 2016 isotopic composition

	%
²³⁸ Pu	2.72
²³⁹ Pu	56.39
²⁴⁰ Pu	26.08
²⁴¹ Pu	7.45
²⁴² Pu	7.35

It can be seen (Figure 3) that for a 50-50 UO₂ CERMET, soluble boron concentrations at beginning of life must be lower than 1 000 ppm to assure a negative moderator coefficient. As control requirements are about 2 000 ppm, it is obvious that considerable recourse to burnable poisons would be necessary. This is why studies [9,10] have been performed with the aim of poisoning the cladding and the matrix with a few per cent erbium. The results are encouraging, from both the neutronics and technological aspects.

Figure 3. Moderator coefficient for various boron contents

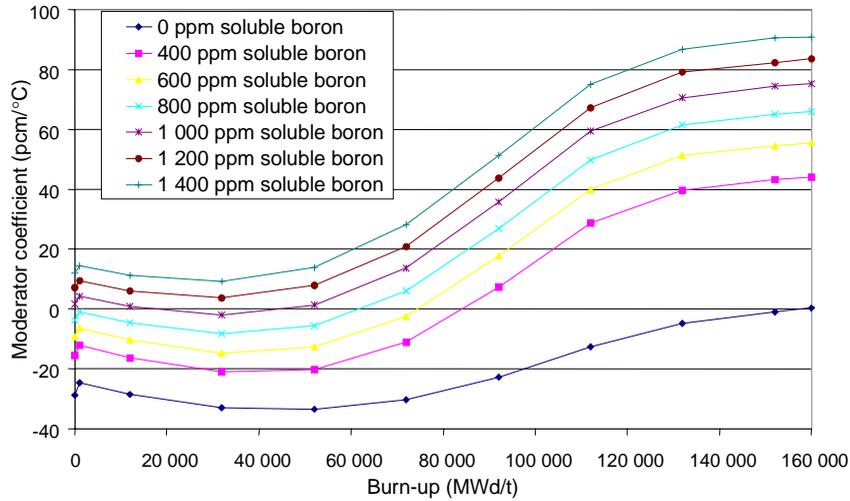


Figure 2 shows that for a MOX-Zry 50-50 CERMET a discharge burn-up of 120 GWd/t requires assuring one-third core (24% Pu), one-quarter core (22.5% Pu) or one-fifth core management (22% Pu). Figure 4 clearly shows that for these types of management the draining coefficient remains negative and Figure 5 that concentrations in soluble boron in these cases are about 2 800 ppm for the one-third core, 2 200 ppm for the one-fourth core and 1 500 ppm for the one-fifth core. The last two types of management are possible with, however, considerable recourse to burnable poisons for one-quarter core management.

Figure 4. Draining coefficient/Pu contents

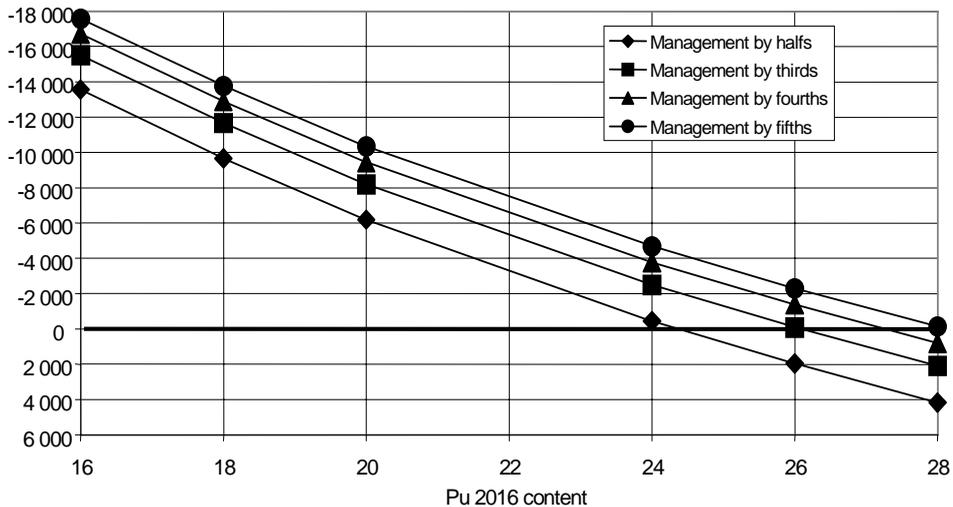
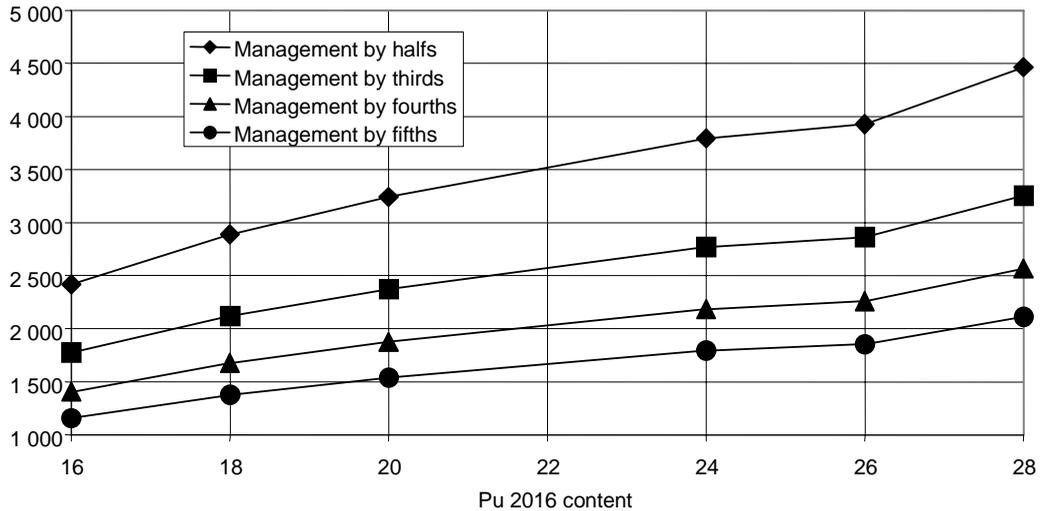


Figure 5. Boron content BOL



Conclusion

A MOX, Zry CERMET seems possible as fuel for a 900 MWe PWR.

For a burn-up objective of 120 GWd/t, only management by one-third core, one-quarter core and one-fifth core is possible. For these contents, the draining coefficient does not pose a problem, as it is always negative. However, the quantities of soluble boron that are needed limit management to one-quarter or one-fifth cores, while it should be noted that one-quarter core management requires the intensive use of burnable poisons [9,10].

Finally, the Doppler coefficient is better ($-3.2 \text{ pcm}/^\circ\text{C}$) than that of UO_2 ($-2.6 \text{ pcm}/^\circ\text{C}$) which would somewhat compensate for a low effective beta in the event of reactivity insertion.

The case of a Pu CERMET fuel is different; it is not evident that a core using this type of fuel can be defined. As a result, research has been oriented towards the heterogeneous advanced Pu assembly (APA) [11,12,13], to ensure core control under acceptable conditions.

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INERT MATRIX AND THORIA FUELS FOR PLUTONIUM BURNING IN LWRs

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Abstract

Proliferation risks, radiotoxicity, environmental safety and energy conservation are reasons which suggest the burning of excess Pu in existing available LWRs world-wide.

Inert matrix fuel is a non-fertile oxide fuel consisting of PuO₂, either weapons-grade or reactor-grade, diluted in inert oxides such as stabilised ZrO₂. Its primary advantage is that it does not produce new plutonium during irradiation, because it does not contain uranium (U-free fuel). An addition of thoria in the matrix (thoria-doped fuel) may be required for coping with reactivity feedback needs.

The full thoria-plutonia fuel – though still a U-free variant – can no longer be defined as non-fertile because of the generation of ²³³U. The advantage of such a fuel option resides in the already existing remarkable technological background and a potential acceleration in the process of disposing of the Pu stocks.

All U-free fuels are envisaged to be operated under a once-through cycle scheme (the spent fuel is supposed to be sent directly to the final disposal in deep geological formations without requiring any further reprocessing treatment).

Calculations show that IMF allows an almost complete burning of fissile plutonium, thus eliminating any proliferation concern. However, whole core loading is not feasible, thus the yearly amount of burnt plutonium is limited. On the other hand, thoria-plutonia fuels have a lower capability for plutonium burning, alleviated by the possibility of accepting a whole core loading. Some proliferation concerns still exist.

An R&D activity for fuel validation is in progress in Italy, in view of irradiation testing in the Halden reactor and/or a Russian PWR. Fabrication tests on simulated fuels via the GSP process are in progress. Corrosion tests confirmed the high chemical stability of these materials.

A new method for thermal parameters measurement has been set up and thoroughly tested.

Introduction

Four generic fuel types could be used for burning weapons-grade (WG) or reactor-grade (RG) plutonium in LWRs:

- a) Uranium-based mixed oxides (MOX) $(U,Pu)O_2$.
- b) Inert matrix fuels (IMF), in which PuO_2 is dispersed in a neutron transparent ceramic inert carrier.
- c) Thoria-doped inert matrix fuels (TD), in which a given amount of thorium oxide is added to the inert matrix in order to improve the dynamic coefficients.
- d) Thorium-based mixed oxides $(Th,Pu)O_2$.

Inert matrix fuel (IMF) is a completely novel fuel consisting of PuO_2 , either weapons-grade or reactor-grade, diluted in a compound of inert oxides based for instance on stabilised ZrO_2 . This new fuel is expected to respond to the following basic requirements: good chemical compatibility, acceptable thermal conductivity, good nuclear properties, good stability under irradiation, high dissolution resistance and long term chemical stability. As a result of these positive qualities, after discharge from reactor and adequate cooling time, the spent fuel is supposed to be sent, as a HLW, directly to the final disposal in deep geological formations, without requiring any further reprocessing treatment (once-through solution). Moreover the expected very limited solubility under the current fuel reprocessing techniques, coupled with the quality-poor residual Pu in the spent fuel, will make inert matrix fuel an anti-proliferation product.

The substitution of some amount of inert oxide with thoria yields a thoria-doped IMF. Thorium doping enables to obtain much more favourable values of the dynamic coefficients and of the power generation, at the expense of a slight reduction in the plutonium burning capability. The main weakness of the inert matrix fuels, with and without thoria, is that no operational experience is currently available, and any developed would have to be thoroughly tested before commercial application. Thorium-based mixed oxides, $(Th,Pu)O_2$, is an interesting fuel option because of its potentiality of good Pu consumption, even then not as high as for IMF. Moreover, there exists a wide experience on $(Th,U)O_2$ fuels, that indicates a very good behaviour under irradiation, better even than UO_2 , and a limited experience on thoria-plutonia fuels confirms this behaviour. The resulting exhausted fuel exhibits a high stability (this feature is viewed as a drawback from the recycling perspective), thus a direct disposal is also likely to be appropriate for this kind of fuel. Disadvantages for Th-fuels would be proliferation concerns, related to the production of ^{233}U , amenable however to be coped with.

This paper presents a summary of neutron physics calculations on IMF and thoria-based fuels, and a survey of the R&D programme which, starting with a simulation of a fuel sample fabrication and characterisation, are presently underway in Italy as a joint activity between ENEA and Politecnico di Milano.

The inert matrix and thoria fuel concepts

The inert matrix and thoria fuel concepts consist basically of a dispersion of Pu oxide in inert ceramic oxides and thoria, respectively, where the Pu content is set in order to give approximately the

same reactivity performances as obtained in standard ^{235}U enriched fuel for LWRs. The constituent materials are then:

- Pu oxide, under powder or sol-gel microsphere form (metal Pu-WG assumed to be converted to oxide).
- Matrix oxides consisting of: stabilised $\text{ZrO}_2\text{-CaO}$, which is the present preferred solution; MgAl_2O_4 (spinel); mixture of Al_2O_3 , $\text{ZrO}_2\text{-Y}_2\text{O}_3$, MgO ; ThO_2 .
- Burnable poisons, whose presence is foreseen for controlling peaking factors and burn-up reactivity swings, such as a pellet coating of ZrB_2 in the form of an integral fuel burnable absorber (IFBA), and such as Gd_2O_3 or Er_2O_3 inside the pellet.

The possible fabrication processes include the gel supported precipitation (GSP) sol-gel process or the standard dry-blended powders technology process.

A priority is being given to microspheres produced via the GSP “wet route” process, which is less contaminant during the fabrication phase (no fines). Pellets are pressed and sintered following the current LWR fuel technology. Fuel pin and assembly will respect the standard LWR geometry.

Neutronic analysis

The calculations were performed assuming that a reactor having AP-600 design characteristics is fully loaded with four different fuel types: $(\text{U,Pu})\text{O}_2$, inert matrix fuel, thorium-doped inert matrix fuel and $(\text{Th,Pu})\text{O}_2$. Their compositions are shown in Table 1. For each fuel type, two different qualities of plutonium, i.e. weapons-grade (WG) and reactor-grade (RG), were considered. The corresponding isotopic vectors (^{239}Pu , ^{240}Pu , ^{241}Pu , ^{242}Pu) were 94%, 6%, 0%, 0% and 58%, 24%, 13%, 5%.

Table 1. Composition of the analysed fuels (mol.%) for obtaining 1 185 EFPDs

Fuel	Description	PuO_2	UO_2	ZrO_2	ThO_2
<i>PU-RG</i>	$(\text{U,Pu})\text{O}_2$ with Pu-RG	6.60	93.40	–	–
<i>PU-WG</i>	$(\text{U,Pu})\text{O}_2$ with Pu-WG	3.65	96.35	–	–
<i>IM-RG</i>	Inert matrix fuel with Pu-RG	5.09	–	94.91	–
<i>IM-WG</i>	Inert matrix fuel with Pu-WG	4.14	–	95.86	–
<i>PT-RG</i>	$(\text{Th,Pu})\text{O}_2$ with Pu-RG	8.12	–	–	91.88
<i>PT-WG</i>	$(\text{Th,Pu})\text{O}_2$ with Pu-WG	4.92	–	–	95.08
<i>TD-RG</i>	Thoria-doped inert matrix fuel with Pu-RG	5.94	–	64.06	30.00
<i>TD-WG</i>	Thoria-doped inert matrix fuel with Pu-WG	4.32	–	65.68	30.00

For all the fuels containing ZrO_2 , the pellets were supposed to have a central hole with a volume equal to 25% of the total, because of the low thermal conductivity of this matrix. The plutonium inventory at BOL was determined in order to achieve a discharge burn-up of 1 185 effective full power days (EFPDs) for all the fuels, considering a zero-dimension three-batch fuel management scheme. The depletion analysis was performed via the WIMS D5 programme.

In Figures 1 and 2 the reactivity swings are presented for Pu-RG and Pu-WG bearing fuels, respectively. Among the four types of fuel, (U,Pu)O₂ and (Th,Pu)O₂ have almost coincident criticality rundown curves, which show a lower reactivity span than standard UO₂ fuel; the opposite holds for inert matrix fuels.

Figure 1. Reactivity swings for Pu-RG

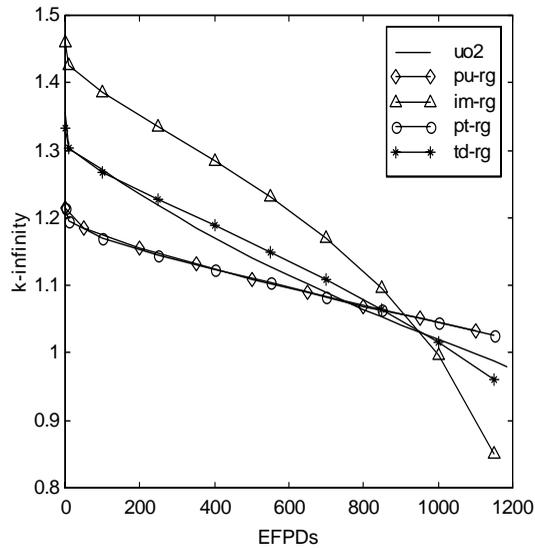
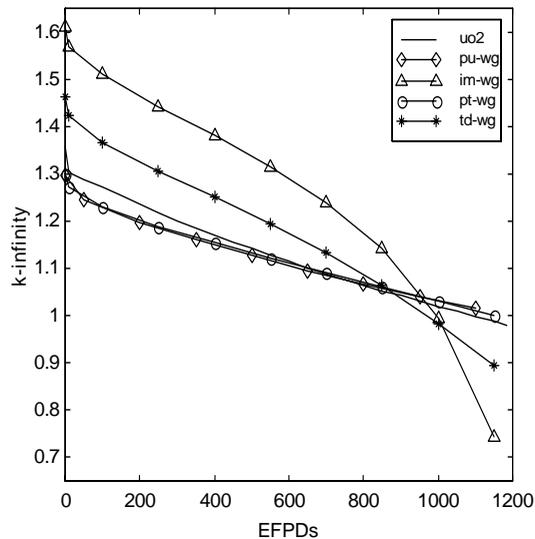


Figure 2. Reactivity swings for Pu-WG



The dynamic coefficients were computed for the four fuel types (Table 2). Because of the lack of any fertile material, the Doppler coefficient for inert matrix fuels is the lowest, while that of thorium-doped fuels is, as imposed from the selected composition, similar to that of UO₂. The same effect can be remarked for the void coefficient (calculated for 10% voidage). The boron coefficient is strongly influenced by the presence of plutonium: its high thermal cross-sections harden the neutron spectrum and cause partial shielding of boron. The resulting coefficients for the plutonium bearing fuels are within 1/3-1/2 of the UO₂ one. This drawback, together with the lower delayed neutron

Table 2. Reactivity coefficients

	DC, pcm/°C		VC, pcm/%void		BC, pcm/ppm
	BOL	EOL	BOL	EOL	BOL
<i>UO₂</i>	-2.64	-2.96	-132	-184	-9.99
<i>PU-RG</i>	-3.00	-2.79	-174	-203	-3.08
<i>PU-WG</i>	-3.19	-3.01	-177	-211	-4.36
<i>IM-RG</i>	-0.99	-0.36	-87	-67	-4.61
<i>IM-WG</i>	-0.98	-0.01	-49	+58	-4.92
<i>PT-RG</i>	-3.29	-3.15	-167	-175	-2.98
<i>PT-WG</i>	-3.55	-3.36	-160	-155	-3.99
<i>TD-RG</i>	-2.32	-2.08	-138	-153	-4.28
<i>TD-WG</i>	-2.33	-2.03	-105	-82	-4.95

fraction, makes more difficult the control of a plutonium-loaded reactor, thus probably requiring, for a whole core loading, a different control rod design and the adoption of enriched boron as a soluble poison.

A whole core IMF loading is not feasible, due to the foreseeable very large power peaks and the dynamic coefficients close to zero.

The plutonium balance for the various fuel types is shown in Table 3. The fraction of burnt plutonium increases going from MOX to (Pu,Th) to thorium-doped to IMF.

Table 3. Plutonium balance (kg/TWh th)

	Pu _{fiss}				Pu _{tot}			
	BOL	EOL	balance	burnt %	BOL	EOL	balance	burnt %
<i>PU-RG</i>	60.6	39.5	-21.0	34.8	84.1	64.5	-19.6	23.3
<i>PU-WG</i>	43.6	21.8	-21.7	49.9	46.4	33.2	-13.1	28.3
<i>IM-RG</i>	42.9	3.4	-39.5	92.0	59.6	15.0	-44.6	74.8
<i>IM-WG</i>	45.5	1.6	-43.9	96.4	48.4	7.5	-41.0	84.6
<i>PT-RG</i>	69.0	28.5	-40.5	58.7	95.9	54.4	-41.5	43.3
<i>PT-WG</i>	54.2	12.6	-41.7	76.8	57.7	22.6	-35.1	60.8
<i>TD-RG</i>	45.3	7.5	-37.8	83.5	62.9	21.7	-41.1	65.4
<i>TD-WG</i>	43.0	3.1	-39.9	92.8	45.8	9.5	-36.3	79.2

Assembly and core calculations

Inert matrix fuels do not appear to be suited for a full core loading; a simple pin cell calculation is not enough to assess their behaviour, but more detailed neutronic calculations are needed. We assumed a core fully loaded with fuel assemblies in which a regular pattern of 56 rods over a total

of 264 (-21%) were replaced with inert matrix fuel pins containing RG plutonium. The neutronic calculations of the fuel assembly were performed with the CASMO-3 code. Due to this limited amount of U-free fuel, both reactivity swings and reactivity coefficients were verified to be only slightly worse than those of a UO₂ fuelled reactor [1].

In order to dampen the peaking factors and then reduce the maximum rod linear heat rate, the utilisation of burnable poisons was considered. The best performing one was IFBA which permits obtaining only a moderate increase of the power peaks, with respect to a standard fuel assembly, and a significant reduction of the soluble boron content of the core.

The inert matrix fuel confirmed the very good plutonium annihilation capabilities: more than 98% of the loaded ²³⁹Pu was burnt, and 90% of fissile plutonium and 73% of the total loaded reactor-grade plutonium was consumed. The fissile fraction of the small amount of remaining plutonium is only 27%, thus the residual plutonium is unattractive for any attempt of recuperation for misuse. These figures are close to those obtained in the pin cell calculations.

Variants on (Pu,Th) fuel

Inert matrix pins, despite the high fraction of burnt plutonium, can only be partially loaded in the reactor core; this imposes restrictions on the yearly amount of destroyed plutonium. Then we considered again the utilisation of (Pu,Th) fuels, and performed new pin cells calculations, seeking enhanced burning performances.

It seems reasonable to reduce the ThO₂/PuO₂ ratio, for lowering the initial loss of reactivity and then obtaining an increment in the incinerated plutonium. More cases are here presented for Pu-WG and for Pu-RG, and namely: reduction of pellet diameter, adoption of hollow pellets (both with the same plutonium content), increase in Pu content, and a combination of them. The results are detailed in Table 4. The 7.2 var. 3 yields for both plutonium types almost the same plutonium elimination capability as the standard inert matrix fuel. Therefore it seems that this thorium fuel can be viewed as a viable alternative to IMF.

Table 4. Combination of several modifications on the (Pu,Th)O₂ pellet design

Ø mm	EFPD		Pu _{fiss} BOL		Pu _{fiss} burnt		Pu _{tot} BOL		Pu _{tot} burnt	
	days		kg/TWh		%		kg/TWh		%	
	RG	WG	RG	WG	RG	WG	RG	WG	RG	WG
8.2 *	1 185	1 185	69.0	54.2	58.7	76.8	95.9	57.7	43.3	60.8
8.2, var. 1	1 499	1 387	54.6	46.3	71.4	85.8	75.8	49.3	54.5	70.8
8.2, var. 2	2 026	2 082	60.6	46.3	66.2	84.9	84.1	49.3	50.3	70.4
8.2, var. 3	2 316	2 243	53.0	43.0	74.4	90.0	73.6	45.7	58.0	77.0
7.2	1 652	1 427	49.5	45.0	77.8	88.2	68.8	47.9	59.3	72.6
7.2, var. 1	1 791	1 509	45.7	42.6	83.2	91.9	63.4	45.3	65.0	77.6
7.2, var. 2	2 513	2 298	48.8	42.0	80.0	92.0	67.8	44.6	62.4	78.7
7.2, var. 3	2 645	2 344	46.4	41.1	84.0	94.2	64.5	43.8	66.8	82.1
8.2, IM	1 185	1 185	42.9	45.5	92.0	96.4	59.6	48.4	74.8	84.6

* Reference case

var. 1: hollow pellet (volume reduction 25%)

var. 2: increase in the Pu content (+50%)

var. 3: combination var. 1 + var. 2

Proliferation issues

During irradiation, thorium-bearing fuels generate the highly fissile ^{233}U , a major concern from the proliferation standpoint. A potential deterrent to diversion, however, is the gamma activity of daughter products of ^{232}U , which emit highly penetrating photons, rendering the spent fuel difficult to handle and easy to safeguard. Another countermeasure for the proliferation risk could be the addition of small amounts of ^{238}U in order to denature the uranium mixture.

The effect of adding some ^{238}U is displayed in Figures 3 and 4 for reactivity and burn-up, respectively. The reactivity is decreased for lack of self shielding of the ^{238}U . This penalty is translated in terms of burn-up reduction, which turns out to be less important by increasing initial plutonium content. The reported reactivity and burn-up values are normalised to those of a fuel without ^{238}U . In both figures the two curves refer to the 8.2 ref. case and 8.2 var. 2 case, respectively.

Figure 3. Reactivity vs. ^{232}Th fraction

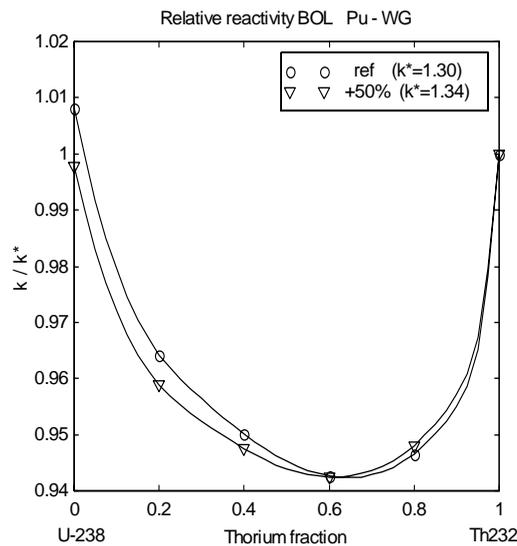
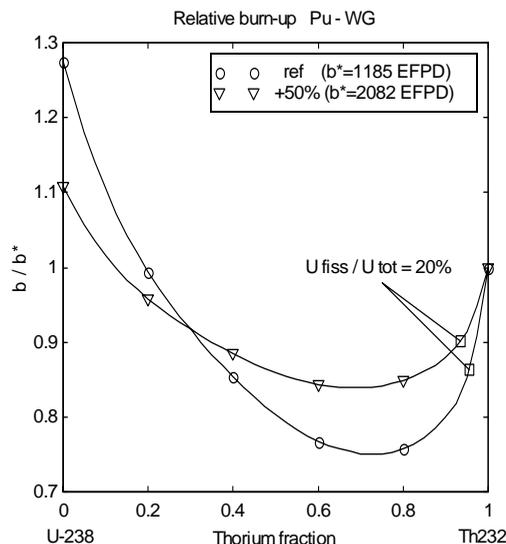


Figure 4. Burn-up vs. ^{232}Th fraction



Fabrication – characterisation and irradiation

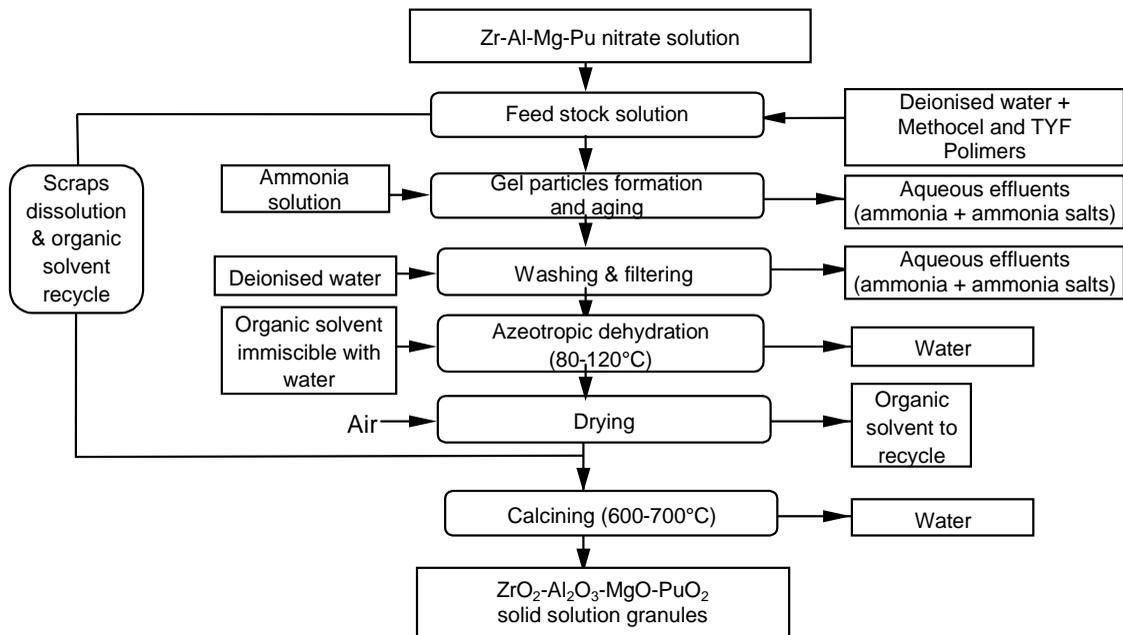
An R&D activity aimed at assessing the experimental response has been undertaken, starting from fabrication-characterisation tests on simulate cold material, with the aim of fabricating a small lot of Pu bearing inert matrix fuel and thoria to be irradiated in a LWR.

The main steps of the programme are listed below:

1. Fabrication of pure matrix and sim-fuel sample pellets (underway).
2. Thermo-physical characterisation, ion irradiation damage (IID) testing and solubility testing on “cold” material (underway).
3. Fabrication of HEU bearing pellets of IMF, including ThO₂, with characterisation and irradiation in Halden reactor.
4. Design, fabrication and irradiation of a representative Pu inert matrix and thoria fuel test experiment, expected to take place in Halden experimental HWBR and/or in a Russian PWR.

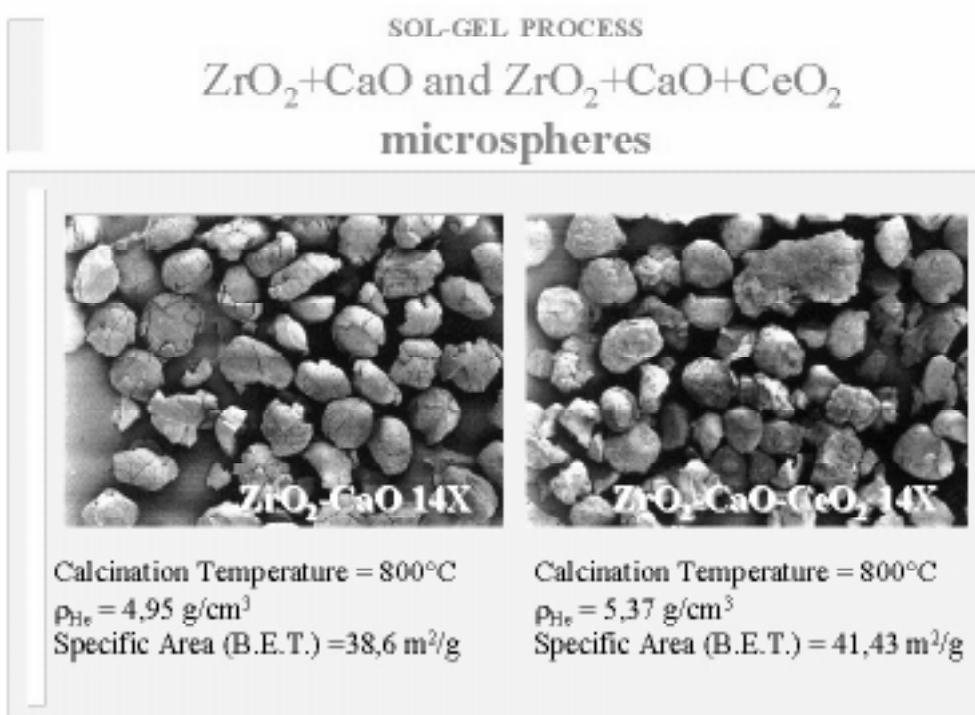
Simulate-fuel fabrication tests are progressing on ZrO₂(CaO), MgAl₂O₄, Al₂O₃-ZrO₂(Y₂O₃)-MgO and ThO₂ matrices. The PuO₂ is simulated by CeO₂. The sol-gel GSP is assumed as the reference process however (Figure 5).

Figure 5. The GSP process for inert matrix fuel microsphere production.



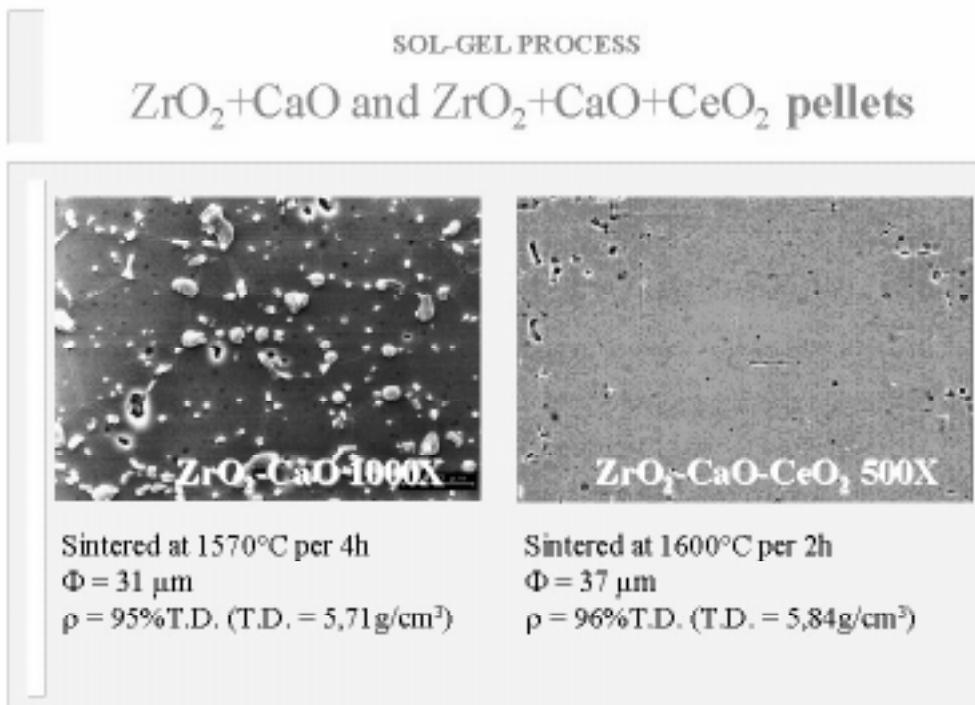
Fabrication tests carried out so far have demonstrated the feasibility of the IMF with respect to the most relevant nuclear grade specifications such as for elemental content, pellet density, geometry and microstructure (Figures 6 and 7). The pre-irradiation picture of the materials is going to be completed through characterisation tests as reported below.

Figure 6. Microsphere microstructures



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Figure 7. Pellet microstructures



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Characterisation testing on simulate fuel refers to the following:

- Measurement of thermo-physical properties such as: thermal conductivity, specific heat, thermal expansion, melting point and molten material behaviour.
- Ion irradiation damage (IID) testing: first exploration on targets by using accelerated 180 keV ions (He, Xe), for different doses ranging from 10^{14} to 10^{17} ions/cm².
- Solubility tests (corrosion and dissolution). Different conditions are simulated: long term geological disposal (about room T, pH ~ 8); fresh/spent fuel reprocessing (HNO₃-8M, 120°C); PWR operation (300°C, 155 bar, pH 7.2).

A multi-property method and apparatus has been developed, qualified and patented. It allows simultaneous measurement of the following thermophysical properties: diffusivity χ , conductivity λ , linear expansion coefficient α , and specific heat c . The method exploits the temperature variations induced via the thermoelastic effect by elastic deformation of a specimen. A homogeneous heat source is produced in a cylindrical sample by uniaxial compression, well within its linear elastic range. The heating process is followed by a thermal relaxation phase at constant stress and null effective thermoelastic source. By recording the temperature of the sample during the thermal transient it is possible to determine directly from the experimental curve the diffusivity χ and the ratio α/c . This method has been calibrated by certificate reference materials (NIST-CRMs) resulting in precise assessments for the uncertainty (below 3%) and the accuracy (better than 10%) of measurements. Preliminary results obtained at room temperature for several zirconia based materials are shown in Table 5.

Table 5. Room temperature thermophysical data

Material Production process	ρ (*) [g cm ⁻³] (T.D.)		c (♦) [J kg ⁻¹ K ⁻¹]	χ (°) [10 ⁻⁷ m ² s ⁻¹]	k (°) [W m ⁻¹ K ⁻¹]	α (°) [10 ⁻⁶ K ⁻¹]
	ZrO₂ + CaO GSP	5.13	(90%)	510	5.4	1.4
ZrO₂ + CaO + CeO₂ Mixed powders – GSP	4.87	(83%)	500	5.2	1.3	10.5
	5.57	(95%)	530	6.0	1.8	7.9
ZrO₂ + Y₂O₃ Coprecipitation – GSP	5.45	(91%)	490	5.4	1.4	9.7
	5.71	(96%)	500	6.2	1.8	8.7
ZrO₂ + Y₂O₃ + CeO₂ Coprecipitation – GSP	5.69	(94%)	520	6.8	2.0	8.1
	5.45	(90%)	510	5.6	1.5	8.9

(*) Archimede's method

(♦) DSC technique

(°) New thermoelastic method

(T.D.) Theoretical density

From preliminary corrosion and dissolution test results emerges that the investigated inert matrix materials are particularly dissolution resistant. This is especially evident for the simulate fuel material containing Ce. Only in very aggressive conditions such as in the combination HNO₃ + HF solubilisation is partially attained. The extrapolated corrosion velocities are extremely low (Table 6).

Table 6. Solubility tests: Dissolution in HNO₃-8M, 1 hour

Sample type	Sample weight (g)	<i>T = 110°C</i> <i>room P</i>	<i>220°C</i> <i>16.4 bar</i>
		Weight loss (%wt.)	Weight loss (%wt.)
ZrO ₂ (Y ₂ O ₃)-Al ₂ O ₃ -MgO pellet	2.5124	<0.01	0.01
ZrO ₂ (Y ₂ O ₃)-Al ₂ O ₃ -MgO+CeO ₂ pellet	2.5633	<0.01	0.03
ZrO ₂ (CaO) pellet	4.9872	<0.06	0.2
ZrO ₂ (CaO)+CeO ₂ pellet	5.2836	<0.01	0.1
ZrO ₂ (Y ₂ O ₃)-Al ₂ O ₃ -MgO Microgranules sintered at 1 550°C	2.5217	<0.11	0.6
ZrO ₂ (Y ₂ O ₃)-Al ₂ O ₃ -MgO+CeO ₂ Microgranules sintered at 1 550°C	2.5066	<0.06	0.4

Concluding remarks

Several nuclear fuel cycles were analysed and discussed with the main emphasis on plutonium consumption, dynamic coefficients and proliferation. The utilisation of the (Th,Pu) fuel cycle appears advantageous due to the following reasons: a) the lack of ²³⁸U prevents the production of new plutonium and then increases the fraction of burnt total plutonium; b) the dynamic coefficients are comparable with the standard UO₂ fuel; c) the fuel shows a good behaviour of the reactivity as a function of burn-up; d) experience exists indicating its excellent behaviour under irradiation and a very stable behaviour in deep disposal conditions. Moreover, the percentage of burnt plutonium is substantially increased by the reduction of the ThO₂/PuO₂ ratio, i.e. reduction of the pellet diameter, the adoption of hollow pellets and the increase of the plutonium content, in order to better exploit the in situ produced ²³³U. Notwithstanding these attractive factors, some drawbacks are also evident in the (Pu,Th) fuel cycle: a) the high thermal cross-sections of plutonium isotopes significantly reduce the worth of control mechanisms such as control rods or soluble boron, thus requiring a different control rod design or the adoption of enriched boron; b) the high fissile ²³³U is generated during the fuel irradiation and this might pose proliferation problems, mitigated by the presence of ²³²U. In any case, the addition of a limited amount of natural uranium (4-7%) eliminates this problem at the expense of a modest reduction of fuel burn-up. However, if the technological aspects of IMF can be positively assessed, this fuel appears particularly attractive for its high plutonium burning capability and complete solution of proliferation concerns.

It can be well imagined that the above options are on a different time scale and then are not one against the other, but complementary in the long run. Moreover the preference for one of these solutions can be determined also by the fuel cycle adopted for future programmes on actinide incineration (via ADS). Thoria-doped fuel can be a reasonable compromise, but it does not eliminate the need for a thorough technological assessment, even worsened by their complex composition.

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CONCEPTS AND FIRST FABRICATION STUDIES OF INERT MATRIX FUEL FOR THE INCINERATION OF PLUTONIUM

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Abstract

Recent aims to destroy plutonium in the most effective way have led to the development of a uranium-free fuel based on zirconia. Concepts for this so-called “inert matrix fuel” (IMF) are presented including first fabrication studies of ceramics, cermers (ceramic + ceramic) and cermets (ceramic + metal). The IMF is foreseen to reduce the amount of fissile plutonium in the spent fuel far beyond the spent fuel standards.

Introduction

Inert matrix fuel has been proposed in order to utilise plutonium in the most effective way and to reduce the amount of fissile plutonium in the spent fuel [1] far beyond the spent fuel standard. By using existing fabrication technology in combination with so-called “inert matrix materials” like zirconia and spinel, disposal of spent fuel would become easier, and public acceptance of peaceful utilisation of plutonium for nuclear energy might be increased. PSI uses the internal gelation process [2] since it takes many years to prepare experimental amounts of a variety of nuclear fuels like UO_2 , MOX and nitrides. This technology could also be used for the fabrication of uranium-free fuel. Concepts of a solid pellet with the drawback of higher centreline temperature as a result of the low thermal conductivity as well as annular pellets [1] and ceramic/ceramic (CERCER) and ceramic/metal (CERMET) material combinations have been proposed. By selecting an appropriate zirconia/spinel ratio, it is possible to overcome the low thermal conductivity of zirconia. Also the feasibility of a CERMET consisting of fine particles bearing plutonium in a cubic zirconia dispersed in a metallic matrix is being investigated as a second option to increase the low thermal conductivity of zirconia. In the following paper a status report of ongoing work is given.

Concepts for the fabrication of ceramics, cercers and cermets

Stabilised zirconium oxide as a fissile material carrier consists of many components like zirconium, yttrium or calcium as stabilising elements, erbia as burnable poison for some fuel concepts and of plutonium as fissile material. The solid solution might be achieved by a co-milling or a coprecipitation process. Different concepts for the fabrication of inert matrix materials, including some effects of parameter changes, are outlined in the following text.

Ceramics

For the fabrication of homogeneous single phase material ceramics, there are two possible fabrication procedures recommended by PSI. Coprecipitated ceramics for classical pellets can be fabricated either from microspheres (internal gelation) or using a powder coprecipitation route. The experimental set-up for the gelation of microspheres has already been described elsewhere [2]. The process used here is one of several sol-gel type processes and is called “internal gelation”. Its principle is a conversion of a highly concentrated metal nitrate solution to a spherical particle of metal oxide with hexamethylene-tetramine as key reactant. The equipment in the PSI laboratories allows sphere fabrication with sizes varying from 800 μm down to 40 μm .

The standard procedure for the fabrication of ceramic pellets used at PSI for zirconia is based on coprecipitated powder which is afterwards milled, pressed into pellets, calcined and sintered [4].

The precipitation is performed with ammonia, i.e. by adding drop by drop to the nitrate solutions until the complete precipitation is finished. Afterwards the precipitate is filtered on a glass filter and washed with Milli-Q-water until a pH-value of 8 is reached. Then a washing step with ethanol follows. The final result is a wet cake containing the hydroxides of the simulated fuel system. The wet cake is dried in a furnace at 120°C, and then the material is crushed. The process is continued by a milling procedure (1.5 h, zirconia jar, Retsch), afterwards a calcination (800°C, 5 h, air), another milling step (2.5 h) and final drying.

The pelletising is carried out by using an automatic press with a pressure between 150 and 300 MPa for 10 seconds. Usually the powder is pressed to pellets with a diameter (d) of about 10 mm and a height (h) in the range of $1.0 \leq h/d \leq 1.5$. The sintering needs to be done in an oxygen containing atmosphere (air) at a minimum of 1400°C. Similar low temperatures are applied for the material prepared by gelation, and high densities have been reached.

Cercer materials

Zirconia microspheres embedded in a spinel matrix is one cercer concept which is currently being examined in several institutes [5]. The flowsheet in Figure 1 outlines the different steps for sphere fabrication and the compaction of a compact pellet based on spray coated microspheres resulting in an optimal distribution of the spheres in the matrix. In this work, a cercer material consisting of a zirconia/spinel combination in a very intimate mixture was also fabricated according to the internal gelation procedure. Table 1 displays the fabrication parameters for a compound with 60% $(Zr_{.76}Y_{.12}Ce_{.12})O_2$ -40% $MgAl_2O_4$ (spinel), in which cerium is used as simulant for plutonium in this scouting experiment. The result in this case is a fine dispersion of the fissile material carrier in the conducting matrix.

Figure 1. Flow diagram for manufacturing of ZrO_2 -Zr cermet and ZrO_2 - $MgAl_2O_4$ cercers

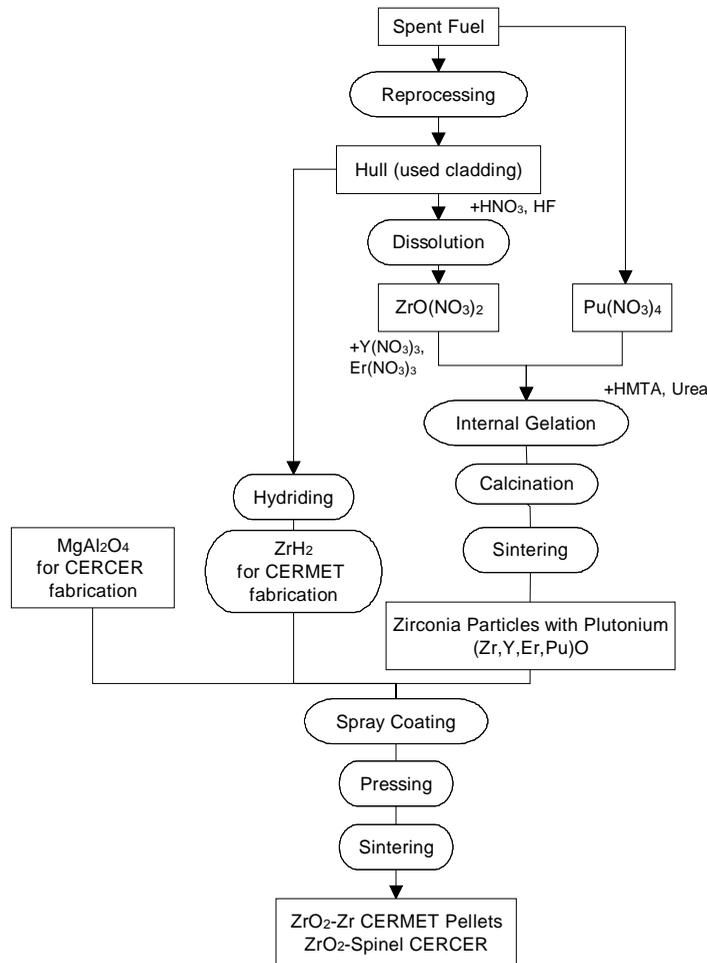


Table 1. Fabrication parameters for microspheres (internal gelation procedure)

Composition of oxides	Concentration of metal [mol/kg solution]	HMTA*/metal	HNO ₃ / (urea + metal)	Remarks
(Zr _{.80} Y _{.10} Ce _{.10})O ₂	0.621	1.00	0.158	About 800 g materials were fabricated (size fractions: 800 μm and 250 μm)
60%(Zr _{.76} Y _{.12} Ce _{.12})O ₂ - 40%MgAl ₂ O ₄ (spinel)	0.800	1.12	0.242	Only the coarse fraction (diameter 800 μm) was fabricated

* Hexamethylene-tetramine

Cermets

A further concept for producing a suitable inert matrix material with good thermal conductivity would consist of fine particles bearing plutonium in a cubic zirconia matrix, which could be fabricated by the conversion of a ZrO(NO₃)₂ + Y(NO₃)₃ and Er(NO₃)₃ solution and a Pu(NO₃)₄ solution (together with HMTA and urea as additives). After the gelation, washing and an appropriate thermal treatment plutonium bearing cubic zirconia particles would be obtained; by using a spray coating technique for adding ZrH₂, followed by pressing, dehydrating and sintering it is feasible to fabricate ZrO₂-Zr cermet pellets (see Figure 1).

Results

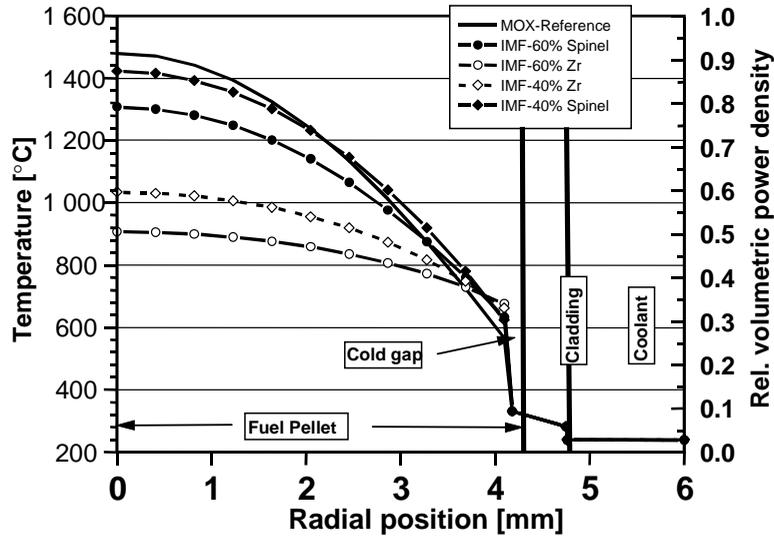
Experimental investigations on this topic are still ongoing, therefore the emphasis will presently be upon (a) the calculations performed to characterise the temperature distribution in a ZrO₂-MgAl₂O₄ cermet and ZrO₂-Zr cermet fuel compared to ZrO₂ and MOX fuels; and (b) on the application of the internal gelation process.

Calculations of the temperature distribution in a fuel pin

As a result of the low thermal conductivity of zirconium-based inert matrix fuel, the centreline temperature of a solid pellet calculated for a standard Halden BWR geometry and with linear power (of the fuel pin) of 400 W/cm is about 300°C higher than that of MOX, see Ref. [1]. Figure 2 shows the temperature distribution of cermet and cermet in two different ratios for fuel pellets, again calculated with a linear power of 400 W/cm. The calculations were performed with the programme PINTEMP [3]. Calculations for MOX are added for comparison. The lowest temperatures (< 1 000°C) were obtained for the pellet IMF-60% zirconium metal (IMF: inert matrix fuel); also for the IMF-40% zirconium metal the temperatures are even lower than for MOX and cermet materials. The higher the metal composition, the lower the centreline temperature of fuel, as shown in Figure 2.

Figure 2. Calculated BOL temperatures in fuel pins with pellets of homogeneous IMF (zirconia-based) and heterogeneous IMF (i.e. zirconia in spinel, cercer, and zirconia in Zr-metal, cermet)

Calculations are valid for Halden BWR conditions and a linear power of 400 W/cm



Gelation studies

To prepare the starting solutions for fabricating zirconia spheres (doped with Y_2O_3 as stabiliser for the cubic modification and CeO_2 as stand-in material for Pu), oxide powders of ZrO_2 , Y_2O_3 and CeO_2 were dissolved in nitric acid.

Typical compositions of the solution used for the preparation of ceramic microspheres are shown in Table 1 for the oxide compound $(Zr_{.80}Y_{.10}Ce_{.10})O_2$. The effects of the variation of the key parameters during the gelation procedure have been evaluated in small size gelation experiments and are described as follows.

Metal concentration

A high metal concentration exhibits a high gelation tendency, which leads to better spherical structures with a high density and crystallinity afterwards. However, the solubility of the different metal nitrates limits the maximum concentration of the feed solution.

Hexamethylene-tetramine content

HMTA decomposes into an amino and hydroxyl group at elevated temperatures; an excess HMTA content will ensure the complete precipitation of metal hydroxide. However, it also increases the possibility of previous gelation of the feed solution at low temperature. A high HMTA content leads to an amorphous material after calcination. The organic component HCOH left inside the spheres is not preferred and will increase the porosity of spheres during the heat treatment.

Acid content

The acid content strongly inhibits the gelation process by modifying the pH value of the solution far from the so-called point of zero charge (PZC). Consequently, a polymer network within the gel spheres will be lost to some extent and thus the structure will contain more NH_4^+ and organic groups, which results in a high porosity after heat treatment. Also inorganic Cl^- or NH_3^- ions are not easy to be washed off, except by using water (which is not recommended here). On the contrary, the acid has a catalytic effect on the decomposition of HMTA.

Urea content

A low urea content will help to stabilise the feed solution at low temperature. However, it contains also unwelcome organic components. Urea will lightly hydrolyse into a NH_4^+ and OH^- group, more or less induce previous gelation slowly. A very high urea content seems to be feasible to produce more rigid spheres (urea works as a binder in the gel network), of course accompanied by more acid in the feed solution in order to prevent earlier gelation.

Gelation temperature

The influence of the gelation temperature is usually considered prior to other parameters, from the point of view of conservative sol-gel science. Here it can be expected that the gelation temperature might affect both the gelation kinetics and the decomposition rate of HMTA or urea, and it will have in addition a catalytic effect of acid on HMTA. The gelation temperature will also be limited later by the plutonium content of the inert matrix. Up to now the internal gelation temperature on the lab scale has varied from 100°C to 105°C.

Heat treatment

The cracking of spheres is usually caused by high surface tensions and a large capillary pressure of liquid inside the structure. The choice of the sintering parameters is therefore important, in particular when regarding the high densification and phase transformations.

The drying of the spheres is usually done at 80-100°C in air. A constant atmosphere will prevent the huge difference between capillary force and pressure outside the pores.

During the calcination a temperature according to the thermo gravimetric analysis (TGA) results for the transformation of the amorphous zirconia to the cubic structure is applied, a slow heating rate and long holding time in air will help transforming into oxide. During the sintering step there is a competition between densification and crystallisation. The former should be controlled to take place earlier than the latter. Due to the fact that the crystallisation is a relatively time consuming process, a previous fast sintering at a temperature lower than the sintering temperature, followed by a slow crystallisation procedure at the sintering temperature is recommended.

As a main result, the gelation technology has been shown to work successfully for both zirconia and zirconia/spinel. Figures 3 and 5, respectively, show the gelled products after drying. The zirconia spheres look very homogeneous, whereas the zirconia/spinel spheres still have to be optimised

Figure 3. G-80Zr-10Y-10Ce – microspheres of zirconia (after drying), photo taken from light microscope; sphere diameter about 800 μm

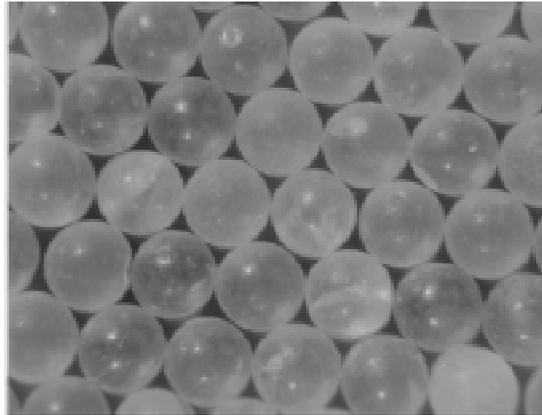


Figure 4. G-80Zr-10Y-10Ce – microspheres of zirconia (after sintering), SEM photography, sphere diameter about 250 μm

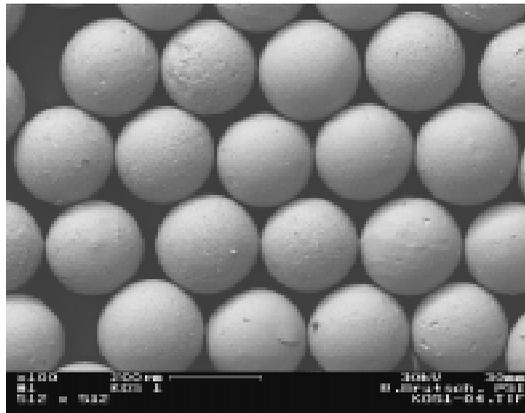
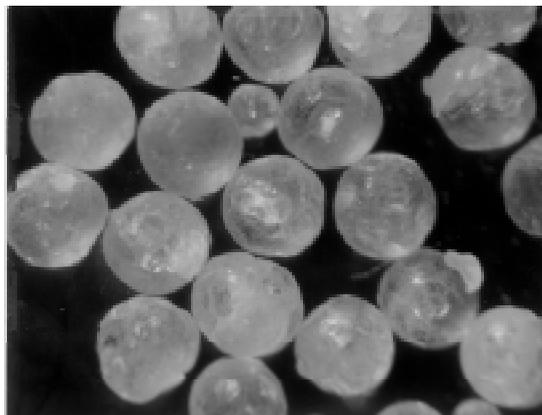


Figure 5. G-48Zr-6Y-6Ce-40MgAl₂O₄ – microspheres of zirconia/spinel (after drying), photo taken from light microscope, diameter of spheres about 800 μm



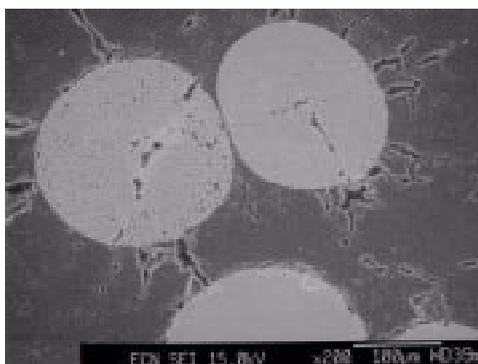
concerning their shape and size (work on this is still ongoing). The zirconia spheres were calcined (500°C, 4 h, air) and afterwards sintered (1350°C, 4 h, air). Figure 4 shows a scanning electron microscopy photo of small zirconia spheres (diameter about 250 μm) after sintering. The X-ray diffraction results confirm that we obtained a cubic single-phase material with a lattice parameter $a = 0.5168 \text{ nm}$. The density of the sintered zirconia product was measured with a helium pycnometer and found to be 5.898 g/cm^3 ($> 95\%$ of the theoretical density value).

Figure 6 shows the prototype of a cermet material, i.e. zirconia microspheres in a zirconium metal matrix. However, the sintering procedure for this samples is still to be optimised, since X-ray results indicate that the metal matrix is not pure zirconium but $\text{ZrH}_{0.25}$. Another problem during the fabrication and characterisation of the cermet was the undesirable reaction of zirconium metal with nitrogen coming from the air. As a result, ZrN was formed and caused heavy problems; it was nearly impossible to yield a highly densified material. Figures 7 and 8 show the prototype of a cermet material, i.e. zirconia microspheres in a spinel matrix. If the zirconia spheres are only calcined (and not sintered), the result is quite satisfactory concerning the contact between spheres and matrix; however the distribution of the spheres in the matrix could be optimised in future experiments. If the microspheres (see Figure 7) are sintered before embedding them into the spinel matrix, significant cracks are formed in the material. This is due to the different thermal and mechanical behaviour of presintered zirconia and unsintered spinel during the heat treatment of the pellet.

Figure 6. O-889: Cermet consisting of zirconia-spheres in zirconium hydride environment

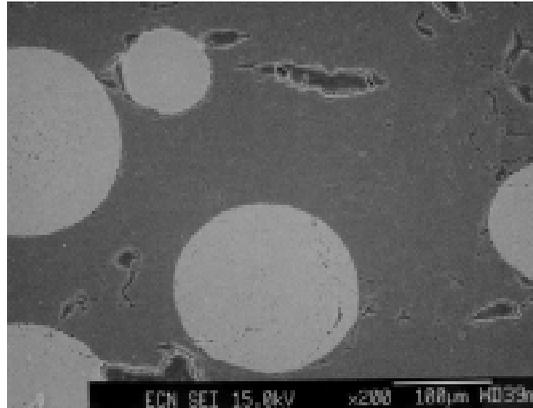


Figure 7. Cermet material: calcined and sintered zirconia spheres in spinel matrix*



* Microspheres fabricated by PSI, cermet pellet and photos by R. Konings (ECN Petten).

Figure 8. Cercec material: Calcined zirconia spheres in spinel matrix*



Discussion and outlook

The motivation for the investigation of inert matrix materials is mainly the reduction of fissile plutonium in the spent fuel. To show that the fabrication of ceramics for this purpose is feasible, spheres of zirconia (doped with ceria and yttria) and spinel were gelled, dried, calcined and sintered followed by X-ray analysis, chemical analysis, ceramography and technological characterisation. The gelation parameters like gelation time, temperature conditions, pH-value of the solution and the influence of nitric acid, HMTA (hexamethylene-tetramine) and urea contents on the gelation behaviour were investigated in detail.

A concept combining microsphere fabrication with afterwards spray coating and sintering has been proposed for the cermetts (ZrO_2 in Zr-matrix) as well as for the cercers (ZrO_2 in spinel matrix). Temperature profile calculations prove the theory that the thermal conductivity of “pure” zirconia can be significantly increased by combining it with metals or other ceramics. For the future, work at PSI will consist of further characterisation of the IMF fuel (ceramics, cercers and cermetts) with emphasis on the description of the irradiation behaviour.

* Microspheres fabricated by PSI, cercec pellet and photos by R. Konings (ECN Petten).

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URANIUM-FREE BURNER REACTOR DEDICATED TO MINOR ACTINIDES TRANSMUTATION*

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Abstract

This paper proposes a large (1 500 MWe) uranium-free fast reactor devoted to the transmutation of minor actinides, with high initial minor actinides loading, homogeneously mixed with the fuel. To restore acceptable values of the Doppler effect and the sodium void reactivities, ^{238}U is replaced by other nuclides which have capture resonances at appropriate energies (tungsten or ^{99}Tc , is a long-lived fission product and can thus be transmuted with profit) and by a hydrogenated moderator separated from the fuel. The transmutation performance of such a core is 70 kg/TWhe for plutonium (due to ^{238}U suppression) and 30-35 kg/TWhe for minor actinides; that is, a consumption rate of about 100 kg/TWhe of transuranic element. Approximately 14 kg/TWhe of ^{99}Tc are destroyed when this element is present. The activity of such subassemblies has been investigated in comparison to standard fast reactor subassemblies (neutron sources, decay heat and gamma dose rate). Finally, a transmutation scenario involving PWRs and minor actinide burning fast reactors has been studied to estimate the necessary proportion of burner reactors and the achievable radiotoxicity reduction with respect to a reference open cycle.

* This work was performed during the Ph.D. work of the first author at CEA/CE, Cadarache, France.

Introduction

The management of long-lived nuclear waste, such as minor actinides (MA) (^{237}Np , ^{241}Am , ^{243}Am , etc.) and some fission products (^{99}Tc , ^{129}I , etc.) is a major strategic issue for future nuclear power generation.

Within the framework of the law passed in December 1991 concerning a pluriannual programme called SPIN (a French acronym for improved SeParation and Incineration), France is currently investigating several nuclear waste management options: chemical separation and recovery, burial in deep depositories, interim storage, incineration in present and/or innovative reactor systems.

In the field of MA incineration in a conventional fast reactor, studies have shown that the initial MA enrichment has to be limited to about 2.5% and 5% for EFR type cores and CAPRA type cores, respectively; the MAs are directly mixed with the fuel (homogeneous recycling). Within these limits, MAs do not significantly alter the safety parameters, particularly the sodium void and the Doppler effects [1,2]. In fact, loading a per cent of MA homogeneously in the core means elimination of the same quantity (or more, if moderator presence is needed) of ^{238}U . ^{238}U has a strong influence on the Doppler effect (~ 95%), on the neutron reactivity loss (by conversion into ^{239}Pu) and on the delayed neutron fraction (~ 50%) [3].

As a result, in the conventional fast reactor, we cannot recycle an important quantity of minor actinides mixed with the fuel (e.g. to increase the MA consumption) in keeping ^{238}U in the core. Thus the scope of this paper is to propose a special core dedicated to MA transmutation, by completely replacing ^{238}U with other materials. This will permit a two-fold aim: (1) keeping adequate values of safety parameters on the one hand; and (2) loading an important quantity of MA to be transmuted on the other hand. This MA incinerator fast reactor will then be integrated into a nuclear park to evaluate its efficiency in terms of waste radiotoxicity reduction of this park.

The reference core design is an oxide CAPRA type with dilution approach [4,5]. This core is highly enriched in plutonium (average Pu enrichment is about 42.21%). The choice of this kind of core for our study is related to: (1) its large power level (1 500 MWe); and (2) its dilution design (e.g. 469 pins per S/A and only 320 of them contain fuel pellets: version 9/93). This design will allow us to introduce a moderator in the fuel free pins in order to improve the sodium void and the Doppler effects which, we expect, will be affected by the presence of MAs in the core. Using this core design, the moderator is separated from the fuel.

Technetium (^{99}Tc) and natural tungsten (W) were chosen to replace ^{238}U in the core because both of them have high temperature dependent capture cross-sections ratio at appropriate energies (higher than that of ^{238}U in the case of technetium) (see Figures 1, 2 and 3). Their capture resonances are located between some keV and 100 keV, where the neutronic density is important [6]. In addition, technetium is a long-period fission product (2.13×10^5 yr. of half-life and 2.94 kg/TWhe of production in PWRs) and can thus be transmuted with profit [7].

To test their performances in terms of the Doppler reactivity feedback without ^{238}U , ^{99}Tc and W were added individually to the oxide fuel (PuO_2) in the reference CAPRA core. Due to the high capture rate of ^{99}Tc and W (compared with that of ^{238}U), the initial reactivity decreased, therefore the fuel enrichment (PuO_2) was adjusted as required to keep the same initial reactivity of the reference case.

The following calculation schemes are used: the JEF-2 cross-section data set, the ECCO code for cell calculations, and the ERANOS (European Reactor ANalysis Optimised System) code system for core calculations. A diffusion theory calculation with a two-dimensional model in RZ geometry has been performed for these preliminary studies.

The results are shown in Table 1. The Doppler effect is established and it is better than the reference case with ^{238}U at the end of life. The ^{238}U suppression increases the loss of reactivity. ^{99}Tc and W are so absorbent that the sodium void decreases their absorption rate. Therefore the sodium void reactivity increases. These effects will be improved by introduction of both minor actinides and a moderator.

Table 1. Safety parameters at beginning and end of life (BOL/EOL)

Safety parameters	Reactivity loss	Doppler effect		Sodium void	
		BOL	EOL	BOL	EOL
CAPRA ($\text{PuO}_2 + ^{99}\text{Tc}$)	$\times 1.5$	-21.5%	+34.5%	$\times 2.1$	$\times 2.0$
CAPRA ($\text{PuO}_2 + \text{W}$)	$\times 2.1$	-25.6%	+77.0%	$\times 1.6$	$\times 2.0$

Definition of uranium-free core for minor actinides transmutation

As we noticed above, the introduction of a moderator is necessary to counterbalance the neutron spectrum hardening induced by minor actinide loading. The moderator calcium hydride (CaH_2) will fill the empty pins in the fuel assembly. To increase the MA transmutation rate and to take advantage of fissile isotopes generated by MA incineration, the total fuel residence time becomes 1 500 EFPD (versus 795 EFPD for the CAPRA reference case). The introduction of both minor actinides and ^{99}Tc (or W) in the core requires an increase of the Pu enrichment. Hence, the composition of the fast reactor core dedicated to a massive MA transmutation is defined as follows.

For NpO_2 or AmO_2 recycling:

- Average Pu content is 66% and 63% (volumetric enrichment) for inner core in the ^{99}Tc case and W case respectively.
- The remainder is half of ^{99}Tc (or W) and half of NpO_2 (or AmO_2).

MA and ^{99}Tc (or W) are mixed homogeneously to PuO_2 .

Safety parameters of the designed core

With the composition defined above, Tables 2 and 3 show that safety parameters are satisfied with respect to the reference case. In effect, Doppler effects are much better for both neptunium and americium recycling at the beginning and the end of life (more than +12%). Minor actinides do not change the Doppler reactivity because their capture resonances are located at some eV, where the neutronic density is very weak. Sodium void reactivity remains almost the same and the reactivity

Table 2. Safety parameters of uranium-free MA (Np) incinerator core with ⁹⁹Tc or W

	CAPRA ((Pu + Np)O ₂ + ⁹⁹ Tc)		CAPRA ((Pu + Np)O ₂ + W)	
Reactivity loss	-29%		-30%	
	BOL	EOL	BOL	EOL
Doppler effect	+56%	+29%	+24%	+12%
Sodium void	×1	×0.7	×1	×0.7

Table 3. Safety parameters of uranium-free MA (Am) incinerator core with ⁹⁹Tc or W

	CAPRA ((Pu + Am)O ₂ + ⁹⁹ Tc)		CAPRA ((Pu + Am)O ₂ + W)	
Reactivity loss	-32%		-33%	
	BOL	EOL	BOL	EOL
Doppler effect	+50%	+39%	+20%	+17%
Sodium void	×1	×0.7	×1	×0.7

loss decreases by about 30% due to fissile isotope generation by MA transmutation. The delayed neutron fraction decreases (~ 30%) due to the ²³⁸U suppression. Indeed, ²³⁸U has a strong influence (some 50%) on this parameter.

Consequences on fuel cycle physical parameters

Tables 4 and 5 show the consequences on cycle physical parameters (α , β , γ , and neutron activities, decay heat and γ dose rate) of dedicated core subassemblies. These values were determined at the fabrication and five years after irradiation. Contributions of both heavy nuclides and fission products were considered (except for γ dose rate).

Table 4. Consequences on cycle operations at the fabrication

		CAPRA (Pu,U)O ₂	CAPRA ((Pu + Np)O ₂ + ⁹⁹ Tc)	CAPRA ((Pu + Am)O ₂ + ⁹⁹ Tc)
Fabrication	Decay heat	×1	×1.6	×2.7
	Activity	×1	×1.6	×1.7
	γ source	×1	×1.6	×7.9
	γ dose rate	×1	×17	×440
	Neutron source	×1	×1.6	×2.3

Table 5. Consequences on cycle operations after five years of cooling time

		CAPRA (Pu,U)O ₂	CAPRA ((Pu + Np)O ₂ + ⁹⁹ Tc)	CAPRA ((Pu + Am)O ₂ + ⁹⁹ Tc)
EOL + 5 years	Decay heat	×1	×3	×4
	Activity	×1	×1.6	×1.8
	γ source	×1	×1	×1.2
	γ dose rate	×1	×1.5	×7.7
	Neutron source	×1	×0.7	×4.7

As we can see, the neptunium recycling has no strong impact on cycle operations; at fabrication, γ dose rates increase due to ^{233}Pa in equilibrium with ^{237}Np (strong γ emitter at about 300 keV). After cooling time, decay heat is about three times that of the reference case because of ^{238}Pu and ^{244}Cm contributions. However, for the americium recycling, at fabrication, the γ dose rate is very high (penalising) because of ^{239}Np in equilibrium with ^{243}Am , and the γ source is eight times higher than the reference case: ^{241}Am is the major contributor. After cooling time, both decay heat and neutron source are important due to Cm isotopes generation.

We can conclude that neptunium recycling does not induce special constraints on cycle operations, which is not the case for americium recycling. The latter case will demand special handling and enhanced protection against γ and neutron emission.

Transmutation performances

The MA incineration performances are expressed in terms of consumption rate which represents the disappearance of the element by fission and by neutron capture over the irradiation period. The total actinides (Pu + MA) consumption reaches about 100 kg/TWhe; more than one-third of this quantity includes the MA consumption (36 kg/TWhe) (see Table 6). These values are higher than the MA production rate in a standard UO_2 fuel PWR (about 3 kg/TWhe) [8]. It is interesting to note that the quantity of long-lived fission product incinerated simultaneously in the MA incinerator core is about 14 kg/TWhe.

Table 6. Transmutation capacities of uranium-free MA incinerator core

	Core with ^{237}Np		Core with Am	
	+ ^{99}Tc	+ W	+ ^{99}Tc	+ W
MA recycled	^{237}Np		Am	
MA consumption (kg/TWhe)	-35.6	-39.8	-31.5	-36.0
MA consumption rate (%)	56	58	45	48
Pu consumption (kg/TWhe)	-68.0	-64.0	-78.4	-75.5
^{99}Tc consumption (kg/TWhe)	-14.0	–	-14.0	–
Cm production (kg/TWhe)	+2.4	+2.8	+14.0	+15.6
Total actinides* consumption (kg/TWhe)	-101	-101	-96	-96

Management of actinides produced by a nuclear park

Introduction

In the above sections, a minor actinides burner core design with a specific initial composition in (Pu + MA) was proposed and its transmutation capacities as an individual reactor were studied. It would be more interesting to speculate upon its transmutation potential face to nuclear park output of (Pu + MA + ^{99}Tc) and its efficiency in terms of radiotoxicity reduction once integrated in a nuclear park.

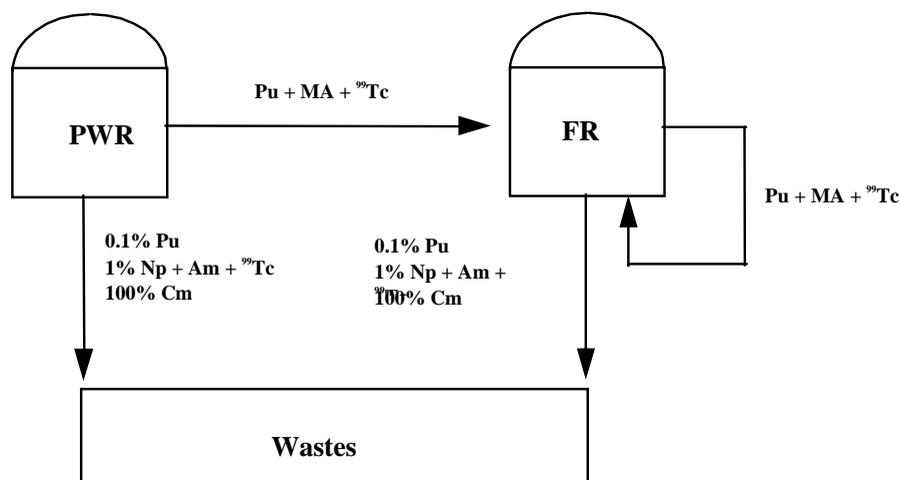
* Total actinides: Pu + Np + Am + Cm.

Description of nuclear park and scenario envisaged

The nuclear park is composed of:

- A proportion of uranium-free fast reactor cores dedicated to MA transmutation (with both americium and neptunium oxide recycled simultaneously).
- A number of (1 500 MWe) PWRs fed with uranium oxide fuel at 4.5% enriched, the burn-up is 55 GWd/t.

A proposed management scenario (e.g. closed cycle) is outlined below:



PWR reactors are sending their output (Pu + MA + ⁹⁹Tc) into incinerator FRs. Multiple recycling is achieved in order to reach an equilibrium state, in which (Pu + MA + ⁹⁹Tc) production and consumption (destruction + losses) balance each other in the whole park. Losses at reprocessing are assumed to be 0.1% for Pu, 1% for Np and Am, and 100% for Cm (Cm not recycled).

Results

After multiple recycling (six cycles, each one 1 500 EFPD), equilibrium is reached. The proportion of uranium-free fast reactor devoted to massive MA incineration needed in a nuclear park is 22.6%. Table 7 shows that at equilibrium, the incinerator core has a different composition (low loading in MA and ⁹⁹Tc) and transmutation potential, but globally the total actinide consumption remains the same.

Table 7. Transmutation performances of incinerator core at equilibrium

	Incinerator core		Core at equilibrium	
	Initial mass (kg)	ΔM (kg/TWhe)	Initial mass (kg)	ΔM (kg/TWhe)
(Am + Np)	3 782	-31.5	2 746	-18.7
Pu	13 374 (27% of ²⁴⁰ Pu)	-78.4	16 006 (40% ²⁴⁰ Pu)	-89.1
Cm		+14		+10
Actinides		-96		-97
⁹⁹ Tc	4 005	-14	2 411	-10.0

There are no noticeable modifications of safety parameters with the core at equilibrium. In fact, the initial ^{99}Tc quantity decreases, but this does not affect the Doppler reactivity due to the ^{240}Pu contribution (the PWR output, which feeds the uranium-free incinerator reactor, is highly enriched in ^{239}Pu , ^{240}Pu and ^{241}Pu) [9].

In terms of radiotoxicity, only TRU radiotoxicity is taken into account because only TRU are recycled in MA fast burner reactors. Besides this, the uranium contribution to waste radiotoxicity would be significant only after 10^6 years.

According to open cycle (e.g. all irradiated PWR fuel is stored without reprocessing), radiotoxicity is reduced by a factor of 25 at around 100 years, but this factor decreases between 10^3 and 10^4 years due to the Cm contribution (Cm is massively present in waste). In fact, Cm is the major contributor to radiotoxicity waste at any time scale (see Figure 4).

If Cm is also recycled, radiotoxicity is reduced by a factor of 100 from 100 years. Up to this time, Cm is the main contributor (Cm is the most active among minor actinides). After 100 years, we notice the contribution of both americium and plutonium isotopes at different degrees. In this case, the proportion of uranium-free fuel incinerator required to reach equilibrium in a nuclear park is 24%.

Conclusions

In the previous sections, it was stated that the design of a uranium-free fast reactor dedicated to a massive MA transmutation is possible and attractive from the point of view of: (1) safety parameters (Doppler reactivity, sodium void effect and loss of reactivity); and (2) transmutation performances (actinides (Pu + Np + Am + Cm) consumption reaches about 100 kg/TWhe, plus 14 kg/TWhe: the quantity of long-lived fission product incinerated). However, consequences on cycle operations and characteristics of uranium-free fuel (e.g. high Pu and Am content) will require the development of new technologies at both fabrication and reprocessing states.

Further safety studies for this new concept of fuel are necessary to demonstrate its feasibility and to evaluate its safety features (e.g. stability of the core, behaviour during a rapid insertion of reactivity, etc.).

Figure 1. Capture cross-section ratio at two different temperatures for ^{238}U and ^{99}Tc

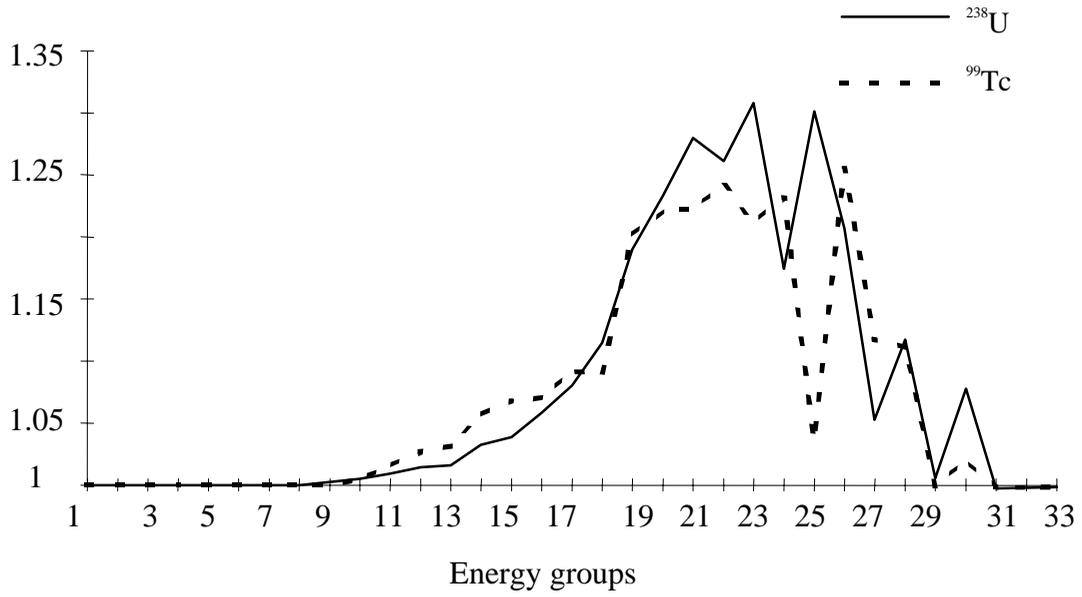
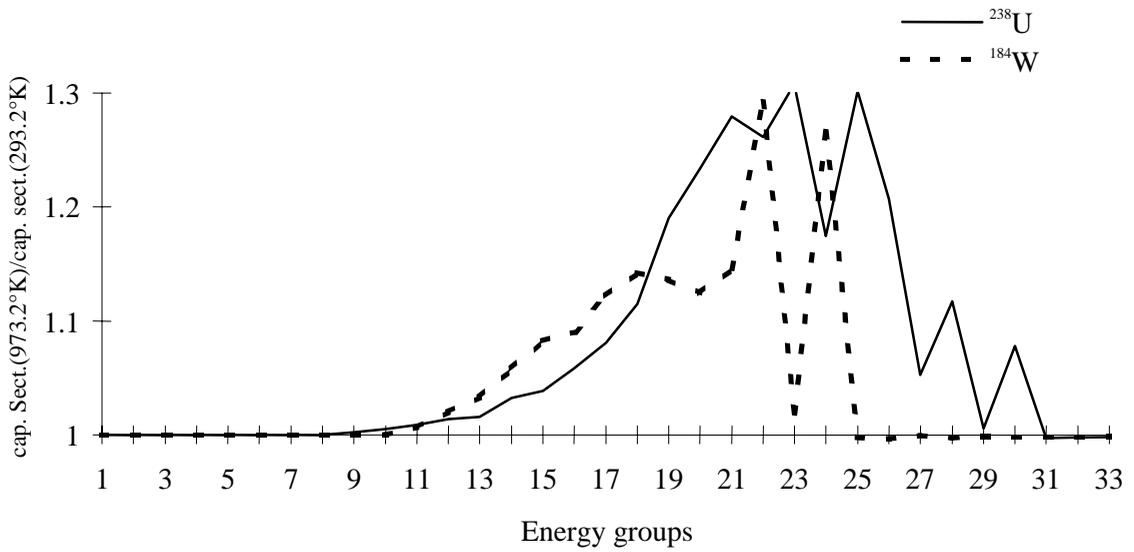


Figure 2. Capture section ratio at two different temperatures for ^{238}U and ^{148}W



Group 11 is equivalent to 111 keV
 Group 18 is equivalent to 3.35 keV

Figure 3. Capture section ratio at two different temperatures for ^{238}U and ^{186}W

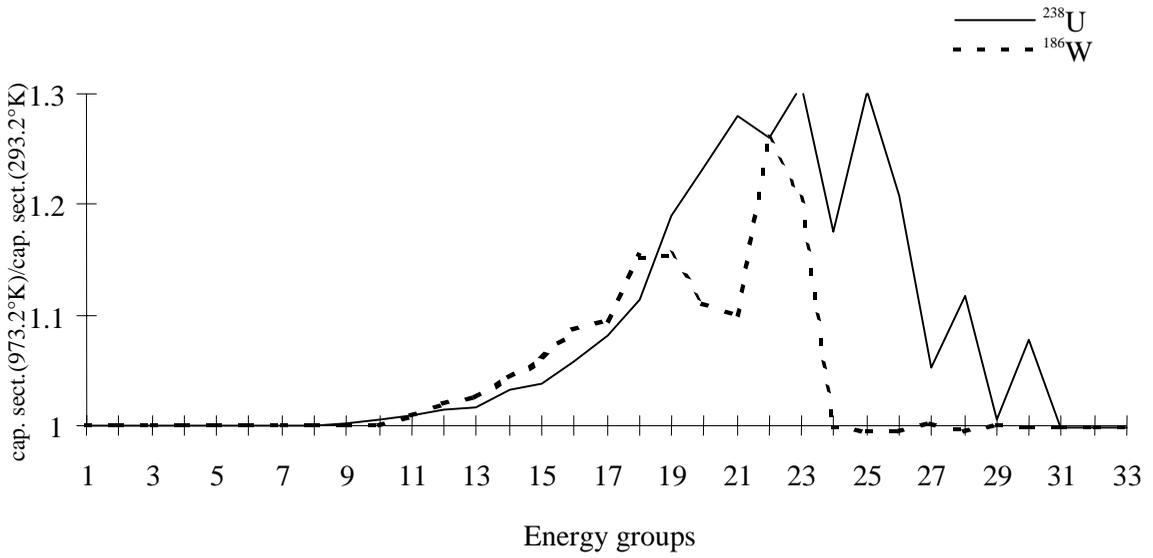
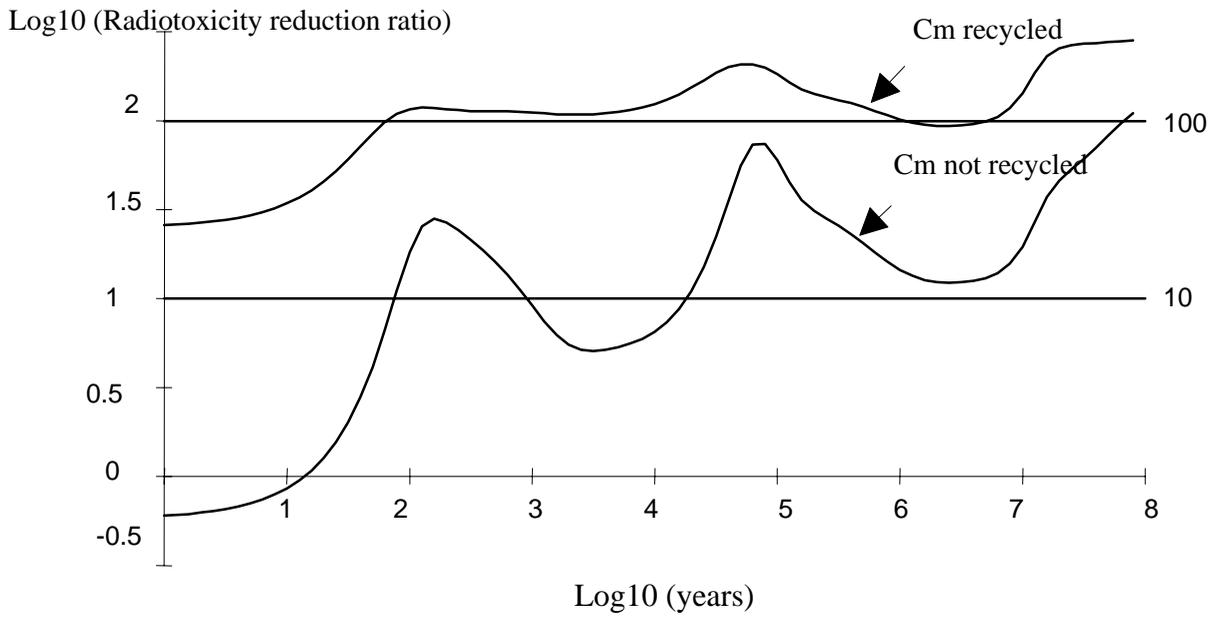


Figure 4. Radiotoxicity reduction ratio of closed cycle (according to open cycle)



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SILICON CARBIDE AS AN INERT MATRIX FUEL FOR CANDU REACTORS

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Abstract

AECL has been studying silicon carbide as part of the current world effort to develop an inert matrix material to burn actinide-waste or military or civilian plutonium, while generating little or no additional actinide wastes or fissile isotopes. This paper reports on the progress of work to develop methods of fabricating SiC to achieve high densities at low sintering temperatures; specific heat and thermal conductivity measurements of as-fabricated SiC; compatibility of SiC with coolant and zircaloy-4; and accelerator simulations of in-reactor fission-fragment damage. Calculations, based on the measured thermal conductivity, show that the inert matrix fuel could operate at 55 kW/m linear power at only 100°C above coolant temperature. In all regards, SiC appears to be a very promising candidate as an inert matrix fuel for water-cooled reactors.

Introduction

Silicon carbide has the following well-established properties, making it a promising candidate for investigation as an inert matrix material:

- Very high thermal conductivity (approximately as high as aluminium for very pure SiC with no porosity), giving the promise of low fuel operating temperatures and improved safety.
- High melting temperature.
- Good chemical stability, e.g. strong resistance to oxidation in air and air-moisture atmospheres (fewer data are available on corrosion in high-temperature water).
- No unacceptable phase changes.
- Low neutron absorption.

In this paper, we report on initial efforts to fabricate SiC with sintering aids and cerium as a plutonium substitute, specific heat and thermal conductivity measurements of SiC, compatibility of SiC with coolant and with zircalloys, and accelerator-based simulations of in-reactor fission-fragment damage to SiC specimens. Reactor physics aspects are discussed in a companion paper at this workshop [1].

Fabrication

One difficulty of fabricating SiC is that it generally requires very high temperatures for pressureless sintering. In addition, it is desirable to use conventional fabrication processes and equipment, e.g. furnaces, that are compatible with UO_2 and mixed-oxide (MOX) fuel fabrication. To address these issues and test fabrication of SiC with a substitute for plutonium, three fabrication campaigns with different objectives were conducted under contract to AECL, at Queen's University between 1996 and 1998.

First fabrication campaign

The first campaign utilised Al_2O_3 , TiO_2 and SiO_2 as sintering aids and aimed to produce silicon carbide at 93-95% of theoretical density (TD) at sintering temperatures below 2 000°C. In addition, cerium was added as a plutonium substitute in two forms, CeO_2 and CeC_2 , in a wide concentration range¹ to determine what densities could be achieved and how the cerium would be incorporated in the silicon carbide matrix. Hardness and fracture toughness tests were also performed, but are not reported here because of space limitation.

In general, the following processes were used for silicon carbide fabrication:

- Blend starting powders, binder (polyethylene glycol), and lubricant (oleic acid), with extra carbon to partially reduce oxide starting powders.

¹ The concentration of CeO_2 equivalent to the reference concentration, 250 g weapons-grade Pu in the outer 30 elements of a standard 37-element CANDU[®] bundle [2], is 3.43 wt.% CeO_2 .

- Ball mill in methanol with ZrO₂ grinding media.
- Dry, crush and screen through a sieve with 1.6 mm openings.
- Uniaxial press (cold) at 50 MPa.
- Isostatic press (cold) at 250 MPa.
- Sinter in graphite resistance furnace in static argon at 1 MPa.

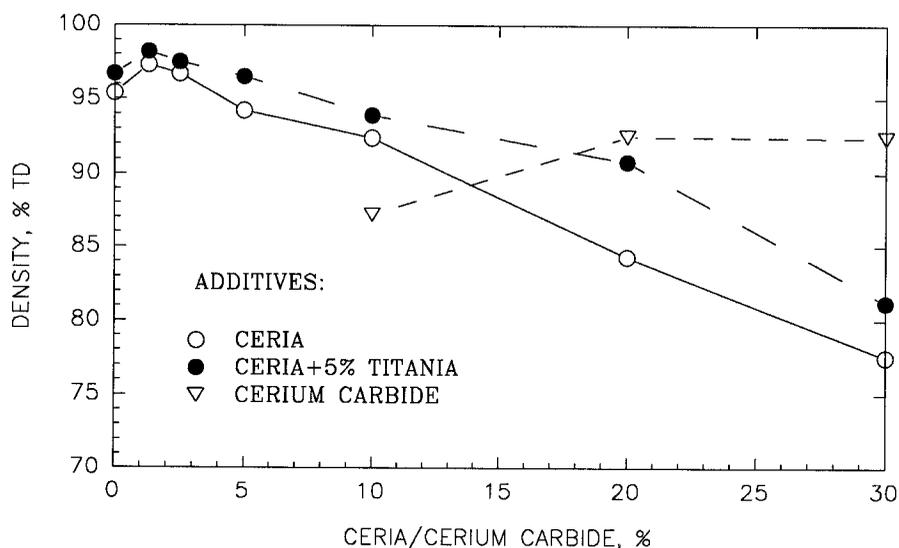
In this first campaign, two groups of sintering powders were used, in addition to SiC powder, carbon black and cerium (used in the form of either CeO₂ or CeC₂):

A) Al₂O₃ (8 wt.%) and TiO₂ (0 and 5 wt.%).

B) Al₂O₃ (2.55-12.5 wt.%), Y₂O₃ (1.08-10.92 wt.%) and SiO₂ (0, 1.3 and 2.6 wt.%).

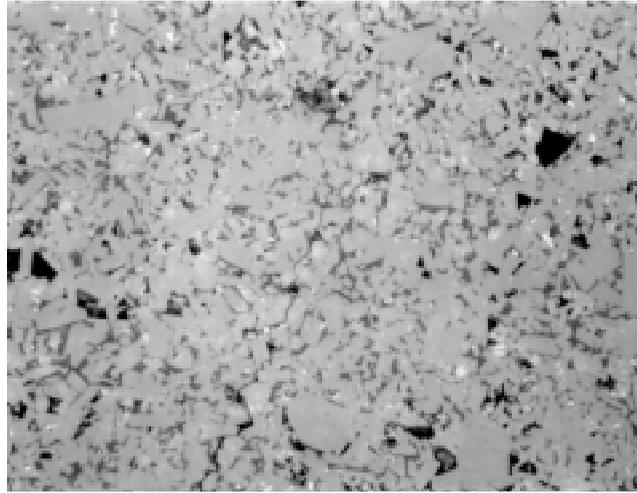
The powders of Group A with CeO₂ concentrations of 0-5 wt.%, sintered at 1 990°C to 2 035°C achieved densities² of 94-97% TD (Figure 1). With increasing CeO₂ concentrations, the densities decreased to about 80% of TD at 30 wt.% CeO₂. The addition of 5 wt.% TiO₂ increased the densities by a few per cent, the amount depending on the amount of ceria used. X-ray diffraction (XRD) analysis showed the formation of AlCeO₃ phase in the samples made with CeO₂, and AlCeO₃ and the intermetallic phase, Al₃Ce, in the samples made with CeC₂. The presence of these phases was confirmed by optical microscopy (Figure 2). In addition, optical microscopy showed the presence of Al₃Ce in the samples made with CeO₂, even though that phase was not detected by XRD.

Figure 1. Densities of SiC with various additives (all utilised Al₂O₃) sintered at ~2 000°C



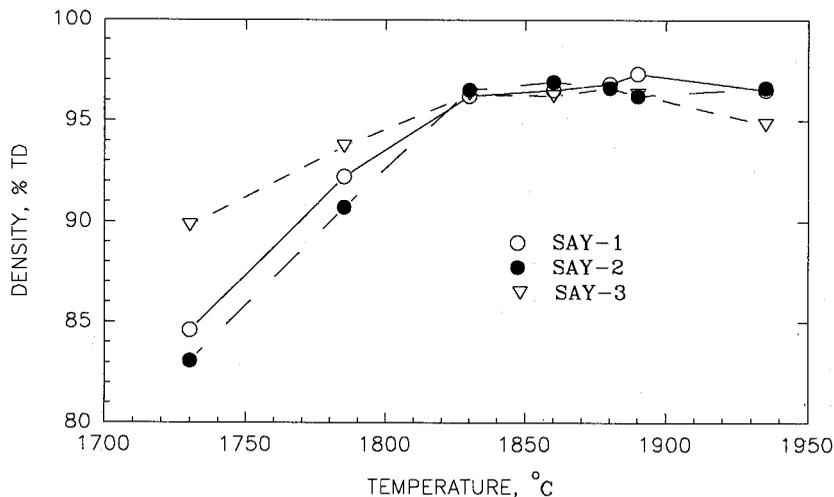
² The theoretical densities were calculated using the knowledge from XRD that TiO₂ transformed to TiC and assuming that CeO₂ transformed to Ce₂C₃. The volume fraction of Al₂O₃ remaining in the sample (some is volatilised during sintering) was determined from weight loss measurements before and after sintering; residual Al₂O₃ ranged from 0 to 5.3 wt.%.

Figure 2. Optical micrograph of a polished section of SiC fabricated with 8 wt.% Al₂O₃ and 5 wt.% CeO₂, sintered at 2 000°C, showing AlCeO₃ (dark grey) and Al₃Ce (white phase). Black spots are sintering porosity; light grey spots in the background are SiC.



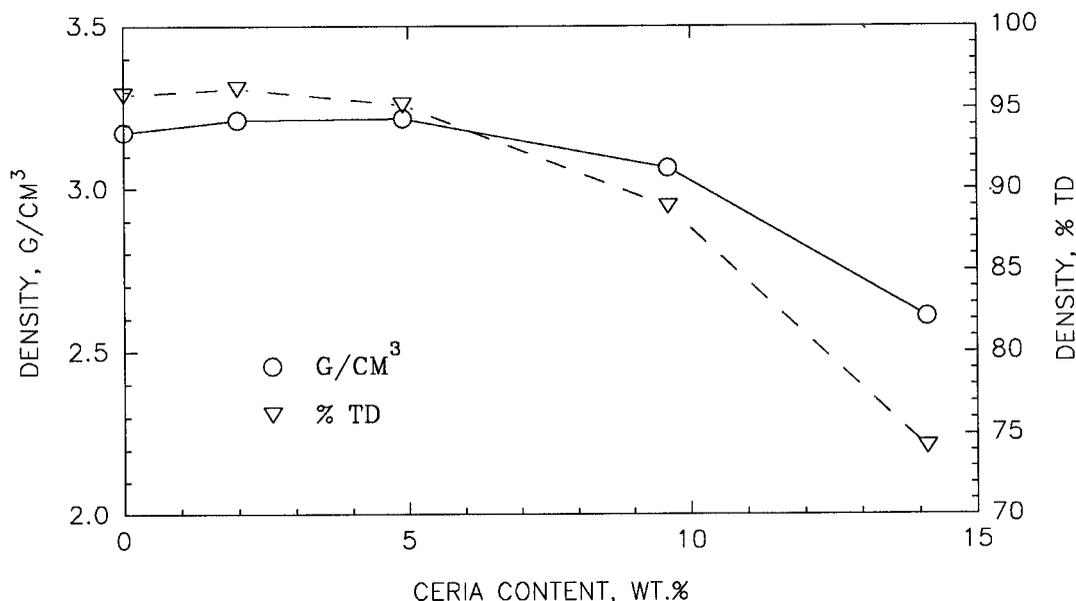
The powders of Group B (with Al₂O₃, Y₂O₃ and SiO₂) yielded high densities at significantly lower temperatures. Figure 3 shows the densities obtained with no ceria; above about 1 830°C, 95-97% of TD was achieved with little dependence on temperature and almost no dependence on the amounts of Al₂O₃, Y₂O₃ or SiO₂ used. Below 1 830°C, the density depended on sintering temperature and the amount of sintering additives, falling at 1 730°C to 90% of TD with 12.5 wt.% Al₂O₃, 10.9 wt.% Y₂O₃ and 2.6 wt.% SiO₂ and to 83% of TD with 9 wt.% Al₂O₃, 3.8 wt.% Y₂O₃ and 0% SiO₂. XRD showed that a yttria-alumina garnet (YAG) phase with formula 3Y₂O₃·5Al₂O₃ formed. This result, improved sintering with Al₂O₃, Y₂O₃ and SiO₂ at 1 830°C and above, had been expected; it is known that YAG, with relatively low melting temperature and surface characteristics that lead to wetting of SiC particles, can form and enhance sintering.

Figure 3. Variation of density with sintering temperature of SiC fabricated with Y₂O₃, SiO₂, but no CeO₂ or CeC₂ (0, 1.3 and 2.6 wt.% SiO₂ for SAY-1, -2, -3, respectively)



When ceria was added below 5 wt.% concentration, the densities were largely unaffected (Figure 4), but above that amount the density decreased to 75% of TD at 14 wt.% CeO₂ (1 860°C sintering temperature).

Figure 4. Variation of density with ceria content for SiC fabricated with 2.55-8.0 wt.% Al₂O₃, and 1.1-3.4 wt.% Y₂O₃; sintering at 1 860°C for 30 min.



Thus, good results were obtained in the first fabrication campaign, but further progress seemed possible.

Second fabrication campaign

The objective of the second campaign was to improve on the previous results with Al₂O₃, Y₂O₃, and SiO₂ sintering additives, i.e. reduce the sintering temperatures even more by varying the amount of SiO₂ used. For a second step, the sintering additive SiO₂ was replaced by CeO₂, the plutonium substitute, with concentrations from 0-30 wt.%. Once again, hardness and toughness measurements were made, but are not reported here.

For the first step, Al₂O₃ and Y₂O₃ were added in the required proportion to produce 10 vol.% YAG, and 0-8 wt.% SiO₂ was used. Figure 5 shows the densities achieved. At 1 730°C, the densities achieved were all low (85-87% TD). At higher temperatures, 1 770°C and above, the addition of SiO₂ made a significant effect. With no SiO₂, the density achieved at 1 770°C was only about 91%, whereas with 2.6-8 wt.% SiO₂ densities of ≥94% TD were achieved. The density achieved was independent of the amount of the SiO₂ addition in the range tested – 2.6-8.9 wt.%. Above 1 820°C, higher sintering temperatures were ineffective at further increasing densities.

In the second step, when CeO₂ was added as a plutonium substitute, and SiO₂ was not included as a sintering agent, the surprising result was that higher densities were achieved at lower temperatures than had been obtained with SiO₂. Figure 6 shows the results. At 1 670°C, densities of 97-99% were

Figure 5. Variation of density with sintering temperature of SiC fabricated with 5.8 wt.% Al₂O₃, 7.7 wt.% Y₂O₃ and 0, 2.6, 5.0 and 8.0 wt.% SiO₂ (SAY-6, -6A, -6B, and -6C, respectively)

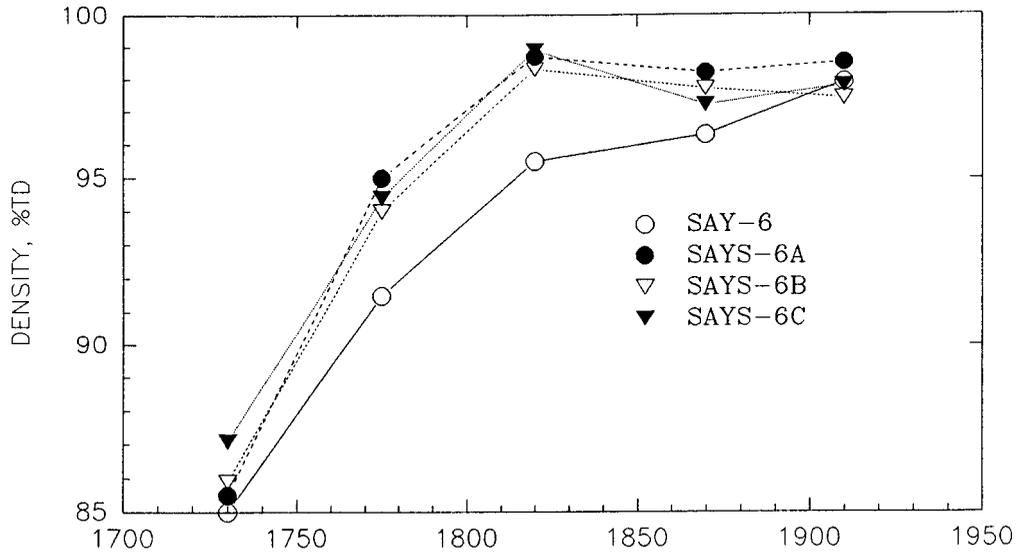
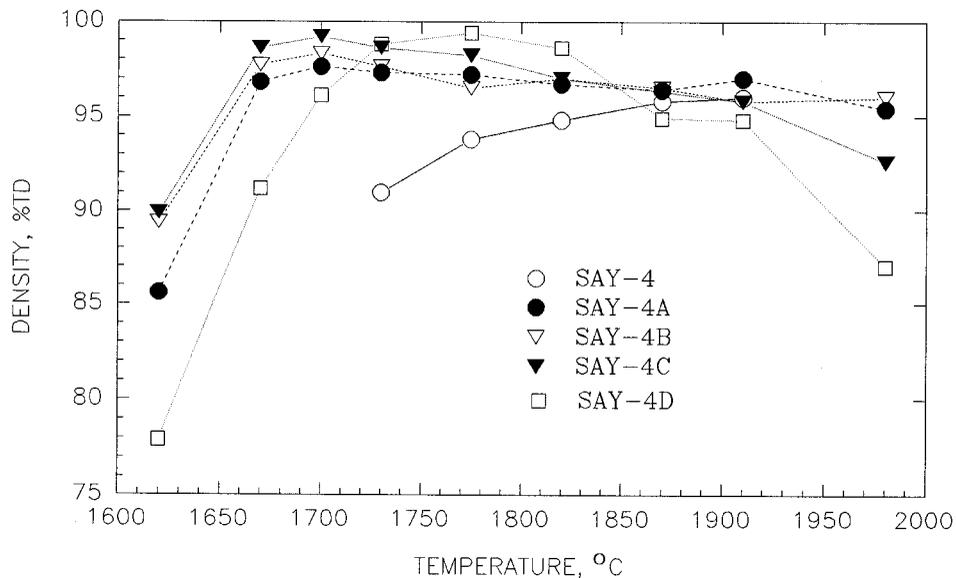


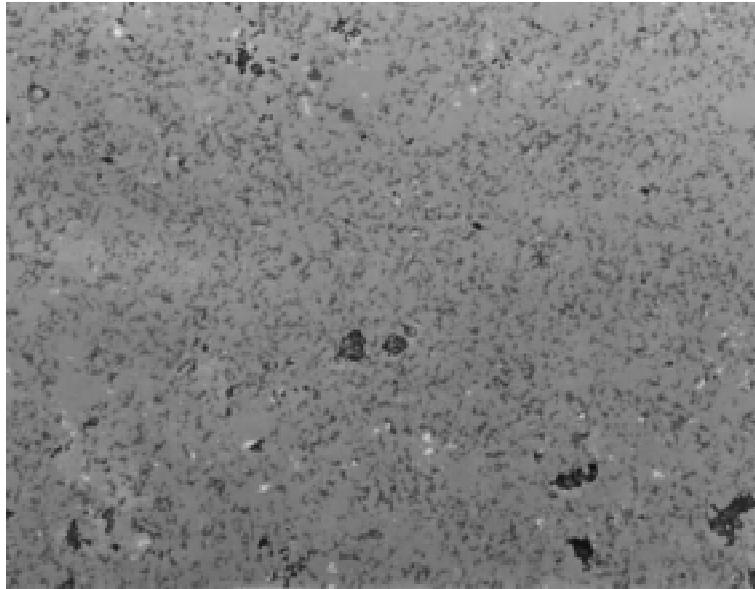
Figure 6. Variation of density with sintering temperature of SiC fabricated with Al₂O₃, Y₂O₃ and 0, 5.0, 10.0, 20.0, and 30.0 wt.% CeO₂ (SAY-4, -4A, -4B, -4C, -4D, respectively)



achieved for all ceria concentrations used, except the highest – 30 wt.%. Even for this highest concentration, 96% of TD was reached at 1700°C and 98.5% at 1740°C. X-ray diffraction showed that Al₂O₃, Y₂O₃ and CeO₂ reacted during sintering to produce two clearly identifiable compounds, both of which had been seen in the first campaign: 3Y₂O₃·5Al₂O₃ garnet (YAG) and cerium aluminate (CeAlO₃). The structure of these two phases was dependent on the composition and the sintering temperature. For example, between 1620°C and 1870°C the garnet phase XRD peaks were not present for the sample with 30 wt.% ceria and were significantly reduced in intensity for the sample

with 20 wt.% ceria. Surprisingly, above 1 870°C XRD did not show the presence of either of the two phases, garnet or cerium aluminate. The reason for this phenomenon is not understood. Optical microscopy (e.g. Figure 7) showed that the phase identified as garnet plus cerium aluminate (dark grey) was finely distributed between micrometer-sized β -SiC grains. At the 20 and 30 wt.% concentrations of ceria, the dark grey phase was larger in size (6-8 μm).

Figure 7. Optical micrograph of a polished section of the sample (SAY-4B) with 10 wt.% CeO_2 . Phase identification is the same as for Figure 2.



Third fabrication campaign

The main objective of the third campaign was to find an acceptable fabrication route that did not require the isostatic pressing stage that had previously been used in all fabrication tests. Isostatic pressing is not used in commercial UO_2 fabrication methods for power reactors, and the objective was to make SiC more acceptable to fuel fabricators. In addition, somewhat lower amounts of sintering aids, Al_2O_3 and Y_2O_3 , were used.

The starting materials were 7 wt.% Y_2O_3 , 7-8 wt.% Al_2O_3 , 5-10 wt.% CeO_2 , 1 wt.% oleic acid (lubricant) and 3-11 wt.% polyethylene glycol (binder). Sintering temperatures ranged from 1 710°C to 1 890°C. Cold uniaxial pressing at pressures of 50, 70, 100, 150 and 200 MPa were used. In addition, for comparison, tests were made with an isostatic pressure of 300 MPa.

Sintered densities of 95-97% of TD for the samples with 5 wt.% CeO_2 were achieved at all pressures (and for the isostatically pressed samples). However, the pressure used affected the formation of cracks in the green compacts, which generally became visible to the naked eye after sintering. No cracks formed at 50 MPa (or for the isostatically pressed samples), at all concentrations of binder. At 70 MPa pressing pressure, only samples made with 5% binder formed with no cracks (four samples made at each composition and pressing pressure). Results were generally the same for the 10 wt.% ceria samples.

Specific heat and thermal conductivity

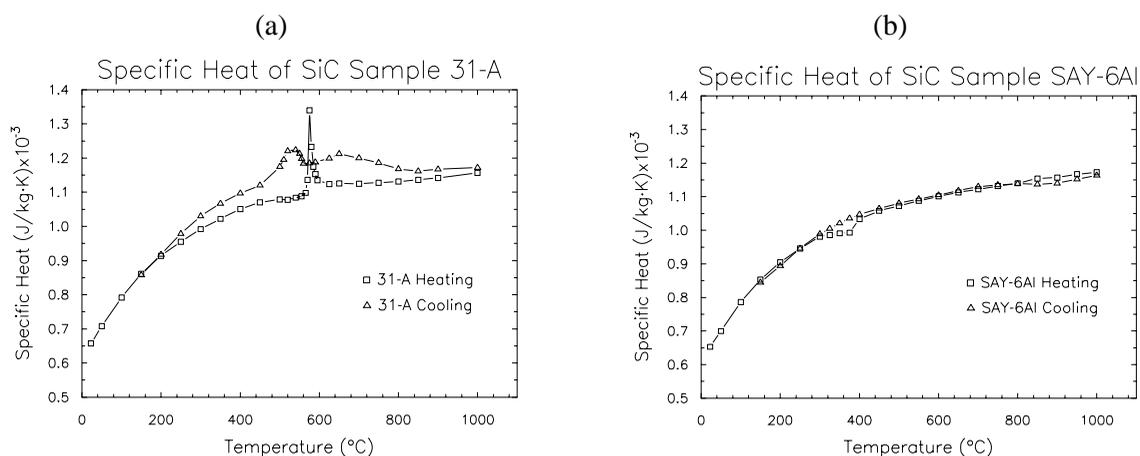
The CRC Handbook of Chemistry and Physics [3] lists the thermal conductivity of synthetic single crystal silicon carbide at 100°F (38°C), 200°F (93°C) and 300°F (149°C) as 90, 86 and 85 W/m·K respectively. The same reference gives the specific heat as 0.67 J/g·K for α -SiC (cubic) and β -SiC (hexagonal).

Measurements of specific heat and thermal diffusivity were made on two of the samples fabricated at Queen's University:

- One from the first fabrication campaign (31-A) with 8.0 wt.% Al_2O_3 , 5.0 wt.% CeO_2 and 2 wt.% C (plus lubricant and binder) starting powders and sintered at about 2 000°C to about 94% TD.
- One from the third fabrication campaign (6-AI) with 6.9 wt.% Al_2O_3 , 5.0 wt.% CeO_2 , 7.2 wt.% Y_2O_3 and 5 wt.% binder, pressed uniaxially at 70 MPa (no isostatic pressing), sintered at 1 800-1 890°C to about 97% of TD.

Specific heat measurements were made from ambient room temperature (RT) to 1 000°C³ using both a Perkin-Elmer Model DSC-2 Differential Scanning Calorimeter with sapphire as the reference material for measurements from RT to 700°C and a Netzsch Model 404 DSC to 1 000°C. Plotted results are a smooth curve through the combined results. Measurements were made during both heating and cooling of the sample. Results are given in Figure 8. The values at 25°C are close to the CRC values given above. The apparent specific heat of sample 31-A showed a spike at about 600°C on the heating curve and an unusual shape over a wider temperature band on the cooling curve. These results may indicate that aluminium (melting temperature 660°C) has separated out, possibly on grain boundaries. These thermal features are therefore likely due to exothermic reactions and are not representative of the true specific heat. Because alumina combined with yttria to form a garnet phase in sample 6-AI, these anomalies were not seen for that sample. For subsequent calculations of thermal conductivity involving the specific heat, smoothed values were used.

Figure 8. Apparent specific heat capacities of two samples of SiC with alumina and ceria additives (a) and alumina, yttria and ceria additives (b)



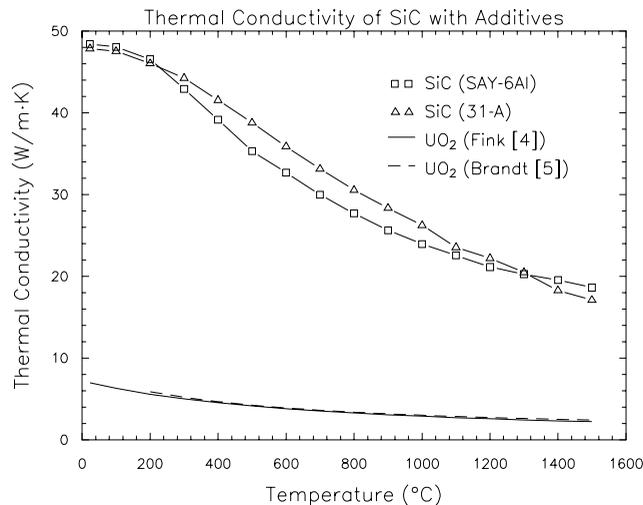
³ Thermophysical Properties Research Laboratory, Inc., 2595 Yeager Rd., West Lafayette, IN 47906, USA.

Thermal diffusivity measurements (α) were made using the laser flash diffusivity method. These values were then combined with measurements of the geometric density (ρ) of the samples and specific heat (C_p) to arrive at thermal conductivity (k), according to the formula:

$$k = \rho \cdot \alpha \cdot C_p$$

Results are given in Figure 9.

Figure 9. Thermal conductivity of two samples of SiC with various additives. The figure also shows literature values of thermal conductivity of UO₂ for comparison.



These values of thermal conductivity can be used to obtain an approximate value of the centre temperature of SiC-based matrix materials using the formula:

$$P = 4\pi \int_{T_{Sfc}}^{T_{Cr}} k dT$$

where P is the linear power of the fuel element and T_{Sfc} and T_{Cr} are the surface and centre temperatures of the fuel. The validity of this formula depends on uniform heat production in the fuel rod, and so loses accuracy when the fissile loading is such that self-shielding is significant. With this limitation, the formula provides linear power as a function of centre temperature. For example, if the surface temperature is 300°C and the linear element power is 55 kW/m, the centre temperature is only 400°C. This graphically demonstrates a key advantage of using a high thermal conductivity material – low operating fuel temperatures. For UO₂, the same conditions would produce a calculated central temperature of 1 500°C.

Compatibility of SiC with coolant and zircaloy

Compatibility measurements of SiC in deoxygenated H₂O at 320°C, pH 10.3 for 90 d [6] and at 300°C, pH 3 for 32 d [2] have been previously reported. At 320°C, pH 10.3, an 880 mg sample showed a linear (with time) weight loss of about 2% (16 mg) after 90 d (after an initial apparent weight increase of 0.5%). This weight change converts to a loss of about 20 µm from the surfaces.

For the pH 3 tests, eleven samples of SiC, 75-400 mg fabricated for the inert matrix programme were also tested at 300°C. Some samples broke into two or more pieces, and a greyish appearance formed on the surfaces, but weight changes were smaller than for the pH 10.3 tests – 1-1.5 mg, versus about 16 mg. Thus the corrosion rate from the surface is down by about an order of magnitude.

Other experimenters measured even lower corrosion rates for de-oxygenated water. Hirayama, *et al.* [7] measured corrosion of 1.5 g samples of sintered SiC (mainly α -SiC) containing boron and carbon as sintering additives. They used water solutions of pH 4, 6 and 10 at 290°C. At all three pH values, deoxygenated and oxygen-saturated solutions were tested, and in each case, the oxygenated solutions corroded the SiC faster than the deoxygenated solutions did, by factors of 10, 15 and 100 for pH values of 4, 6 and 10, respectively. The fastest corrosion rate, 3 μm over 72 h, was that for oxygen-saturated water at pH 10. For de-oxygenated water, the rate, about 0.03 μm over 72 h, was considerably slower than for our tests, but different additives may have an effect, and the level of de-oxygenation may have been greater. In any case, corrosion of SiC, in case of cladding breach, would not be a cause for concern.

Initial tests of SiC interaction with zircaloy-4 were performed by pressing polished disc specimens of SiC and zircaloy-4 under a light pressure and an argon atmosphere in a molybdenum cell at 1 000°C, 1 500°C and 1 700°C. Temperatures were maintained for 1 h at 1 000°C and 1 500°C and for 15 min. at 1 700°C. Two types of SiC specimens, fabricated at Queen's University, were used; both contained alumina as a sintering aid; and one contained titania, and 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 1 000°C, at 1 500°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 1 700°C test specimens, where the amount of molten eutectic phase was sufficient to cause partial dissolution of the Mo-cell sidewalls.

The test results indicate that formation of a molten Zr-rich phase could conceivably occur during a hypothetical reactor accident at temperatures lower than the melting point of unoxidised zircaloy cladding (1 760°C). This would allow further attack on the SiC carrier material, thereby allowing any fission products that had exsolved from the plutonium phase to be released. On the other hand, UO_2 interaction with zircaloy begins at about 1 200°C [8] and dissolves into molten zircaloy at 1 760°C. The rate and extent of UO_2 -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_2 interaction. Tests using SiC pellets clad with zircaloy-4 sheathing, rather than pressed discs in a Mo cell, would provide a better comparison.

Accelerator simulations of in-reactor fission-fragment damage

The Tandem Accelerator at the Chalk River Laboratories was used to produce a beam of 72 MeV (typical fission-fragment energy) of iodine. This beam was used to bombard the surfaces of candidate inert matrix materials over a wide dose-range (10^{14} - 10^{17} ions/cm²)⁴ and a wide temperature range

⁴ 10^{17} ions/cm² is approximately equivalent to a fuel burn-up of 12 MWd/kg heavy element.

(RT to 1 200°C). Samples tested were ZrSiO₄ (zircon), MgAl₂O₄ (spinel), CeO₂ (ceria), CePO₄, ZrO₂ doped with Ca, Ce, Er or Y, as well as SiC. 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.

Results, with figures showing surface relief of many of the samples, were reported in Ref. [6]. Usually samples showed either no surface relief or relief of 1-2 µm (ion-bombardment depth was 10-20 µm, depending on material). A benchmark test on Al₂O₃, which is well-known to vitrify and swell in-reactor when used as a fuel-containing matrix, showed surface relief of about 1.6 µm. Similarly, tests on UO₂ did not show swelling (no observable surface relief). Of all the materials tested, the least amount of swelling was observed for SiC and doped ZrO₂. These candidate materials did not show *any* swelling; i.e. laser profilometry could not detect any surface relief at the implantation spots. All other candidate materials showed some surface relief, implying that swelling could be expected when used in-reactor as a fuel. However, in contrast with the accelerator simulation of spinel, Porta [9] reported that spinel performed well in an in-reactor irradiation in France. Possibly, in some cases, cladding helps to confine the test material and thereby limit swelling. Thus, if a material has tested poorly in an accelerator simulation test, it may still perform adequately in-reactor; if a material performs well, however, in an accelerator simulation test, it seems highly probable that it would not swell in-reactor.

Conclusions

Progress has been made in establishing methods of fabricating SiC containing a wide range of cerium, which was used as a plutonium substitute. The methods are similar to those in current use for UO₂ fuel production. High densities were achieved and the required sintering temperatures have been reduced from over 2 000°C to approximately 1 700°C by judicious use of sintering additives. In fact, ceria itself has proven to be a most effective sintering additive. High density material was fabricated without using cold isostatic pressing, a step not used in UO₂ fuel production. Cerium is usually incorporated into the SiC as an AlCeO₃ phase, often as micron-sized particles, but as larger particles (6-8 µm) under some fabrication conditions. Above 1 830°C sintering temperature, with Al₂O₃ and Y₂O₃ sintering aids, XRD did not show the evolution of any phases except SiC. This suggests a total solid solution of all additives, and is not understood.

Measurements of specific heat capacity and thermal conductivity of SiC specimens fabricated in this programme were made; although the thermal conductivity is reduced from pure, fully dense SiC, the conductivity is still so high that the central temperature in an operating fuel at 55 kW/m would only be about 100°C higher than the coolant temperature.

Silicon carbide containing cerium (plutonium substitute) and a few weight per cent sintering additives was fabricated using equipment that is compatible with conventional UO₂ or MOX fabrication technology. The thermal conductivity of such SiC-based inert matrix fuels is very high – a clear benefit from the perspective of in-reactor fuel performance and safety considerations. Accelerator simulations of in-reactor damage indicate that such a fuel would be dimensionally stable. Conventional pressurised water coolants and zircaloy claddings are also compatible with SiC.

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PLUTONIUM ROCK-LIKE FUEL BEHAVIOUR UNDER RIA CONDITIONS

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Abstract

The safety characteristics of rock-like (ROX) fuel systems are studied for the dispositional use of excess plutonium (Pu) in light water reactors (LWRs). The transient behaviour of ROX fuel composed of a two-phase mixture of stabilised ZrO_2 and spinel ($MgAl_2O_4$) is investigated by transient analysis of reactivity initiated accidents (RIAs) with the EUREKA-2 code. Pulse irradiation tests of the simulated ROX (Pu was replaced by U for easier realisation) under RIA conditions was conducted in the Nuclear Safety Research Reactor (NSRR).

The LWR core with U-free ROX is evaluated to have reactor kinetic parameters less moderate than those of an ordinary UO_2 core, which results in considerably higher peak fuel enthalpies in design base RIAs. The ROX fuel, however, has lower density (1/2 of UO_2), larger specific heat (2/1 of UO_2) and lower melting temperature (1 940°C). Because of these properties, the ROX fuel and cladding temperature tended to be lower than that of UO_2 fuel in the three tests at peak fuel enthalpies of 240, 390 and 530 cal/g-fuel conducted in the NSRR. The fuel rod did not fail at 390 cal/g-fuel, even though partial fuel melting (15%) was observed. The lower melting temperature of the ROX as compared to UO_2 (2 840°C) kept the cladding temperature lower than its melting point (1 825°C) and prevented thermal failure of the cladding, thus defining the failure threshold of UO_2 fuel (212 cal/g-fuel). Cladding rupture and considerable fuel dispersion occurred at the peak fuel enthalpy of 530 cal/g fuel (the fuel melt fraction is 55%). However, mechanical energy generation (prevention of which was the main purpose of the current Japanese safety guideline limit for RIA) due to molten-fuel/water interaction was not observed. These pulse irradiation tests indicate that ROX fuel behaviour under RIA conditions is quite different from that of UO_2 , and that the safety limits could be higher for the ROX fuel.

Introduction

As a choice for the use of plutonium (Pu) from fuel recycling and from nuclear weapons, inert matrix fuels or uranium-free (U-free) fuels are under development in many countries [1-2]. At the Japan Atomic Research Institute (JAERI), research on inert matrix fuel started in 1994 [3]. The research aims at development of a chemically and geologically stable rock-like oxide (ROX) structure fuel for once-through use in light water reactors (LWRs) and for high transmutation of Pu, which has high proliferation resistivity and environmental safety. For the ROX system, quasi three-component systems of $ZrO_2(Y,Gd)-Al_2O_3-MgO$ (Zr-ROX) and $ThO_2-Al_2O_3-MgO$ (Th-ROX) have been studied.

Among the research, fabrication techniques, material properties, chemical stability and irradiation stability are studied [4-8], in addition to nuclear characterisation of the ROX core [9]. Due to the absence of U, the ROX core tends to have less moderate reactor kinetic parameters which result in higher peak fuel enthalpies in reactivity initiated accidents (RIAs) than for ordinary UO_2 cores. The ROX fuel, however, has material properties quite different from those of UO_2 . Because of these properties, the ROX fuel behaviour could be quite different from that of UO_2 fuel and the current safety limit would not be applicable to ROX. Thus, fuel behaviour under simulated RIA conditions above the current safety limit is studied in the Nuclear Safety Research Reactor (NSRR) pulse irradiation tests. This paper summarises the characteristics of the ROX core and fuel behaviour in reactivity initiated accidents (RIAs).

Nuclear characteristics of ROX core

Characteristics of two types of ROX, Zr-ROX and Th-ROX with weapons-Pu, in an LWR core arrangement are evaluated by cell burn-up calculations and 2-D core calculations using the SRAC [10] code system and JENDL-3 [11] nuclear library. In an LWR of moderator to fuel volume ratio (V_m/V_f) = 1.9, which corresponds to current PWRs, Pu transmutation rates with the two types of ROX are large enough and more than 80% and 99% of Pu and ^{239}Pu , respectively, can be burned. The calculated kinetic parameters indicate less moderate characteristics of ROX cores, especially with Zr-ROX. The fertile ^{232}Th in Th-ROX works like ^{238}U in the UO_2 fuel, making kinetic parameters more moderate and reactivity drop due to burn-up smaller than that of Zr-ROX. The neutron capture of ^{232}Th to generate ^{233}U , however, causes the safeguards problem. Thus, the characteristics of Zr-ROX as a typical example, were investigated further in this study.

In order to obtain characteristics of a realistic ROX LWR core, a PWR of 3 000 MWt with 193 assemblies of 17×17 type ROX fuel under a three-cycle (1 400 days) operation is simulated by the COREBN and HIST codes of the SRAC system. Table 1 compares the kinetic parameters of the ROX core using weapons Pu with those of UO_2 for beginning of cycle (BOC) and end of cycle (EOC) conditions. Due to the absence of fertile nuclides like ^{238}U , the Zr-ROX core has kinetic parameters less moderate than those of an ordinary UO_2 core. With these kinetic parameters, considerably high peak fuel enthalpies (E_p) are evaluated with the point kinetics code EUREKA-2 [12] for design base RIAs, as shown in Table 2. The highest enthalpy is calculated in the one rod ejection RIA simulation, 1% $\Delta k/k$ reactivity insertion, from EOC hot stand-by condition. In order to keep the peak fuel enthalpies below the current Japanese guideline limit of 230 cal/g, five times larger Doppler reactivity is needed. The addition of thoria (ThO_2) by 24 mol% or UO_2 by 15 mol% to the ROX fuel with weapons Pu, and a heterogeneous core with 1/3 ROX and 2/3 UO_2 , are found to mitigate the high fuel

Table 1. Kinetic parameters of PWR core with Zr-ROX fuel evaluated by 2-D SRAC-COREBN calculation

		Zr-ROX [PuO ₂ -ZO ₂ (Y,Gd)-Al ₂ O ₃]		UO ₂	
		BOC	EOC	BOC	EOC
Prompt neutron life time l (s)		1.20×10^{-5}	2.31×10^{-5}	2.09×10^{-5}	2.45×10^{-5}
β_{eff}		2.87×10^{-3}	3.48×10^{-3}	6.14×10^{-3}	5.45×10^{-3}
Void reactivity (% $\Delta k/k$)	0-40% void	-0.32	-9.7	-8.0	
	0-95% void	-17.7	-93.0	-96.0	
Doppler reactivity (% $\Delta k/k$)	600-900 K	-0.13	-0.23	-0.86	
	900-1 200 K	-0.098	-0.20	-0.75	

Table 2. Results of RIA analyses at peak power position of hot stand-by EOC core

Fuel type	Parameter adjusted		Peak fuel enthalpy (Ep), cal/g (kcal/cm ³)	Peak fuel temperature, °C
Zr-ROX	None		>>230 (1.28)	>>1 940
	Doppler reactivity	×3	330 (1.84)	1 940
		×5	220 (1.23)	1 700
	β_{eff}	×1.5	740 (4.13)	
		×2	570 (3.18)	
UO ₂	Guideline limit		230 (2.52)	2 600
	None		93 (1.02)	1 820

enthalpies to a reasonable extent with larger Doppler reactivity. The guideline limit, however, is defined for UO₂ and a different limit should be used for ROX, the material properties of which are quite different from those of UO₂.

NSRR pulse irradiation tests

Test conditions

In order to understand the basic behaviour of the ROX fuel rod under the RIA conditions beyond the guideline limit for UO₂, three pulse irradiation tests were conducted in the NSRR at peak fuel enthalpies (Ep) of 240, 390, and 530 cal/g. Short fuel segments with ZrO₂[Y] (yttria stabilised zirconia: YSZ)-MgAl₂O₄ (spinel)-UO₂ fuel pellets of 17×17 PWR type are subjected to pulse irradiation under stagnant water cooling conditions at room temperature and at atmospheric pressure. The schematic of the test rod is illustrated in Figure 1 and specifications are listed in Table 3. The rod has a 135 mm long fuel stack with 14 pellets surrounded by Zry-4 cladding. Spinel, which is inert from the point of view of nuclear characteristics, is added to obtain larger thermal conductivity and U is used as a Pu simulant in the tests.

The test rods were instrumented with thermocouples(T/Cs) for measurement of temperatures of the cladding surface and the capsule water, sensors of rod internal and capsule pressure, pellet stack/cladding elongation sensors, cladding strain gages and a water level sensor to trace the transient behaviour during pulse irradiation tests. The instrumentation and the capsule configuration are illustrated

Figure 1. Test fuel rod configuration

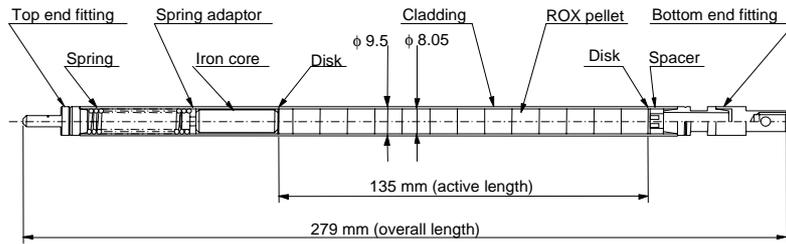


Table 3. Specifications of Zr-ROX test fuel rod for NSRR pulse irradiation test

Element	Overall length	279 mm
	Fuel stack length	135 mm
	Fill gas	He 0.1 MPa
Pellet	Material composition	UO ₂ :YSZ:MgAl ₂ O ₄ /25.4:26.8:47.8 mol% YSZ = ZrO ₂ :Y ₂ O ₃ /0.812:0.188
	²³⁵ U enrichment	20 w/o
	Diameter/length	8.05 mm/9 mm (P/C radial gap = 0.085 mm)
Cladding	Material	Zry-4
	Outer/inner diameter	9.5 mm/8.22 mm (thickness = 0.64 mm)

in Figure 2. The test rod contained in the capsule was pulse irradiated in the experimental cavity of the NSRR, as shown in Figure 3. The NSRR is a TRIGA-ACPR, which utilises U-ZrH fuel for safe pulsing operations with enhanced negative fuel temperature reactivity. The ROX fuel rod power histories in the three tests are illustrated in Figure 4. Peak linear heat rate reached the range of 12×10^4 through 3×10^4 kW/m, and the pulse width at half maximum was 4.5 through 8.4 ms in the tests.

Figure 2. Test capsule arrangement and instrumentation for NSRR pulse irradiation tests

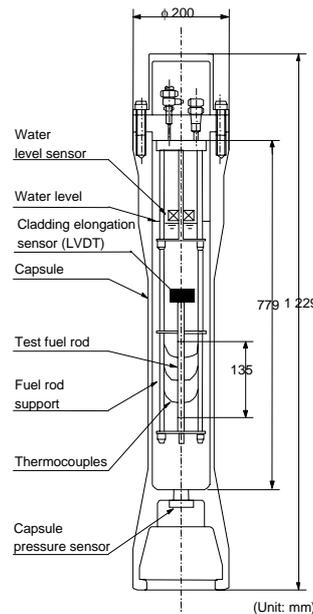


Figure 3. Configuration of the NSRR

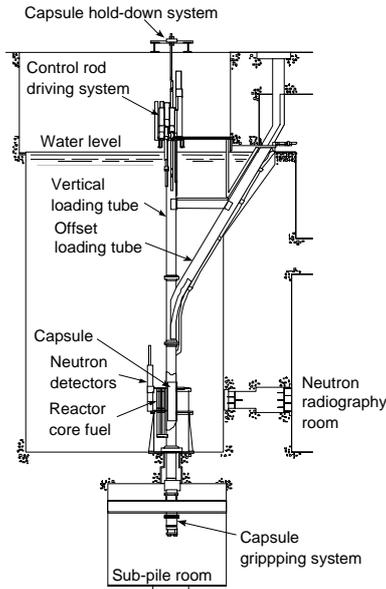
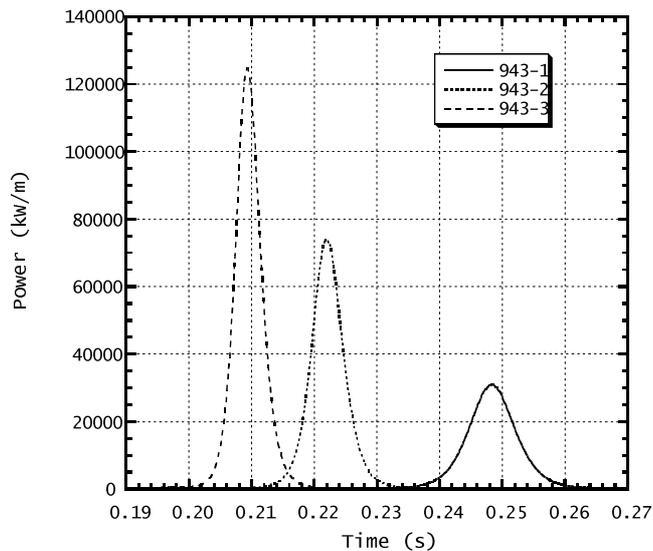


Figure 4. Power histories of test fuel rods



Test simulation with FRAP-T6

The ROX fuel rod behaviour during the pulse irradiation tests were simulated with the FRAP-T6 code [13], which was developed for the thermal and mechanical analyses of the transient behaviour of U(Pu)O₂ fuel rods, and was modified/validated for the NSRR tests [14,15]. The material properties of ROX, e.g. larger specific heat (2/1 of UO₂), were installed in FRAP-T6 for the ROX test simulation. The test simulation was conducted to estimate fuel temperature, etc. which was unable to be measured, and to examine if the models for UO₂ fuel rod applicable for the ROX.

Results

The post pulse irradiation appearance of the three test rod is shown in Figure 5. In Test 943-1 at an Ep of 240 cal/g, local oxidation of the cladding occurred. The cladding surface temperature exceeded 100°C for only about 0.8 s, reaching locally different peaks of 350-600°C. The peak fuel temperature was estimated to be 1 600°C by the FRAP-T6 simulation. The cladding strain gage indicated quite limited elastic strain below 0.05% at the timing of pulse irradiation, suggesting limited pellet/cladding contact in the test. These results suggest that the threshold enthalpy of departure from nucleate boiling (DNB) for ROX fuel under the tested condition is around 240 cal/g. Results of the test are summarised in Table 4.

Figure 5. Appearance of ROX fuel rod after the pulse irradiation



Table 4. Summary of the ROX pulse irradiation test results (peak values are listed)

Test no. (\$)	943-1 (\$2.7)	943-2 (\$3.75)	943-3 (\$4.5)
Ep, cal/g	240 (1.0 kJ/g)	390 (1.6 kJ/g)	530 (2.2 kJ/g)
Cladding t. °C [cal]	600 [750]	1 200 [1 200]	1 600 [1 400]
Fuel t. °C [cal]	[1 600]	[1 940/melt fraction 0.15]	[1 940/melt fraction 0.55]
Pellet elongation, %	2.1 [1.6]	3.1 [2.1]	>3 [>2.1]
Cladding elongation, %	-[0.4]	0.6 [0.9]	Not measured [1.0]
Radial plastic strain, %	0.2	1.5	4
Rod pressure, MPa	–	0.12	>0.3

In the second test, Test 943-2, at an Ep of 390 cal/g, the cladding surface temperature reached 1 200°C and film boiling lasted for about 8 s, as shown in Figure 6. Pellet stack and cladding peak elongation were 3.1% and 0.6%, respectively, which suggested pellet cladding mechanical interaction (PCMI) during the test to some extent. The post-test fuel, as shown in Figure 7, indicated slight bulging of about 1% in residual hoop strain. Fuel temperature was calculated to reach the eutectic point of 1 940°C, and about 15% of the fuel was estimated to be once molten. The cross-sectional micrographs of Test 943-2 fuel in Figure 8 show a central void due to the fuel melting. On the other hand, those of Test 943-1 fuel at an Ep of 240 cal/g show no morphological change except for the cracks. The rod internal pressure increased by 0.02 MPa from the initial pressure of 0.1 MPa by the pulse irradiation. The cladding survived the transient and limited chemical interaction between the fuel was found. The morphological change and the chemical interaction will be studied further by SEM/EPMA.

Figure 6. Transient cladding temperature histories

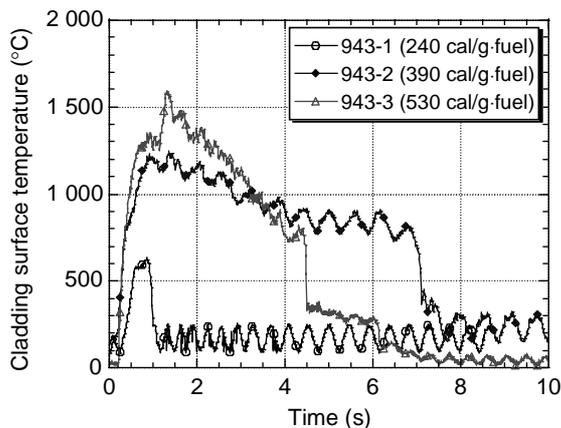


Figure 7. Residual hoop strain of the rods

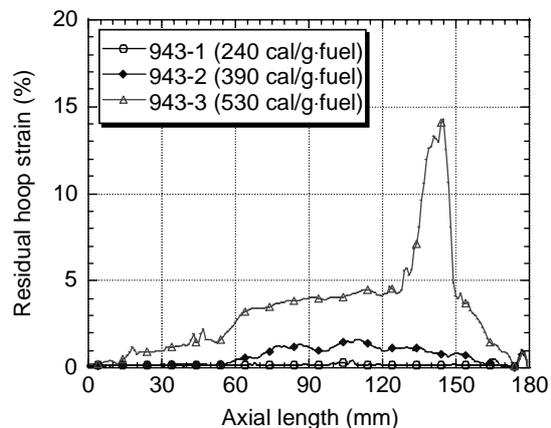
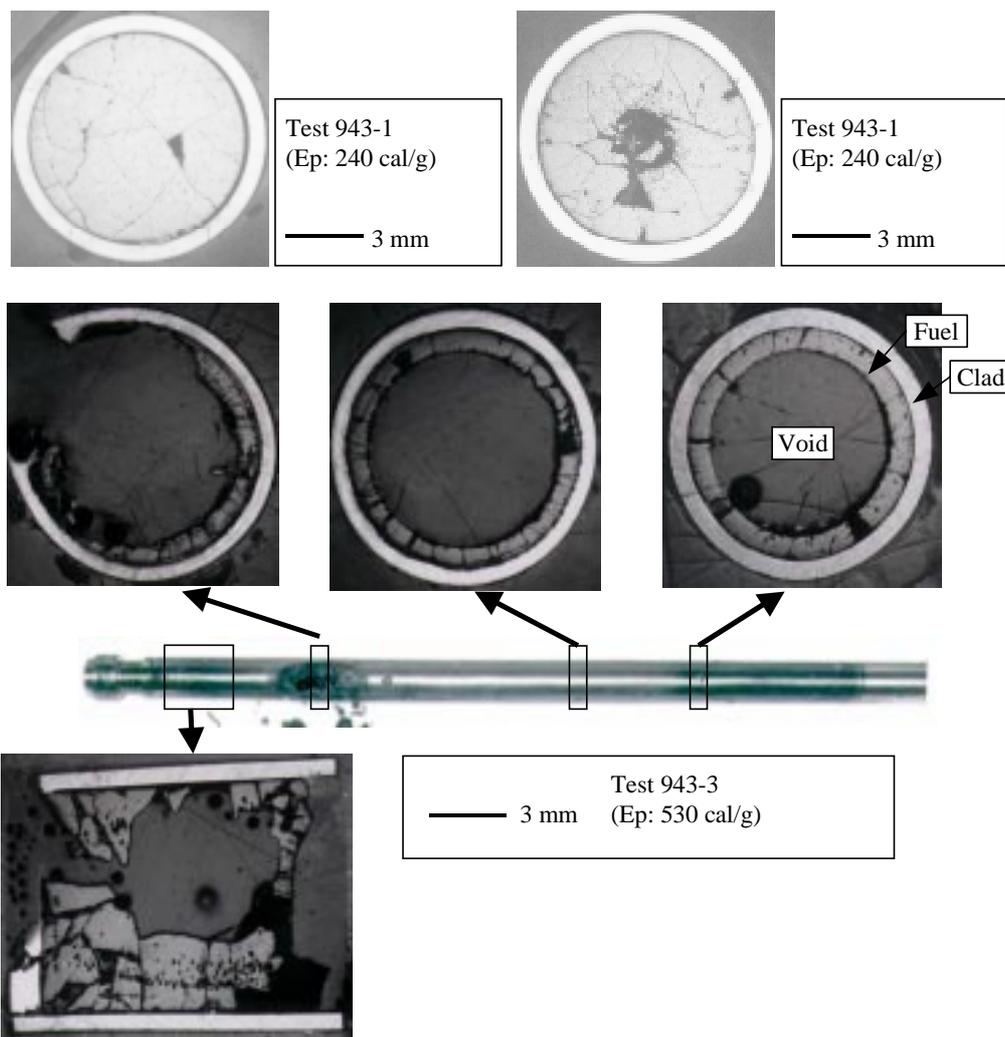


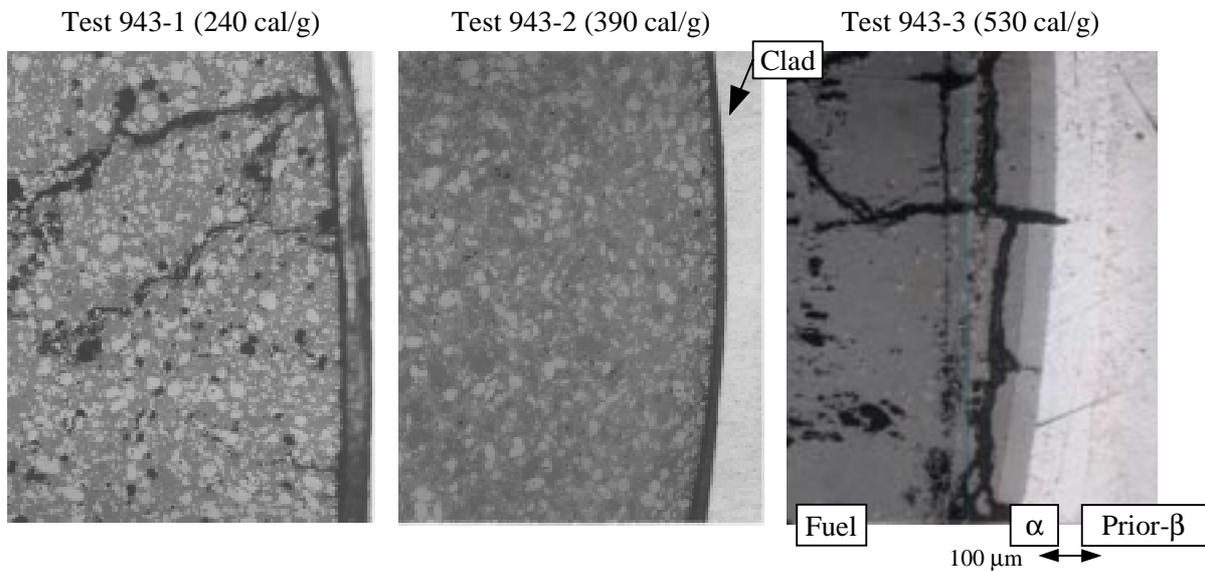
Figure 8. Cross-sections of pulse irradiated ROX fuel rods.
 The central cavity is generated by fuel melting in Test 943-2. Most of the fuel looks once molten and about 70% of fuel was ejected in Test 943-3.



Cladding failure occurred in Test 943-3 at an Ep of 530 cal/g. The cladding ruptured at the lower part of the fuel stack, as shown in Figure 5, and a considerable amount of fuel (about 70%) was dispersed to the water. However, the pressure spike was not measured and the water column movement sensor indicated minor vibrations, suggesting that mechanical energy generation due to molten fuel/water interaction was negligible in the test. The FRAP-T6 calculated 55% of the fuel melted in the test. The cladding reached a peak temperature of about 1 500°C, and showed neither extended embrittlement due to heavy oxidation nor thinning/thickening of once molten cladding, which causes cladding failure of UO₂ fuel. The cladding residual hoop strain was about 4% over the pellet stack region, which suggested that the rupture occurred relatively late during the transient after the cladding temperature rose. The larger cladding temperature decreasing rate, about twice that of Test 943-2 in Figure 6, suggests a smaller heat capacity for the fuel in Test 943-3 after the fuel release, probably at about 1 s.

Figure 8 compares the cross-sections of pulse irradiated ROX fuel rods in the three tests. In Test 943-1 at an Ep of 240 cal/g, no micro-structural change except for cracking of the pellet is seen in Figures 8 and 9. Fuel melting created a central void in the pellet in Test 943-2. A micro-structural change in the zirconia and urania is observed to become finer blocks invading into spinel even at the fuel periphery in the test. Most of the fuel in Test 943-3 looks once molten and considerable interaction with the cladding occurred, which likely contributed to the cladding burst. Further analyses are being performed with SEM/EPMA to understand the material interactions and morphology changes better.

Figure 9. Micro-structures of pulse irradiated fuel rods



Discussion

The unique behaviour of Zr-ROX fuel was observed in the three pulse irradiation tests. Due to the specific heat of ROX – twice as large as that of UO₂ – its fuel and cladding temperature tend to be much lower than those of UO₂. As summarised in Table 5, the threshold enthalpy of DNB is about 240 cal/g, while that of UO₂ is 110 cal/g. Cladding failure of UO₂ occurs at an Ep of 220 cal/g, when the cladding temperature reaches its melting point, and thinning/thickening of once molten cladding

Table 5. Comparison of fresh ROX and UO₂ fuel behaviour under RIA conditions

Peak fuel enthalpy cal/g (J/g)	Fresh UO ₂ fuel rod		ROX fuel rod		Transient behaviour for fuel rod
	Cladding temp.	Pellet temp.	Cladding temp.	Pellet temp.	
110-120 (461-502)	100-700	1 200			UO ₂ DNB threshold
220 (921)	1 850	2 500			UO ₂ failure threshold (UO ₂ cladding embrittlement)
240 (1 005)			600	~1 500	ROX DNB threshold (ROX cladding oxidation)
280 (1 172)	>1 850	2 840			UO ₂ pellet partial melting
325 (1 361)	>1 850	2 840			UO ₂ mechanical energy generation
390 (1 633)			1 200	~1 940	ROX pellet partial melting (pellet melt fraction ~15%)
530 (2 219)			1 500	~1 940	ROX failure without mechanical energy generation (pellet melt fraction ~55%)

and oxidation progresses heavily. The fuel temperature at the failure threshold of UO₂ is estimated to be 2 500°C. On the other hand, ROX fuel did not fail even at an Ep of 390 cal/g, where fuel partial melting (~15%) was observed. The considerably lower melting temperature of ROX (1 940°C) made the difference, keeping the cladding temperature lower than its melting point. The lower fuel temperature, however, makes negative reactivity feedback even smaller in the case of RIAs, when the Doppler reactivity of ROX is smaller than UO₂ due to the absence of a fertile nuclide like ²³⁸U.

The ROX fuel failure occurred at an Ep of 530 cal/g and a considerable amount of fuel ejection was observed. The failure, however, did not cause measurable mechanical energy generation, which occurred at an Ep of 390 cal/g or higher for UO₂ fuel due to molten fuel and coolant interaction. The lower melting temperature of ROX and the relatively late dispersion might have contributed to the mild failure even with fuel melting. The safety evaluation guideline for UO₂ fuel was defined to avoid mechanical energy generation in the design bases accidents. The test results suggest higher criteria for fresh ROX than UO₂. The failure might be different for irradiated ROX due to fission gases accumulated in the fuel. The gas pressure released by fuel melting could cause wilder fuel dispersion, resulting in mechanical energy generation for irradiated fuel.

The FRAP-T6 simulation of the tests shows reasonable agreement with the test results, suggesting that the models for UO₂ are applicable to the ROX. The FRAP-T6 models, however, were not designed for liquid fuel, and many estimated properties of molten ROX, such as latent heat for melting, thermal conductivity, etc., were used in the simulation. These models and properties need to be modified and/or validated for a better understanding of ROX behaviour, because molten fuel plays an important role in accidents.

Conclusions

Transient characteristics of the Zr-ROX fuel under RIA conditions were examined by cell and LWR core calculations. Kinetic parameters, such as Doppler and void reactivity, of the Zr-ROX core were less moderate and higher fuel enthalpies than for the UO₂ core were calculated for design base RIAs. The enthalpy limit for Zr-ROX from a safety point of view, however, is not known. Thus, three pulse irradiation tests of Zr-ROX, ZrO₂(Y)-MgAl₂O₄-UO₂, were performed to investigate transient fuel behaviour under RIA conditions. Fuel failure did not occur at an Ep of 390 cal/g, even though about 15% of the fuel was once melted in the test. Cladding rupture and molten fuel dispersion occurred at an Ep of 530 cal/g. The failure, however, was not associated with mechanical energy generation, which should be avoided from the safety point of view. The molten fuel played an important role on the Zr-ROX fuel failure and its mechanism was quite different from that of UO₂. Further research is needed for establishing the criteria. However, these results suggested that the enthalpy limit in the safety evaluation guideline for ROX could be higher than that of UO₂, which was 230 cal/g in a Japanese guideline.

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IRRADIATION DAMAGE IN INERT MATRIX OF URANIUM-FREE FUELS

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Abstract

Several cercers are good candidates for plutonium fuels without uranium or for the targets related to the heterogeneous recycling of minor actinides. These cercers are actinide compounds (for instance PuO_2 or AmO_2) dispersed in inert matrices such as MgO or MgAl_2O_4 . Experimental irradiations of MgO-AmO_2 are expected in the near future in the nuclear reactor Phénix. The behaviour of these new fuels under irradiation, however, is not very well known, and even if experimental irradiations of inert matrices are also in progress at Phénix, it is very useful to study the mechanisms of irradiation damage by simulation. The dominant effects of fission products on inert matrices can be in particular simulated by irradiation with ions.

Different specimens of MgO or MgAl_2O_4 have been irradiated with heavy ions at room temperature at GANIL. The irradiation damages have been characterised by macroscopic and microscopic observation, by optical spectroscopy and by XRD; the influence of fabrication process has also been analysed.

The results will be presented with the equivalent domain in the real fuels. A mechanism for irradiation damage will be proposed and discussed.

REACTORS WITH NON-OXIDE FUELS

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METALLIC FUEL FOR FAST REACTORS

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Abstract

Metallic fuel has certain advantages for use in fast reactors. High thermal conductivity of the fuel leads to passively safe response to anticipated-transient-without-scrum initiators in a sodium cooled pool-type fast reactor. Also, the high thermal conductivity allows the core design option of high linear pin power. The most recent design changes of metallic fuel have yielded burn-up capability in excess of 20 at%.

Transient tests on metallic fuel have eliminated an early concern that in extreme accident conditions metallic fuel would melt and form a critical mass at the bottom of the reactor vessel. Rather the fuel expands axially, prior to cladding failure, with consequent reduction of reactivity. Upon cladding failure the fuel is dispersed into a high porosity mass that is easily coolable. Iodine is the largest contribution to the fission product source term for early release of radioactivity in an accident. The zirconium-containing fuel alloy is unique because it has been found to retain the iodine.

Metallic fuel is easily and economically reprocessed and refabricated remotely, which leads to a diversion resistant and proliferation resistant fuel cycle. A commixed plutonium, uranium and minor actinide product can be separated from the fission products by electrorefining and refabricated by injection casting for reirradiation. The separation of fission products is incomplete such that the product remains highly radioactive. Further, plutonium and higher actinides remain commixed and contaminated with fission products precluding suitability for potential weapons use.

This paper will review the most recent performance information on U-Pu-Zr metallic fuel irradiated to burn-ups of 20 at%. Phenomena such as fuel swelling, mechanical and chemical interactions with various cladding materials, performance during transients, failure mechanisms and behaviour following both benign and complete failure of fuel pins will be presented.

Ultimately it will be shown that the fuel is capable of very high burn-up – exceeding 20 at%. The achievable burn-up depends upon a combination of the pressure exerted on the cladding by

fission gas accumulation in the plenum of the fuel pin and, once the built-in void volume is expended, a fuel volume increase of 1-2% per per cent burn-up will occur from the solid fission product accumulation.

Off-normal performance is best characterised as benign where significant reactivity excursions lead promptly to fuel expansion driven by dissolved and interspersed fission product gases, without fuel-pin failure. For high temperature excursions fuel cladding interaction along with increasing plenum pressure lead to cladding failure. Should a fuel pin fail during normal operation the pin depressurises with no further degradation. Run-beyond-cladding breach irradiations of up to 200 days have shown no further degradation of the breach site.

Introduction

Metallic fuels were the first fast reactor fuels. Ceramic fuels replaced metallic fuels due to early performance limitations of metallic fuels. During the 1970s the performance limitations of metallic fuel for power reactor applications were satisfactorily resolved at EBR-II; in fact, additional attributes of metallic fuel were discovered. The results of the development work, both preceding and throughout the 1970s, have been summarised [1].

During the 1980s metallic fuel became the fuel of choice for the Integral Fast Reactor (IFR) concept proposed by Argonne National Laboratory (ANL). Central to the concept is recognition that the world's reserve of ^{238}U must be utilised as an energy source in the centuries to come. Thus, the fuel system must be able to utilise plutonium as its principal fuel and must have the potential to simultaneously create replacement plutonium by irradiating ^{238}U . U-Pu-Zr was chosen as the most promising fuel alloy based on earlier work in the 1960s.

Metallic fuel has several important attributes. Recycling by electrorefining leads to a simple, proliferation resistant fuel cycle. As well, the fuel cycle allows the actinides to be recycled for useful energy. The actinides are thus transformed to fission products, which avoids their contribution to risk of the geologic repository. Also, the high thermal conductivity of the metallic fuel results in significant safety benefits.

Irradiation performance

Plutonium or uranium-plutonium alloys are not candidates for fast reactor fuel because the melting points are too low. Additions of chromium, molybdenum, titanium and zirconium all resulted in an adequate increase in melting temperature over a satisfactory range of plutonium content in the alloy. However, zirconium was unique in that it enhanced compatibility between the fuel and austenitic stainless-steel cladding materials by suppressing the interdiffusion of fuel and cladding components [2]. Without zirconium, the cladding elements nickel and iron readily diffuse into the fuel to form compositions that result in a lower solidus temperature (the temperature at which melting starts) adjacent to the cladding. Should the solidus temperature at the fuel/cladding interface be exceeded during an off-normal event, the cladding could fail due to penetration by the liquid front. The concentration of zirconium in the U-Pu-Zr alloys was limited to about 10 wt.% for plutonium concentrations of up to 20 wt.%, because too much zirconium would result in a liquidus temperature (the temperature at which melting is complete) that would exceed the softening point of the fused-quartz molds in the injection-casting fabrication equipment used for metallic fuel [3]. By the end of the 1960s, a plutonium-based fuel alloy had been partially developed that had both adequate compatibility with the cladding and a high solidus temperature. Raising the melting temperature solved only a part of the difficulty – there remained the need to achieve high burn-up and long residence time in the reactor. Eventually that was accomplished by a simple change in design, based on theoretical developments in the early 1960s; the initial smeared density¹ was reduced.

¹ “Smeared density”, a dimensionless percentage, is commonly used to quantify the effective density of the fuel within the cladding. As used here, it denotes a planar smeared density calculated by dividing the mass of fuel (including any non-gaseous fission products) in a unit length of pin by the theoretical mass of a unit length of voidless fuel of the same composition, with diameter equal to the inside diameter of the cladding. Low smeared density can be achieved by using highly porous fuel or a large gap between fuel and cladding. Note that the smeared density increases as burn-up proceeds because each heavy-metal atom that fissions becomes two atoms of a less dense material.

The first fuel in EBR-II was called MK-IA. The MK-IA fuel was clad with austenitic stainless steel with an 85% smeared density and had a very small gas plenum. Because of the low burn-up achievable with the MK-IA fuel, the decision was made by the US Atomic Energy Commission (AEC) that mixed-oxide fuel would be developed as the nation's fast reactor fuel. However, continuing to use metal fuel for EBR-II was convenient, which would allow EBR-II to continue as a test reactor for mixed-oxide fuel and advanced cladding material. It was economically desirable to convert the core of EBR-II from the high smeared density MK-IA fuel to a low smeared density fuel that could go to higher burn-up [4]. As a result of that decision, a MK-II fuel design emerged in 1970 for use in EBR-II. The MK-II was U-5 wt.% fissium (Fs)² at a smeared density of 75%, first clad with 304 stainless steel and later with 316 stainless steel, with a plenum-to-fuel volume ratio of about 0.6. By 1974 it was clear that the new design was successful. Cladding breach did not occur until about 10 at% burn-up, more than a factor of three better than the MK-IA fuel.

MK-II fuel failed by an intergranular crack at the restrainer dimples – three small, sharp-bottomed indentations that were placed 120° apart about 20 cm above the fuel column [5]. Their purpose was to prevent the metallic fuel pin from somehow ratcheting upward inside the cladding, then dropping back down at an inopportune time and creating a reactivity insertion. However, post-irradiation examination of a large number of MK-II fuel pins showed only a slight upward motion in a small number of them; thus, future designs eliminated any type of restraining device. Later, fuel irradiated without the restrainer dimples achieved substantially greater burn-up.

More than 30 000 MK-II fuel pins have been irradiated in EBR-II as standard driver fuel [6]. The administrative burn-up limits for the fuel remained at 8 at%, even though consistently no cladding breach occurred below 10 at%. The burn-up limit of 8 at% was chosen for two reasons. First, the “hex” (hexagonal) ducts on the fuel assemblies were initially made of type 304 stainless steel. At a fuel burn-up of 8 at% (about 8×10^{22} n/cm² total), the diameter of the hex ducts had increased from radiation-induced swelling until they could not be handled through the in-vessel EBR-II storage basket. Second, 8 at% was far enough below the ultimate burn-up capability of 10.5 at% to assure that the probability of in-reactor breach was very low during steady-state operation, and to provide a wide margin for containing all anticipated effects of off-normal events. With these safeguards in place throughout the 1970s, EBR-II continued to demonstrate that metallic fuel is capable of high burn-up.

As of 1983 the commercial viability of U-Pu-Zr fuel remained undemonstrated, even though many of the feasibility questions associated with the performance of metallic fuel had already been answered. In fact, additional positive attributes of metallic fuel had been discovered, such as robust performance during off-normal transients. Nevertheless, from 1969 to 1984 no U-Pu-Zr fuel was irradiated and there was no facility available to fabricate the fuel. With only 18 U-Pu-Zr fuel pins irradiated to about 4 at% burn-up, the data base was weak, although these fuel pins did exhibit the performance that would have allowed them to reach high burn-up. Moreover, in addition to the lack of demonstration that the U-Pu-Zr fuel would, in fact, reach high burn-up, a number of other issues required further study for complete resolution.

The capability to fabricate ternary fuel was established, and a fuel programme to demonstrate performance was initiated to gain the information that would be needed to eventually obtain a license for metallic fuel from the Nuclear Regulatory Commission (NRC). A number of assemblies were

² Fissium is an alloy that approximates the equilibrium mixture of metallic fission product elements left by the pyrometallurgical recycling cycle designed for the EBR-II; it consists of 2.5 wt.% molybdenum, 1.9 wt.% ruthenium, 0.3 wt.% rhodium, 0.2 wt.% palladium, 0.1 wt.% zirconium and 0.01 wt.% niobium.

irradiated to establish what the burn-up potential of the U-Pu-Zr fuel could be, and how the fuel pins would perform with alternative cladding materials and with a range of design parameters such as smeared density, plenum-to-fuel volume ratio, operating temperature and linear power. Finally there was a series of tests to help develop the fuel fabrication specifications. In parallel with the irradiation experimentation, analytical modelling and out-of-core testing were undertaken to improve understanding of fuel performance.

For the initial set of three tests of U-Pu-Zr fuel, the irradiation began in EBR-II in early 1985, and a burn-up of 18.4 at% was reached [7]. The test contained fuel of three compositions: U-10Zr, U-8Pu-10Zr, and U-19Pu-10Zr (compositions given in weight per cent). The fuel was clad with an austenitic stainless steel alloy, D9. Later in 1985, a test with fuel identical to the first three, but with cladding of the martensitic alloy HT9, began its irradiation in EBR-II. Fuel elements clad with HT9 reached 19.9 at% burn-up without cladding breach. It was found that although the microstructure of the alloys was strongly dependent on their composition, the quantity of gas released to the plenum as a function of burn-up was consistent for all fuel alloys irradiated, as was the burn-up at which the pores became interconnected. Further, it was observed that the initial swelling of the fuel, up to the point of fuel/cladding contact, was anisotropic, with the radial component more than twice as large as the axial component [8]. Still, the fuel slug had appreciable axial growth as a function of alloy composition and irradiation conditions. As expected from irradiation results in the 1960s, radial redistribution of the alloying elements was observed, particularly uranium and zirconium, although the radial concentration profile of plutonium was largely unchanged [9]. As the radial concentration of zirconium and uranium changed, a radial distribution of porosity developed, with distinct zones that were evident on a macro scale. Up to the burn-ups examined, the changes in the diameter of the austenitic cladding could be attributed primarily to irradiation-induced swelling and creep, the source of stress being the plenum pressure in the fuel pin.

As mentioned earlier, the high smeared density of early metallic fuel prevented it from reaching high burn-up. Even at low burn-up, the cladding deformed and failed when the fuel swelled from accumulation of fission products. Attempts at that time to extend the burn-up concentrated on alloying, on thermomechanical treatment of the fuel to suppress swelling and on the use of strong cladding to resist deformation and the onset of swelling. That work was largely unsuccessful, peak burn-ups of about 3 at% being the best achievable.

The primary cause of swelling is the accumulation of fission-product gas in bubbles that grow as gas pressure increases with burn-up and overcomes the gas-bubble surface tension, causing the fuel matrix to expand. It was known theoretically that when fuel swelling reaches about 30% the bubbles must begin to interconnect, independent of size and number density. Therefore, it was postulated that if the gap between fuel and cladding was large enough to allow the fuel to swell about 30% before fuel/cladding contact, the bubbles would interconnect and release the accumulated fission gas, thus removing or reducing the primary cause of swelling; a large gas plenum above the fuel would capture the fission gas and keep the stress on the cladding reasonably low. By the time the metallic fuel development programme was terminated in the late 1960s, it had been demonstrated that interconnection of pores and subsequent release of fission gas occurred consistently when the smeared density was less than 75% for a range of metallic fuel alloys. Figure 1 shows the fission gas release as a function of fuel-volume increase [10].

U-Pu-Zr has the most extensive data, although substantially older data for U-Fs and limited information on other alloys are available. The general swelling behaviour for these alloys is shown as the increase of fuel length versus burn-up in Figure 2. Swelling proceeds rather rapidly with burn-up, a characteristic of metal fuel.

Figure 1. Fission-gas release versus fuel-volume increase

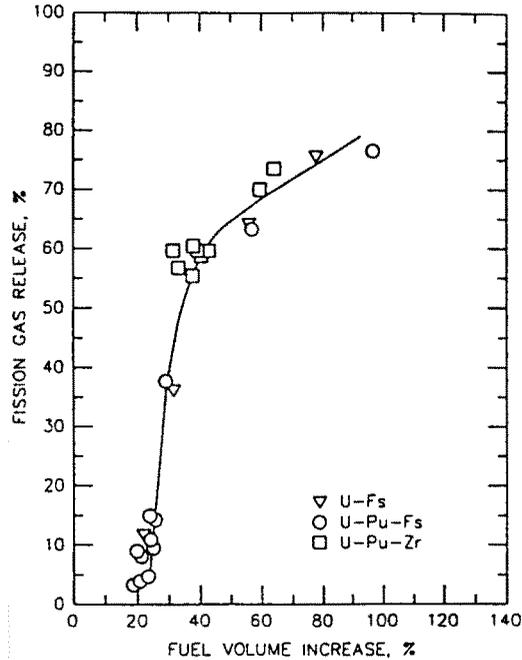
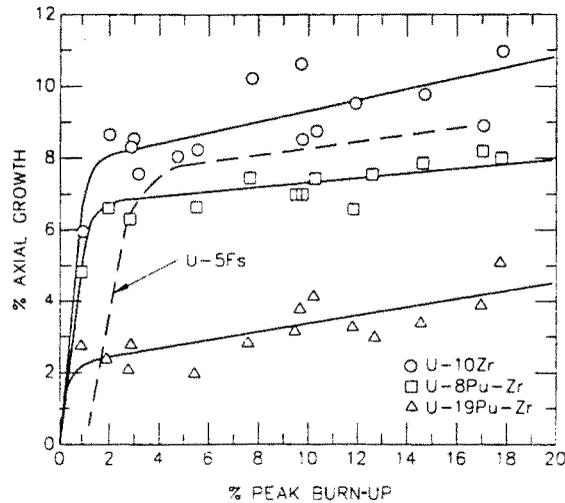


Figure 2. Fuel length increases in various metallic fuels as a function of burn-up (EBR-II irradiation; all the fuel had similar initial smeared density)

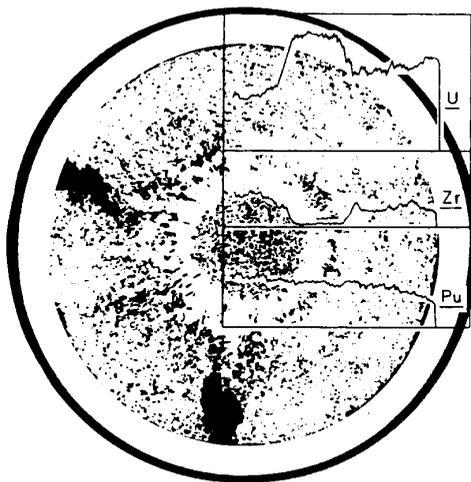


In order to improve the transfer of heat from fuel to cladding, this gap is filled with a liquid metal such as sodium. The plenum above the fuel should be large enough to accommodate the sodium when expelled from the gap by the swelling fuel, while leaving sufficient volume to accommodate released fission gas. The bond sodium also plays an important role in the thermal behaviour of fuel during irradiation by assuring that the thermal conductivity remains reasonably high throughout the burn-up cycle. An important concern is that the large amount of swelling and porosity developed during the first few atom per cent of burn-up may lead to large reductions in the thermal conductivity of the fuel. During this free-swelling stage, bond sodium is displaced from the gap to the gas plenum above the fuel. Eventually, near the point of maximum swelling, the pore interconnections become

large enough to allow the bond sodium to infiltrate the body of the fuel. The ingress of high-conductivity bond sodium into the fuel porosity effectively restores much of the thermal conductivity that the fuel lost in the initial free-swelling stage.

Virtually all length increase takes place during the burn-up interval before the swelling fuel contacts the cladding, which occurs at ~1% burn-up. For a given type of fuel, the levelling off in axial swelling is thus determined by the fuel's initial smeared density. The initial smeared density is typically chosen at approximately 75%, to allow enough swelling (approximately 30%) to release fission gas from the fuel (see Figure 1). This much planar swelling would, for isotropic swelling, translate to a length increase of approximately 15%. However, the observed length increases are consistently smaller, indicating anisotropic swelling. The main reason for this appears to be the difference in swelling behaviour between the hotter centre of the fuel and the colder periphery, illustrated in Figure 3 for U-10Zr [8].

Figure 3. Transverse metallographic section from the high-temperature section of a U-19 Pu-10Zr element of 3 at% burn-up, with superimposed microprobe scans, showing zone formation, cracking and Zr-U redistribution



A possible explanation for the anisotropic swelling behaviour is the following: stress-induced swelling in the peripheral fuel zone (predominantly α phase) would result in a larger diametral than axial strain; hence anisotropic swelling [11]. Anisotropy is especially pronounced and variable in U-Pu-Zr, as can be seen in Figure 2. Note that the diameter of the fuel expanded some 15% to reach the cladding while the axial growth was only 2-8%, as shown in the figure.

The higher-plutonium pins show more anisotropy because of the development of a pronounced radial distribution of alloy phases that is a result of redistribution of zirconium in the radial temperature gradient. This is consistent with the equilibrium phase diagram of U-Pu-Zr, which shows that various phase-field boundaries can be found radially across an operating fuel pin. The solubility of zirconium in these phases varies with temperature; thus, a driving force for diffusion can be created both by the gradient in chemical potential of the fuel constituents in these phases, and by the heat flow. A low-zirconium zone consisting primarily of the low Zr ternary ζ -phase is observed to form at the centre of pins by zirconium migration to the lower temperature δ -phase zirconium at the periphery. In parts of fuel pins that operate at higher temperatures, the zirconium-depleted zone forms at mid-radius by migration of zirconium to both the high temperature γ -phase zirconium at the centre and the δ -phase zirconium at the periphery.

The boundaries of the radial zones follow isotherms in the fuel, which determine the various phase boundaries of the alloy. In the usual situation of upward coolant flow and a cosine-shaped axial power profile in the fuel, the peak fuel temperature occurs between the centre and top of the fuel column. An example of the resulting zone pattern in a moderate power U-19Pu-10Zr fuel pin is shown in Figure 3. In higher power pins where the peak fuel temperature shifts to a higher axial position, the three-zone pattern, with a zirconium-depleted zone at an intermediate radial location, may extend to the very top of the fuel column.

As fission gas is generated, the gas pressure increases in the bubbles within the fuel. This pressure is transmitted to the cladding by the plastic metallic fuel. Because early pin designs did not allow the fuel sufficient room for free swelling, this gas-bubble pressure was transmitted directly to the cladding. As a result, all early designs suffered from cladding deformation and rupture at modest burn-ups. However, as discussed earlier, an as-built smeared density of about 75% allows free fuel swelling of approximately 33%, at which point porosity becomes largely interconnected and open to the outside of the fuel, releasing a large fraction of the fission gas to a suitably large plenum at the top of the pin. The gas pressure in the open pores is then determined by the volume and temperature of the plenum above the fuel.

However, the open porosity will eventually close due to the accumulation of solid fission products. The following discussion contains an approximate calculation of the net volume change due to accumulation of non-gaseous products. The volume changes and total yield of the major non-volatile, non-soluble fission products are shown in Table 1.

Table 1. Volume changes due to non-soluble major fission products

Element group	Fission yield per 100 fissions	State	Average molar volume $\text{cm}^3 \text{mole}^{-1}$	Per cent volume change per % BU*
Alkali (Cs, Rb)	22.2	Liquid, 70% in Na bond	70	0.108
Alkaline earth (Sr, Ba)	14.7	Solid, liquid, in precipitation and 20% in Na	20	0.146
Rare earths + Pd (Ce, Nd, etc.)	51.4	Solid, precipitation	20	0.792
(Tc, Ru, Rh, Ag)	23.3	Solid, precipitation	9	0.162
Total non-soluble fission products				1.18

* For a molar volume of $12.9 \text{ cm}^3 \text{mole}^{-1}$

A rigorous analysis of the change in molar volume of the fuel due to transmutation of uranium and plutonium into solid fission products requires evaluation of the fission products' chemical state in the various metallurgical phases – a difficult task considering the complexity of the alloys. However, an approximation for the major fission products can be made if one partitions these products and makes the following assumptions:

- 1) Zirconium, niobium and molybdenum are all in solution in the fuel phases, and thus contribute some to the swelling by reducing the density of the fuel.
- 2) Alkali elements are partly dissolved in the bond sodium.
- 3) The noble metals precipitate as compounds.

- 4) The rare earths precipitate as a separate compound or alloy.
- 5) The majority of the alkaline earths precipitate separately.

Post-irradiation examinations of high burn-up fuel appear largely to support both this partitioning and these assumptions. Another reason for partitioning the fission products is their different migration behaviour in the fuel; such grouping allows evaluation of the effect this migration has on the properties of the various radial phasal zones. The accommodation of the non-soluble fission products predictably result in expansion as a function of burn-up. However, the lattice is depleted in uranium and plutonium as fissioning proceeds, resulting in a volume decrease that partially offsets the expansion due to fission products.

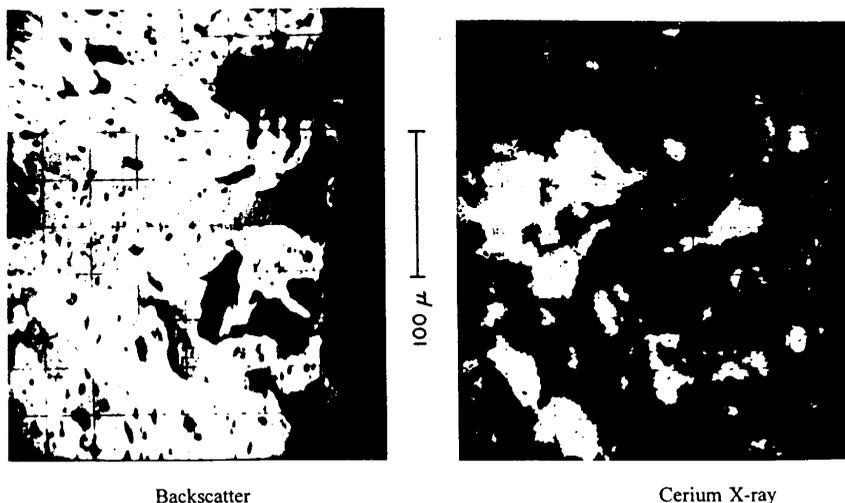
All the fission-product zirconium, niobium and molybdenum are assumed to be soluble and thus they compensate for some of the volume decrease due to uranium and plutonium fission. Therefore, non-gaseous fission products contribute to the volume change in three ways:

- Volume decrease due to the fissioning of uranium and plutonium.
- Volume increase due to the increase of zirconium, niobium, and molybdenum, which are soluble in the fuel matrix.
- Volume increase due to non-soluble fission products.

The result of the first two contributions is estimated as follows: at 10 at% burn-up of the uranium and plutonium, the average composition of the U-19Pu-10Zr (wt.%) fuel matrix has changed from 77(U-Pu)-23Zr at% to 71(U-Pu)29(Zr-Nb-Mo) at%, with an associated volume change of -0.2% per per cent burn-up.

The rare earths, which are insoluble, are found to collect in two separate phases. They collect in the large pores around the periphery of the fuel and in the gaps between fuel and cladding. Figure 4 shows this behaviour for a U-19Pu-10Zr fuel sample with 17 at% burn-up. Evidently these elements migrate down the temperature gradient. One of these two phases contains most of the palladium, but the other noble metals are concentrated in blocky precipitates throughout the fuel.

Figure 4. SEM scans of U-19Pu-10Zr fuel with 17 at% burn-up



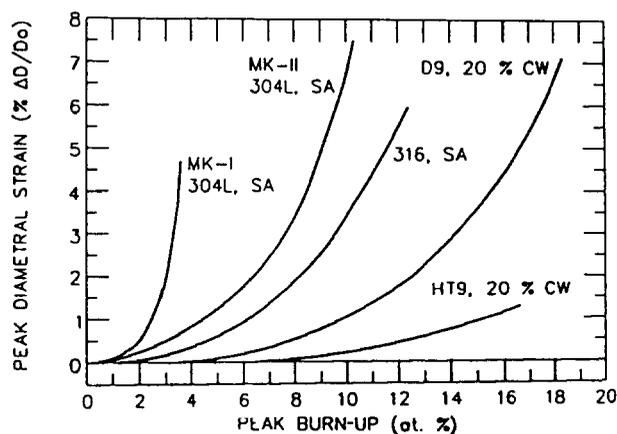
Some elements such as molybdenum, zirconium and niobium are not found as separate phases. These elements are likely in solution rather than separate phases. Chemical analysis of bond sodium indicate that a major fraction of the alkali and some of the alkaline-earth elements can simply be added to the volume of the bond sodium. The changes in fuel volume due to these fission products are also given in Table 1. The increase in total volume due to the non-soluble fission products amount to approximately 1.2% of the fuel volume increase per per cent burn-up.

Also the fission gases xenon and krypton that are still in the matrix must be considered. This gas is in solution and in very small agglomerations that act as a solid, liquid or high-pressure gas. Moreover, the concentration of this resident fraction of the gas is probably very different in the various fuel phases, and has not been well characterised. Post-irradiation examination, however, including wet-chemical analysis of high burn-up fuel, indicate that 10% of the gas volume for an entire pin is a reasonable estimate of the average resident fraction. Such a fraction in solution and in liquid or non-ideal dense gas increases the volume of the fuel by ~0.2% per per cent burn-up. This brings the total volume increase (not counting voids) to approximately $(-0.2 + 1.2 + 0.2) = 1.2\%$ per per cent burn-up.

With a volume increase of 1.2% per per cent burn-up, an original smeared density of 75% would change to 90% at 20 at% burn-up. It appears that in this high burn-up range, the initially open porosity will become increasingly closed off, and fission-gas pressure in the fuel will build more rapidly since less of the generated gas will vent to the plenum. In effect (remembering that the fuel has been in contact with cladding since ~1% burn-up), this would represent the onset of possibly significant fuel/cladding mechanical interaction (FCMI). In irradiation of fuel that has a smeared density of 75%, FCMI has not been observed at burn-ups of up to 18%. This is readily explained for austenitic stainless steel cladding such as type 316 and D9. As shown in Figure 5, the deformation of the cladding steel at high burn-up is so large that it accommodates any increase in the volume of the solid fuel. The deformation in type 316 and D9 cladding is caused mainly by swelling, and any irradiation creep strain can be accounted for by plenum pressure alone; hence, no evidence of FCMI.

Figure 5. Progressive improvement in the deformation (swelling and creep) of the cladding of metallic fuel elements (EBR-II irradiations)

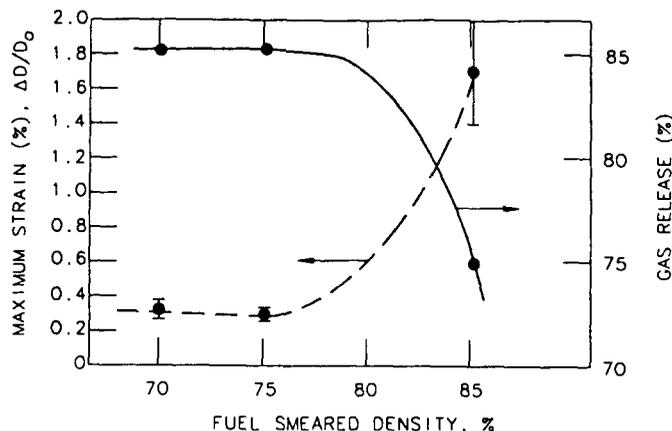
SA: solution annealed, CW: cold worked



Martensitic stainless steel, HT9, was found to exhibit negligible irradiation induced swelling at high neutron fluence. Therefore, this steel was adopted for IFR fuel cladding. As shown also in Figure 5, the deformation at 16 at% burn-up is only approximately 1%, and the smeared density of the fuel at this point should have changed from 75% to approximately 85%. Apparently little FCMI has

occurred at this burn-up, even with the non-swelling material, since the cladding creep strain can be accounted for by plenum pressure only, within the accuracy of the calculations. In order to determine the effective smeared density at which significant FCMI does occur, HT-9 clad fuel pins with an initial smeared density of 85% were irradiated along with the usual 75% [12]. The resulting cladding strains are compared in Figure 6. It is clear that the rate of growth of cladding strains increased for the initially 85% smeared-density pins, somewhere prior to 12% burn-up, when solid fission products increased the smeared density from 85% to approximately 95%. It appears from these data that FCMI became substantial in this interval, although even for initial values of 85%, open porosity apparently develops to some extent, and substantial, albeit less, gas release occurs, as is shown in Figure 6. Only when solid fission products increase the smeared density to well above 85% does FCMI become clearly evident. We may conclude that even with non-swelling cladding, significant FCMI can be avoided to high burn-up if the as-built smeared density is kept to 75%. In this discussion it has been assumed that the porosity is uniformly affected by the accumulation of fission products. Clearly, based on the observed porosity patterns and the location of fission products in the fuel, this may be too simplistic for a precise evaluation of FCMI, but it appears adequate for analysis of both steady-state and transient performance.

Figure 6. Peak cladding diameter increase and gas release fraction for HT-9-clad U-19Pu-10Zr fuel of various as-build smeared densities, at 12.5 at% burn-up



Interdiffusion of the metallic fuel elements with the cladding elements lead to the so-called fuel/cladding chemical interaction (FCCI). Characterisation of fuel/cladding interdiffusion is extremely difficult, because of the number of alloy components involved. With stainless steel cladding, even in the simplest fuel alloys such as U-Fs and U-Zr, at least five major constituents participate in the diffusion process. In addition, minor alloy components such as carbon, nitrogen and oxygen, as well as fission products – particularly at higher burn-up – appear to play an important role. The potential problem of interdiffusion of fuel and cladding components is essentially twofold: weakening of cladding and formation of relatively low-melting-point compositions in the fuel. In an attempt to assess the severity of these two deleterious effects in preparation for irradiation testing of specific fuel pins, one customarily performs diffusion-couple experiments in the laboratory, involving specific interfacing samples exposed to heat or other accelerating influences.

General observations regarding FCCI can be made even though the understanding of FCCI is not complete. Prior to accumulation of significant amounts of lanthanide fission products at the fuel/cladding interface, FCCI depends on the particular fuel/cladding combinations, e.g. the degree of nickel depletion in austenitic cladding and decarburisation of martensitic cladding has a solid-state

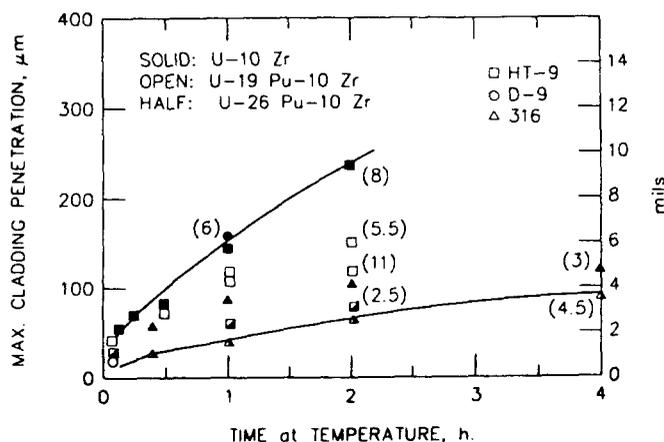
diffusion-type of dependence on time and temperature. Lanthanides ultimately control FCCI, but their presence at the internal cladding surface depends not only on burn-up but very strongly on their radial migration in the fuel. Although radial migration of lanthanides increases with fuel temperature for all fuel alloys, it is more rapid in U-Pu-Zr than in U-Zr.

The in situ formation of Zr-N layers in diffusion couples and the reduced nickel depletion found in type 316 stainless steel indicate the feasibility of improving the FCCI. However, the FCCI problem with austenitic cladding might be largely academic, since swelling alone renders these steels unacceptable for use at high burn-up. High burn-up operation requires a low-swelling cladding material such as HT9, so acceptable FCCI is more important in this steel.

The ex-reactor diffusion tests indicate that liquid-phase formations need to be taken into account if off-normal temperatures reach 700-800°C, even though melting due to FCCI is not expected during steady-state conditions. In the transients that were considered, the temperatures in this range are reached in events that last only minutes, while lower temperature events, such as loss of coolant combined with a disabled heat-rejection system, may last many hours. To better characterise the formation of liquid phases in cladding and fuel at elevated temperatures, sections of various irradiated fuel pins have been heated in an in-cell apparatus over a range of temperature and time [13]. This work has shown no evidence of liquid-phase formation below ~725°C for test durations of up to 7 h, in general agreement with the diffusion experiments discussed before, and has yielded a large body of kinetic data at higher temperature [12]. Figure 7 shows an envelope of the results of measurements on several fuel/cladding combinations, as a function of time at 800°C. The broadness of the data range is due to the number of fuel/cladding combinations tested – 0-26% plutonium and type 316, D9 and HT9 steels, as well as various burn-up levels and irradiation conditions.

Figure 7. Envelope of depth of penetration by fuel/cladding melt into the cladding as a function of time at 800°C during post-irradiation heating test

In parentheses: per cent burn-up of test samples



In general, the penetration depths appear to be parabolic functions of time raised to a power of less than one. The rate of attack is similar for HT9 and D9, but generally lower for type 316 with all fuel compositions. It is interesting to note that for the range of parameters covered thus far in these post-irradiation heating tests, fuel with 0% plutonium has shown the highest rate of attack and 26% plutonium the lowest, opposite to the trend observed in diffusion couples. Metallographic examination of these irradiated samples reveals that the FCCI is similar to that observed in unirradiated diffusion-couple tests, with the exception of a pronounced effect of rare-earth fission products

in high- burn-up fuel samples. This experimental programme has not yet led to development of a satisfactory fundamental model for the observations. Meanwhile, correlations of cladding penetration with temperature and time derived from these preliminary test results are used empirically in evaluating transient behaviour of metallic fuel pins.

Transient testing in TREAT

Safety test considerations

Beginning in 1985, safety studies of metal-fuelled LMRs were undertaken within the IFR programme, and six loop experiments in TREAT have been performed so far on irradiated metal fuel of modern design [14,15]. These have been designated as the M-series tests M2 through M7 [16]. All six were designed to provide data pertaining to transient-overpower-without-scrum (TOP-WAS) conditions in an IFR type of reactor. The specific objective was to study the behaviour of fuel and cladding near the cladding-failure threshold, for a wide range of burn-up and for several fuel/cladding combinations. Achieving this objective required, for some fuel pins, stopping the power transient at the brink of failure, and, for others, stopping the transient immediately after failure.

Analysis of the tests indicates that melting of approximately one-half of the total fuel inventory encompassing over 90% of the cross-section of the fuel slug near the top of the fuel was typical of what had been predicted for test pins in this series. Post-test examination of pins that remained intact confirmed these estimates. As described earlier, these measurements and analyses were also used to deduce the thermal conductivity of the fuel and to confirm the presence of bond sodium in restructured fuel [17].

Fission gas pressure along with penetration by the non-uranium eutectics are phenomena that have been modelled to predict cladding failure. In contrast to ceramic fuels, the lower mechanical strength of metal fuel leads to pressure in the pin plenum as the primary pressure loading of the cladding. Cladding strains, however, reflect not only the pin-plenum pressure but also thinning by eutectic formation. Cladding deformation and strain-to-failure are computed on the basis of stress/temperature history and correlations derived from transient tube burst tests. The high thermal conductivity of metal fuel assures peak cladding temperatures and, therefore, the likely failure sites will always be near the top of the fuel. Temperatures key to the failure-threshold analysis (pin plenum, peak cladding midwall, and cladding inner surface) are close to or easily derived from the measured whole-pin coolant temperature rise. The rise rate is sufficiently rapid that, except at the highest possible burn-ups, failure would not be expected until the temperature of the fuel/cladding interface exceeds a threshold value of 1350°K, the temperature at which eutectic penetration into the cladding becomes very rapid (associated with the melting of a protective, solid, iron-uranium compound). Depending on the particular test or reactor conditions, this temperature is reached at overpower levels around four times nominal.

One uranium zirconium pin survived to about 4.8 times nominal power. This result was unexpected because computed temperatures were much higher than the expected threshold of 1 350°K for rapid melt penetration. However, for rapid melt penetration, it is also necessary that a molten phase rich in uranium be in contact with the cladding. It is possible that the high solidus temperature (around 1500°K) of binary fuel prevents or delays the onset of rapid penetration.

A fast-neutron hodoscope measured the peak pre-failure elongation for each test pin. Measured values, varying from a high of nearly 20% down to 2-3%, were in every case significant beyond the approximate 1% attributable to pure thermal expansion. In most cases, the peak expansion persisted

during cooldown and was evident in post-test remains. Measured expansion of irradiated U-Fs fuel showed strong dependence on burn-up and was especially large at low burn-up. By contrast, expansion of the IFR fuel was typically less and did not show large burn-up dependence. In irradiated fuel, the underlying mechanism is believed to be expansion of fission gas that is initially confined within solid fuel, but freed to expand as fuel melts. (Expansion of the one “fresh” pin tested may have been caused by boiling of bond sodium that had mixed in with molten fuel.) Quantitatively, axial expansion is estimated by a model in which the fission gas in molten fuel simply expands until its pressure equals that of the pin plenum.

Different types of fuel behaved similarly. In all cases, cladding failure was accompanied by a sudden, temporary reversal of coolant flow at the inlet. Pressure spikes were minor (less than 2 MPa) and were correlated with the plenum pressure in the failed pin, but lower by about one order of magnitude. In each case, about half of the fuel inventory was ejected through a small breach at the top of the fuel. The amount of disruption observed seems correlated with the amount of pin depressurisation following failure. Under these circumstances, disruption could be driven either by expansion of trapped fission gas or by sudden boiling of the liquid bond sodium within the fuel.

The following conclusions can be reached from the transient tests on metallic fuel. Failure threshold with the 8-s period overpower conditions is about four times nominal power over a wide range of burn-ups and fuel types. (However, additional modelling issues and questions arise when test fuel is overheated for longer times at lower temperatures.) Pre-failure axial expansion in metal fuel is significantly greater than thermal expansion, with strong dependence on fuel type, especially at low burn-up. Expansion of fission gas trapped in melting fuel seems to be the mechanism at work. For IFR fuel, expansion in the range of 2 to 4% was typical of all burn-ups tested. Finally, on a qualitative basis, post-failure motion was benignly dispersive in all metal fuel types tested.

Operation of fuel pins with breached cladding

Several run-beyond-cladding-breach (RBCB) tests have been performed in EBR-11 with U-Fs, U-Zr, and U-Pu-Zr fuel clad with type 316, D9 and HT9 stainless steels [18]. The purpose of these tests was to confirm the expected benign behaviour of metallic fuel pins during operation following cladding breach, and to characterise the release of fission gas, delayed-neutron emitters and possibly fuel, from breached pins.

The behaviour of failed metallic fuel pins during continued operation is thus governed by the properties of the fuel, its fission products and the cladding, and is not affected by fuel-sodium reactor products. Run-beyond-cladding-breach (RBCB) tests were conducted on metallic fuel in EBR-II to understand the breach characteristics and confirm the lack of fuel reaction with the sodium coolant. The tests were run on U-Fs, U-Zr and U-Pu-Zr fuel clad with various stainless steels.

The cladding was thinned on an irradiated pin to 30-40 μm to stimulate cladding failures. Shortly after reinsertion in the reactor, cladding failure occurs in this thinned area. This is announced by both fission gas and delayed-neutron (DN) signals on the reactor monitoring system. Because the pin is depressurised, and because FCMI is insignificant (since the fuel itself does not stress the cladding and there is no fuel-sodium reaction) there is little or no cladding stress during RBCB operations, particularly with high-swelling austenitic cladding. As a result, the cladding breach remains small, even after approximately 100 to 200 days of operation of the pins in the breached mode. A similar RBCB exposure within HT9-clad pins exhibited more crack extension and widening. This may indicate a certain amount of FCMI resulting from trapped fission gas in closed-off porosity. This trapped gas

also causes the fuel to swell somewhat at the breach site where the external restraining effect of the cladding has vanished. However, it is important to note that no fuel was extruded through the cladding breach even after the 150 days of breached-pin operation.

When the fuel pin depressurises upon cladding breach, the sodium in the open porosity is expelled, which reduces the thermal conductivity of the fuel slug. The main effect of the resulting increase in fuel temperature appears to be enhanced transport of rare-earth fission products to the periphery of the fuel slug, and an increase in FCCI. The depth of rare-earth penetration into the cladding at the top of the fuel after 150 to 200 days of RBCB operation is approximately twice that in unbreached pins at comparable burn-up and cladding temperature. This is further indication that the behaviour of rare-earth fission products plays an important role in FCCI.

Summary

Metallic fuel has been continuously improved for more than thirty years. The last fuel/cladding combination studied was the ternary alloy U-Pu-Zr with the martensitic alloy cladding HT9. When the programme was terminated, this fuel had achieved 20 at% burn-up without failure. Its steady-state performance was outstanding and well understood. The fuel itself appears capable of essentially unlimited burn-up, with lifetime ultimately set by a combination of pressure exerted on the cladding by fission gases accumulating in the plenum of the fuel pin and a fuel volume that increases by 1-2% per per cent burn-up from build-up of solid fission products once they have filled available void space. Performance in off-normal transients is benign, since major excursions lead promptly to fuel expansion driven by dissolved gases, without fuel-pin failure. At excessive temperatures there can be a chemical interaction of fuel and cladding, consisting of interdiffusion of some of their components. This, along with internal pressure from the fission gases in the plenum, is the ultimate mechanism for failure of a fuel pin. If a pin with breached cladding is subjected to continued operation, no further degradation occurs.

Acknowledgements

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FUEL CYCLE SYSTEMS WITH NITRIDE FUEL FOR TRANSMUTATION

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Abstract

Core performances of the nitride fuelled fast reactor were examined with parameters of bonding materials such as He or Na, and ^{15}N enrichment. The cost of ^{15}N enrichment and the possibility of recovery of ^{14}C in the pyrochemical reprocessing were also discussed. Gaseous ^{15}N can be separated from argon purge gas of electrorefining cell to be recycled to nitridation process.

Thermodynamic predictions were made on the molten-salt electrorefining of the irradiated nitride fuel for transmutation reactor systems. Simulating calculations were made to determine the amounts of actinides recovered in the electrorefiner and to estimate decontamination factors of fission products.

Introduction

Due to its high thermal conductivity and heavy metal density, a nitride fuelled LMFBR shows good core performance such as high core breeding or low reactivity swing with burn-up, and can also offer enhanced performance in the area of passive safety. These operational and safety characteristics can provide design flexibility of reactor systems with reactors for fuel breeding and/or reactors for transmuting long-lived nuclides.

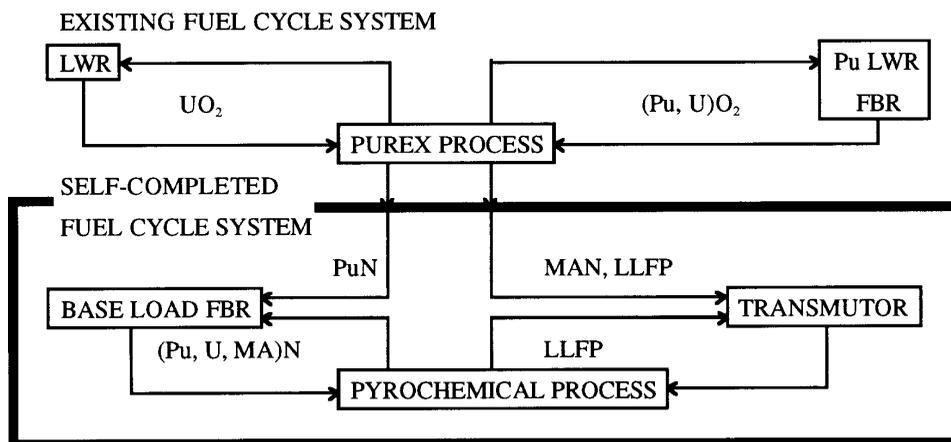
A fuel cycle system coupled with a nitride fuelled LMFBR and a pyrochemical reprocessing has been investigated at the Japan Atomic Energy Research Institute in order to establish a confinement/transmutation system for long-lived radioactive nuclides [1,2]. The pyrochemical reprocessing of nitride spent fuel is adopted for new fuel cycle systems to lower the fuel cycle cost. The main process of this fuel cycle consists of the following steps: disassembling of the nitride spent fuel, fuel element shearing, electrorefining, cathode processing, nitridation of recovered fuel metal and fabrication of nitride fuel assembly.

Core performances of the nitride fuelled fast reactor were examined with parameters of bonding materials such as He or Na and ^{15}N enrichment. The cost of ^{15}N enrichment and the possibility of recovery of ^{14}C in the pyrochemical reprocessing were also discussed. Gaseous ^{15}N can be separated from the argon purge gas of the electrorefining cell to be recycled in the nitridation process. In this paper, thermodynamic predictions were also described on the molten-salt electrorefining of the irradiated nitride fuel for transmutation reactor systems. Simulating calculations were made to determine the amounts of actinides recovered in the electrorefiner and to estimate decontamination factors of fission products.

Self-completed fuel cycle system

Figure 1 shows the schematic view of the fuel cycle system. The reactor system consists of base load reactors and transmutation reactors. The base load reactor produces electric power and extra plutonium fuel if needed.

Figure 1. Schematic view of self-completed fuel cycle system



MA : Minor Actinide

LLFP : Long-Lived Fission Product

The initial fuel inventory of reactors is assumed to be supplied from the existing LWR purex reprocessing plant. Actinides in nitric salt are converted to oxide particles using the sol-gel process, then transformed to nitride using the carbothermic reduction in nitrogen atmosphere in a particle bed. The minor actinides and long-lived fission products such as ^{99}Tc or ^{129}I are also expected to be extracted from the cadmium metal or molten salt of the electrorefiner, and to be incinerated by a specially designed reactor: the transmutator. Gaseous ^{15}N is separated from the argon purge gas of the electrorefining cell to be recycled in the nitridation process.

Nitride fuelled fast reactor

Core performances of the nitride (MN) fuelled fast reactor were examined with parameters of bonding materials (He, Na) and ^{15}N enrichment (^{14}N , ^{15}N), and were compared with that of mixed oxide (MOX) fuelled fast reactors. Two-dimensional reactor models are shown in Figure 2. Table 1 shows the main design and performance parameters for fast reactors examined. Coolant temperatures of inlet and outlet are 380°C and 550°C , respectively.

Figure 2. Reactor model

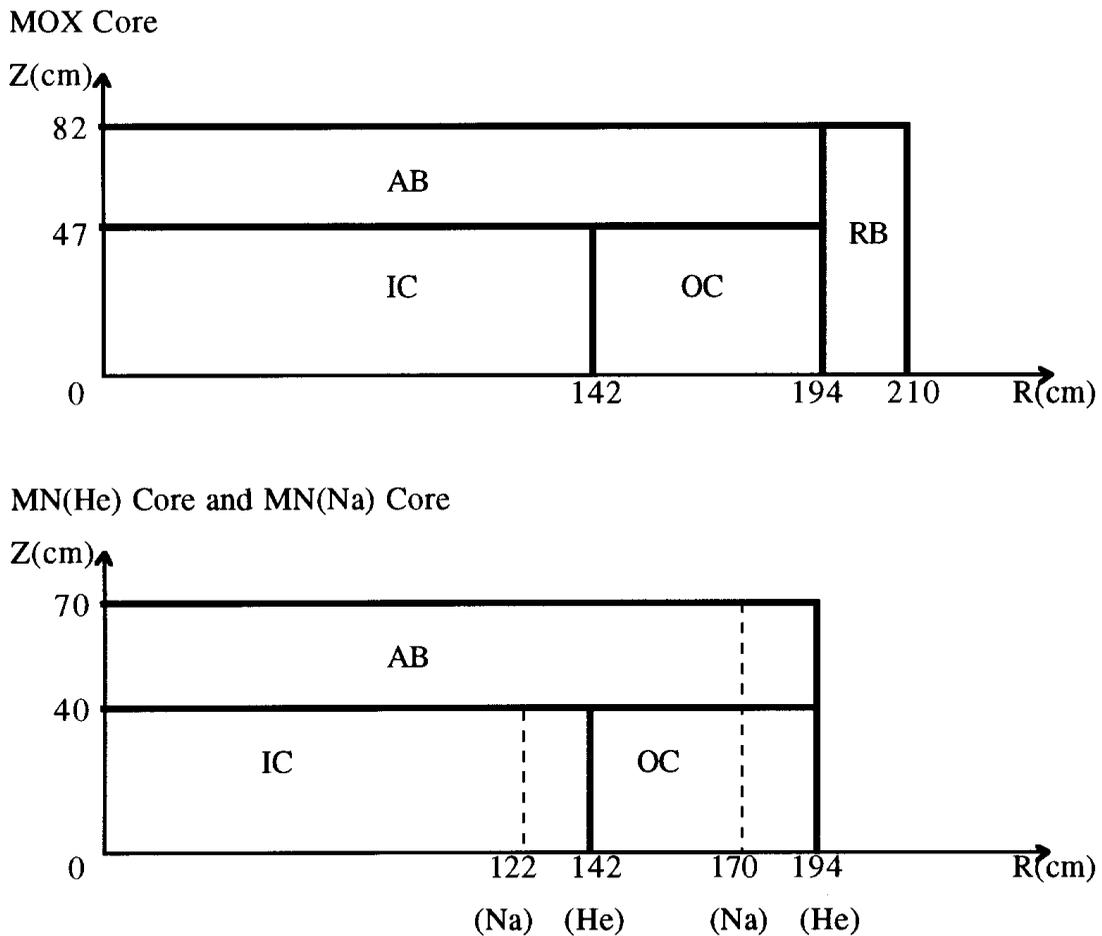


Table 1. Main design and performance parameters for 1 300 MWe fast reactor

	MOX core	MN(He) core		MN(Na) core	
Fuel pin diameter (mm)	8.5	8.5	8.5	8.5	8.5
Can thickness (mm)	0.48	0.48	0.48	0.48	0.48
Gap width (mm)	0.09	0.09	0.09	0.31	0.31
Smear density (%TD)	84	80	80	80	80
Core height (mm)	930	800	800	800	800
Average linear rating (w/cm)	280	330		440	
		N-15	N-14	N-15	N-14
Cycle length (month)	13	13	13	13	13
Number of batch	5	5	5	4	4
Average burn-up (GWd/t)	150	130	130	145	145
Puf enrichment (in/out)	17/19	11/15	14/16	12/17	15/20
Puf inventory (ton)	5.24	5.14	5.50	3.90	4.19
Puf generation (ton/cycle)	0.20	0.34	0.19	0.24	0.09
(Puf breeding)	(1.18)	(1.33)	(1.17)	(1.24)	(1.09)
Burn up reactivity loss (% $\Delta k/k/cycle$)	3.31	0.56	2.28	1.72	3.84
Doppler coefficient* ($10^{-3}T \cdot \Delta k/\Delta T$)	-4.96	-4.69	-5.46	-4.60	-5.39
Coolant temperature coefficient ($10^{-6} \Delta k/k/^\circ C$)	6.85	6.66	5.42	6.78	5.34
Radial expansion coefficient ($10^{-6} \Delta k/k/^\circ C$)	-6.55	-6.95	-6.98	-7.19	-7.35

* MOX core: 1 600°C → 2 200°C, MN core: 1 000°C → 1 600°C

The effect of the ^{15}N enrichment (100% ^{14}N or 100% ^{15}N) on nuclear performance is summarised:

- Smaller fissile inventory is required for the ^{15}N core, by 7%.
- Excellent breeding performance is obtained for the ^{15}N core, and burn-up reactivity loss decreases, less than the half of that of the ^{14}N core.
- Larger negative Doppler coefficient and smaller positive coolant temperature coefficient (sodium void reactivity worth) are obtained for the ^{14}N core.

These differences in nuclear performances come from the big difference in the (n,p) reaction cross-section between ^{14}N and ^{15}N in higher energy region. There may be troublesome problem of ^{14}C production due to the large (n,p) reaction for the ^{14}N core.

Takano, *et al.* [3] estimated the toxicity of ^{14}C for the spent fuel from the ^{14}N core assuming a recovery rate of 99.27% of U, 99.94% of Pu, 99.89% and 20% of rare earths, and pointed out that:

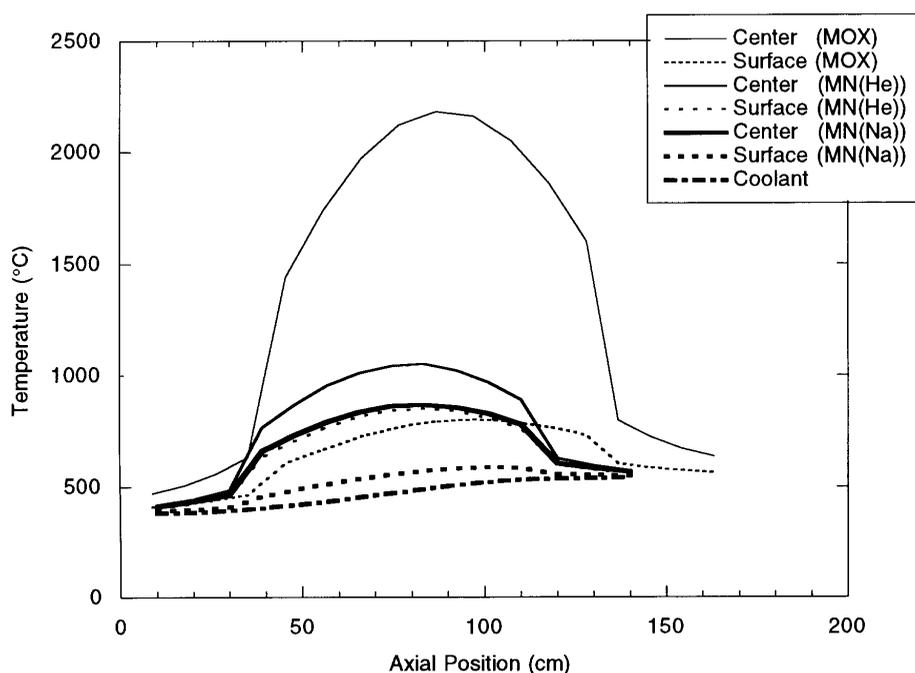
- 1) The toxicity of ^{14}C becomes almost equivalent to that of Am + Cm, and smaller by a factor than that of Pu after 500 years.
- 2) If 90% enriched ^{15}N is used, the toxicity of ^{14}C becomes similar to that of FP after 1 000 years.

From these observations, it was concluded that the problem of ^{14}C is not radiotoxicity but the capability of recovering ^{14}C in a reprocessing system. The recovery of ^{14}C is easier by pyrochemical processing in which almost all of the ^{14}C is kept in salt by chemical reactions with Zr and Mo in FP.

The cost of ^{15}N enrichment is estimated at present as very high: 100 000 yen/g. In order to keep the cost effect less than 10% of the fuel cycle cost, the enrichment cost of ^{15}N should be below 4 000 yen/g. These expensive gaseous ^{15}N can be separated from the argon purge gas of the electrorefining cell and be recycled by the nitridation process in the case of pyrochemical reprocessing.

Fuel and coolant temperature profiles are shown in Figure 3 for various cores. In the case of the nitride core (especially for the sodium bonding case), maximum fuel temperature can be significantly reduced due to its excellent thermal conductivity during normal operation.

Figure 3. Fuel and coolant temperature profile



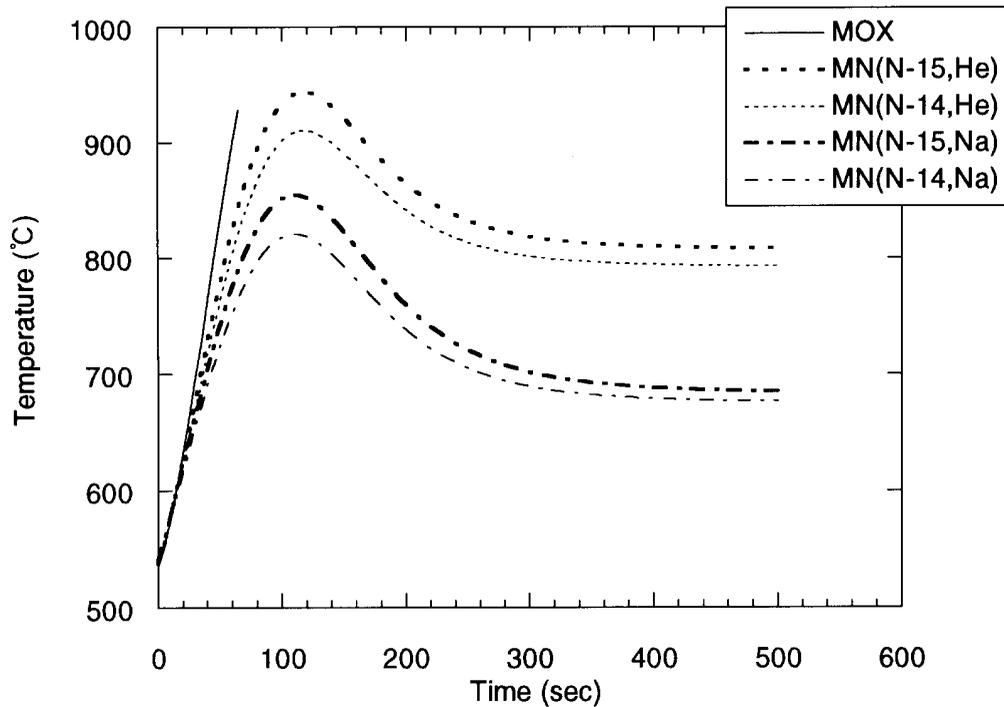
The transient analysis in the unprotected loss of flow accidents was made for each core using the reactivity coefficients shown in Table 1. These cores are not optimum in point of reactivity effects, therefore, additional negative feedback equal to radial expansion effect is assumed for each core in these analyses. The variation of the coolant temperature is shown in Figure 4 as a function of time during transients. Here again, the nitride cores show good dynamic performance without sodium boiling.

Pyrochemical reprocessing of nitride fuel transmutor

Thermodynamic predictions were made on the molten-salt pyrochemical reprocessing of the irradiated nitride fuel from the transmutor [4]. The process is similar to that of the pyrochemical reprocessing of the metal fuel [5]: the nitride is anodically dissolved and the actinide metals are deposited on the liquid-cadmium cathode. Simulation calculations were made with the use of the

Figure 4. Coolant temperature profile at unprotected loss of flow accidents

(Pump coastdown time: 20 sec)



PALEO code to determine the amounts of actinides recovered in the electrorefiner and to estimate decontamination factors of fission products. The PALEO code, similar to the TRAIL code [6], calculates electrochemical behaviour for TRU or U using a diffusion layer model. In this calculation, the basic data such as standard potentials of actinides and lanthanides were taken from Ref. [7], and the rare earths (REs) are represented by Ce which is one of the most difficult nuclides to be separated from actinides. It was found from preliminary calculations that the Ce concentration in the salt should be lower than 3.0 wt% to keep the decontamination factor of about 4 to recover more than 99.99% of TRU, considering the returned TRU + Ce from the Cd-U extraction. Equilibrium concentrations of main elements in LiCl-KCl eutectic were set to be 3.5 wt% for plutonium, 2.0 wt% for uranium, 6.5 wt% for minor actinides (Am) and 3.0 wt% for rare earths (Ce) so that the element concentration in the output actinides from the electrorefining process is almost the same as that in the input actinides. At the first stage of electrorefining, RE (Ce), Pu, MA (Am) and finally U are well dissolved. MA (Am) and RE (Ce) are deposited on the liquid Cd cathode. The process time is about six hours.

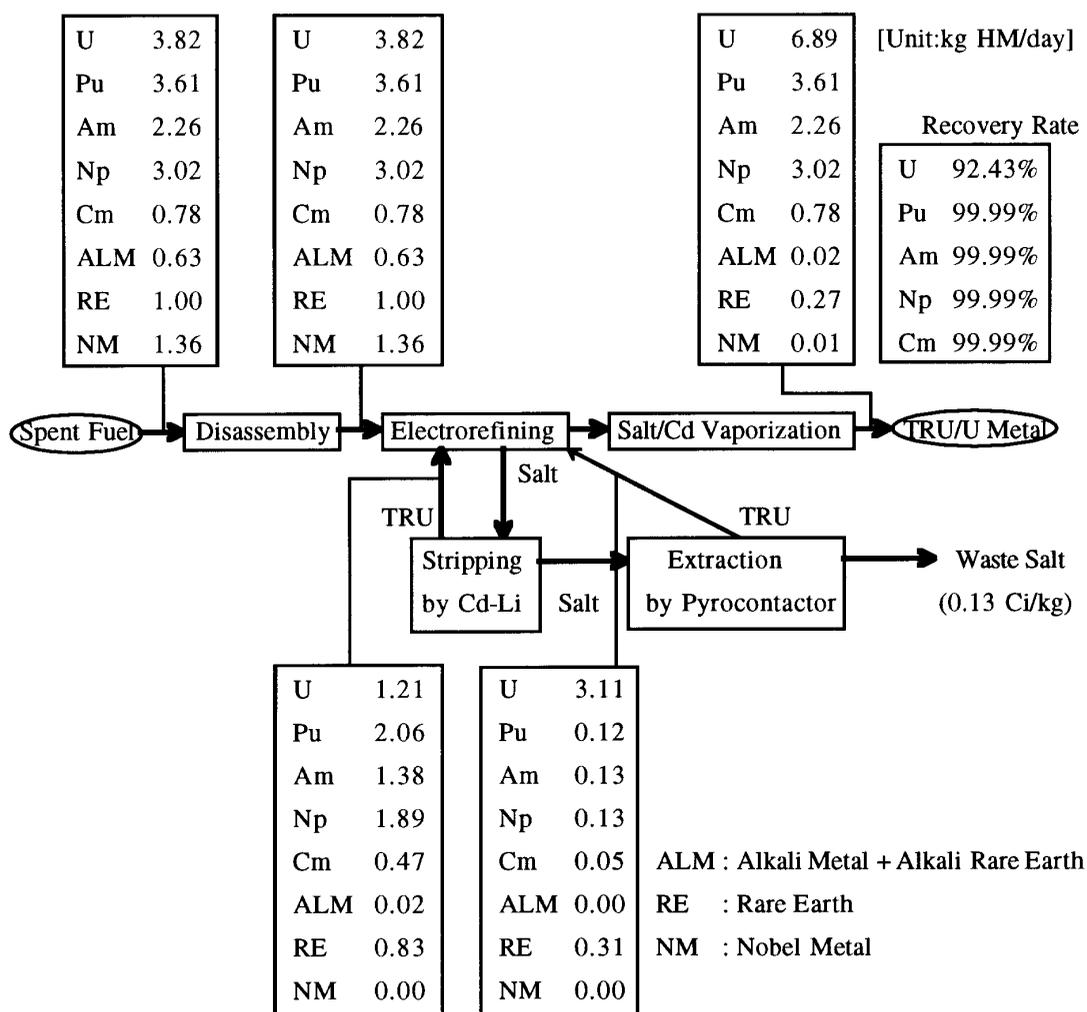
In order to keep the rare earth concentration in salt around 3.0 wt%, some amount of the salt (2.5 wt% of the salt) has to be removed from the electrorefining vessel at each batch. The removed salt first goes through a stripping step with Cd-Li, and then a multi stage extraction step with Cd-U. The recovery rate of the actinides and the rare earth concentration in heavy metal in the stripping and extraction steps are evaluated as a function of uranium distribution coefficient or Cd/salt ratio. Considering the recovery rates of the actinides with rare earth concentration in HM, the values of 0.07 for U distribution coefficient and 3 for the Cd-Li/salt ratio in the stripping process were selected. The stripping and extraction steps remove almost all the actinide from the salt to the cadmium, however, with the amount of 64% of the rare earths. Actinides recovered in the cadmium solution are returned to the electrorefiner.

The recovery rate of the actinides during the process are shown below:

Element	Electrorefining	Stripping process	Extraction process
U	75.40%	96.96%/99.95%	-/92.43%
Pu	62.34%	94.44%/97.90%	99.99%/99.99%
Np	59.94%	93.77%/97.50%	99.98%/99.99%
Am	59.94%	91.20%/96.47%	99.88%/99.99%
Cm	59.94%	90.07%/96.02%	99.76%/99.99%

Figure 5 summarises the mass flow in the electrorefining and salt treatment process. Annual processing capacity is set to be 3.5 tonne-HMeq (heavy metal equivalent) corresponding to the amount of spent fuels from 3×800 MWt P-ABR [8]. About 17.5 kg-HMeq per day are processed when the annual working day is assumed to be 200 days. Without considering the process loss, more than 99.99% of actinides can be recovered after adding those from the salt process except uranium.

Figure 5. Mass flow in electrorefining and salt treatment process



In order to separate the actinides more efficiently from lanthanides, Ogawa, *et al.* proposed a new process, the LINEX process [9], in which nitridation of actinides was made directly in the salt by using a reaction with Li_3N . Ogawa, *et al.* also discussed the application of the LINEX process in various pyrochemical processes, such as removal of actinides and lanthanides from the waste salts, and recycling of the expensive ^{15}N [10].

Concluding remarks

Core performances of the nitride fuelled fast reactor were examined with parameters of bonding materials and ^{15}N enrichment. We must still address the various problems or unknown factors such as irradiation performance of nitride fuel, ^{14}C toxicity, the higher cost of ^{15}N enrichment, etc., but the nitride core shows good core performances under normal operation and accidental condition.

The pyrochemical process can be adapted to reprocessing the nitride spent fuel of the transmutor. A new method, for example the LINEX process, is needed to remove the actinides more efficiently from the fission products in the salt.

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A FAST SPECTRUM Pu BURNER REACTOR WITH NITRIDE FUEL

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Abstract

A large fast spectrum plutonium burner design involving pure PuN fuel has been investigated. A detailed study has been performed, showing the overall neutronic feasibility of such a core. The use of a moderator ($^{11}\text{B}_4\text{C}$) in the unfuelled pins allows to recover a significant Doppler feedback despite the absence of ^{238}U ; the sodium void reactivity remains modest and the delayed neutron fraction quite high. According to the cycle management retained, shutdown requirements would be met assuming an intermediate mid-cycle shutdown for diluent management. However, the fuel peak burn-up is very high (46%), and the uncertainties on data and methods may call for further, more accurate assessments. Some of these are presented. A detailed transient analysis has also been performed.

Introduction

Ways to reach large plutonium consumptions in a fast reactor have been investigated in the frame of the CAPRA programme. A reference core has been designed, using a MOX (U/Pu) fuel with a high (45%) plutonium content. This content has been limited to 45% to ensure spent fuel solubility during PUREX reprocessing operations. Such a plutonium content already allows to achieve high plutonium consumptions (≈ 75 kg/TWhe), but a residual conversion from ^{238}U still remains and prevents reaching the potential fission rate of ≈ 110 kg/TWhe. A radical way to get rid of this residual conversion would be to use uranium-free fuels.

Indeed, some uranium-free designs have been examined, with or without inert matrix, and among them pure PuN fuels are considered. A pure PuN fuel is expected to dissolve without significant residues using the PUREX process. The main contribution to the Doppler feedback in conventional MOX fast reactor cores comes from ^{238}U . In uranium-free plutonium fuels, only the even isotopes of plutonium can contribute to the Doppler feedback. A degraded plutonium composition has therefore been considered which contains large concentrations of ^{240}Pu and ^{242}Pu ; in addition, parametrical studies using other Pu qualities have been performed.

A pure PuN fuel would need to be diluted by using an inert matrix, or by resorting to very innovative pin designs (such as a very thin annular fuel concept). For the purpose of neutronic calculations, a homogeneous composition will be taken into account hereafter, so that this choice remains open (provided the inert matrix is really inert).

Core description – calculation tools and methods

The core is rated at 1 450 MWe (3 600 MWth), has a fissile height of 1 m, and contains neither axial nor radial breeders. There are 366 fuel S/As, 120 in the inner core and 246 in the outer core. In addition, there are 52 steel diluent S/As included within the core (see Figure 1). The S/A pitch is of 181.4 mm. This corresponds to the standard CAPRA core layout (the so-called 04/94 version of the core [1]). But at the S/A level, the PuN core differs greatly from the standard CAPRA 04/94 core.

Each fuel S/A consists of 318 fissile pins and 151 moderator pins containing $^{11}\text{B}_4\text{C}$. As pure PuN fuel is used, the dilution of the fuel in the core must be greater than even the conventional MOX CAPRA design. Very innovative annular pin designs have been devised for the pure PuN fuel (i.e. with no inert matrix); e.g. each fissile pin may consist of two concentric steel cladding tubes, with low diameter (typically 50 μm) PuN fuel particles poured into the annular space between them. In any case, the neutronic calculations involve a two-zone 1-D cylindrical model for cell calculations, and homogenised S/A compositions for core calculations, and are mostly sensitive to the PuN volume fraction, so as to be able to represent a PuN + inert matrix concept as well. Indeed, the core has two radial zones containing two different PuN volume fractions.

The control and shutdown rod (CSD) and the diverse shutdown rod (DSD) positions are the same as for the CAPRA oxide reference core [1]. There are 24 CSDs and 9 DSDs. The boron carbide in the control rod absorber is modelled with a ^{10}B enrichment of 30%, this low value being used to compensate for the lack of an accurate rod homogenisation.

A six-batch fuel cycle was used, with the six inner core batches being half the size of the six outer core batches. Only half the batches are refuelled each cycle, using two alternate refuelling schemes; each fuel S/A thus remains in its position for two cycles before being moved. The cycle length used was 170 EFPD (and thus the fuel residence time 1 020 EFPD), and a load factor of 80% was assumed.

As said before, a degraded feed plutonium composition was used (w%):

^{238}Pu	3.37	^{241}Pu	8.04
^{239}Pu	32.64	^{242}Pu	15.38
^{240}Pu	39.78	^{241}Am	0.80

The European fast reactor code system ERANOS [2] has been used to perform the core neutronic calculations and nuclear cross-section data is taken from the CARNAVAL-IV library in a broad 25 neutron energy group structure. To achieve a higher accuracy on the contributions of the even Pu isotopes to the Doppler feedback, a more recent library using JEF-2.2 data, with a 33 energy group structure, has also been used. A hexagonal 3-D model of a 120° sector of the core was used, with an axial mesh size of 5 cm for flux calculations and 10 cm for burn-up calculations. Each S/A is modelled radially by seven points at each axial height, one at each vertex of the hexagonal S/A and one at the centre.

The core is modelled with each fuel batch having its correct burn-up in order to give a realistic burn-up profile. Data corresponding to the mean irradiation conditions were produced by a series of seven iterations starting from the configuration with all batches having clean core isotopic compositions. The volume fractions were adjusted by having an end of cycle k_{eff} of 1 with all rods withdrawn, and balancing the peak linear ratings between the two fuel zones.

Main results

The main results are summarised in Table 1. Comments and complements can be found in the sections that follow.

Ratings, temperature differences and damage rates

The peak pin rating is assumed to be approximated by the maximum of all the radial points of all the radial heights of the mesh used. The overall peak value is 561 W/cm, at start of cycle conditions. This is higher than the upper limit for EFR.

The maximum temperature differences between adjacent S/As is 78°C at start of cycle conditions. This has been calculated assuming a 3°C temperature rise for each 10 W/cm^3 rating increase. The 78°C value is close to the upper limit above which thermal striping could damage the above core structure.

The Peak pin damages over life were calculated taking into account the shuffling schemes. The maximum values obtained were 138 dpa NRT Fe.

Peak burn-up

It was calculated to be 45.8% both in the inner and outer cores. This is an extremely high value, to be compared to the 20% peak burn-up used for the EFR or CAPRA cores. This may well require the use of an inert matrix for the fuel.

Table 1. Main neutronic parameters – comparison to the 04/94 oxide core

		PuN core	Ref. core
Cycle length	EFPD	170	285
Number of cycles	–	6	3
Load factor	–	0.80	0.80
Fuel volume fractions (PuN core)	%	10.76/12.29	21.40/21.40
Enrichments (oxide core)	%	–	42.97/44.72
Number of fuelled pins per S/A (out of 469)	–	318	336
Peak linear ratings (inner/outer)	SOC	W/cm	516.6/560.5
	MOC	W/cm	510.1/525.9
	EOC	W/cm	481.5/470.5
Max. rating diff. between adjacent S/As	W/cm ³	266.7	–
Max. temp. diff. between adjacent S/As	°C	80.0	–
Sodium void (SOC/EOC)	fuel only	pcm	597/1 146
	fuel + diluents	pcm	-585/-133
Doppler constant (temp. reduced to 180°C)	SOC	pcm	-323
	EOC	pcm	-409
Peak pin burn-up	% h.a.	45.8	20
Total delayed neutron fraction	pcm	303	324
Prompt neutron lifetime	s	1.23 10 ⁻⁶	8.4 10 ⁻⁷
Peak damages (inner/outer core)	dpa NRT Fe	138 / 138	124
Reactivity loss over cycle	pcm	7 534	9 158
CSD + DSD worth	pcm	12 583	13 433
CSD worth	pcm	10 799	11 902
CSD + DSD worth (2 rods missing)	pcm	10 091	8 946
CSD + DSD worth (1 rod replaced by new fuel)	pcm	10 537	9 881
Rod withdrawal worth	pcm	983	–
Power rise from rod withdrawal	%/pcm	0.132	–
Pu burn-up rate (in core)	kg/TWhe	125.9	74.2

Reactivity loss over cycle

It has been calculated between end and start of cycle conditions with zero CSD rod insertion. The reactivity loss obtained, 7 534 pcm over one cycle or 44.3 pcm/EFPD is very important, as could be expected from the lack of conversion from ²³⁸U to ²³⁹Pu.

Doppler constant

It has been computed at start and end of cycle conditions by changing the mean core fuel temperature from 700°C to 180°C. It should be noted that the Doppler constant at start of cycle is significantly lower than at end of cycle. This will have implications for safety studies.

The Doppler constant has been computed assuming a $1/T$ relationship to the reactivity change as in oxide fuelled cores: $K_D = \frac{1}{T} \frac{dk}{dT}$. This dependence is not related to an underlying physical attribute, but has been found to closely represent the observed temperature dependence for uranium containing cores. However the PuN core considered here contains no uranium. Hence, further calculations have also been performed, using JEF-2.2 data to examine the temperature dependence, with four sets of cross-sections, computed at different temperatures: 573 K, 973 K, 1473 K and 2973 K, which are tabulated temperatures in the JEF-2.2 cross-section library. The results indicate a constant Doppler coefficient in the temperature range of 573-1473 K, although this comes from the compensation of many positive and negative contributions from both capture and fission in different energy ranges. In the range 1473 K to 2973 K, there is an indication that the Doppler coefficient increases by about 10%.

The Doppler effect of ^{242}Pu and of iron is not included in the data set used, which furthermore was established for oxide fuels with a low Pu content rather than for Pu without U fuels. Studies have shown that these calculations underestimate the Doppler constant. For example, to use a library with the resonance temperature behaviour of ^{242}Pu leads to increase the Doppler constant by almost 20%.

The moderator in the unfuelled pins helps much in recovering a significant Doppler constant. It has been computed that without it the Doppler constant would cut by half.

Sodium void reactivity

Two voiding calculations have been performed: firstly voiding from the fissile regions of the fuel S/As alone, and secondly from the fissile regions of the fuel and from the full height of the diluents. As the reactivity calculated on voiding the sodium from both the fuel and diluent S/As is very sensitive to the diluent design and to the calculation scheme (e.g. diffusion vs. transport), the values quoted should be interpreted in terms of general trend only.

Doppler and sodium void vs. Pu isotopic composition

In this part of the study, the PuN core has been loaded with three different Pu isotopic compositions (composition C2 is very close to the one used above):

	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am
C1	1.8	57.9	22.5	11.1	5.6	1.1
C2	3.4	32.6	40.1	8.1	15.5	0.8
C3	2.8	16.9	50.4	8.3	20.8	0.8

Changes in the fuel volume fraction needed, sodium void reactivity, Doppler constant, delayed neutron fraction and prompt neutron lifetime have been assessed. All these calculations have been performed using a JEF-2.2 library.

	X_{fuel}	$\Delta\rho_{\text{Na}}$ (EOC)	K_D (EOC)	β_{eff} (EOC)	Λ (EOC)
C1 vs. C2	-30%	-70%	-40%	-1%	+50%
C3 vs. C2	+25%	+44%	+50%	+5%	-15%

This shows that the Doppler constant and the sodium void worth are closely related to the quality of the feed plutonium, and mainly to the ^{240}Pu content in the feed plutonium.

Delayed neutron fraction

Despite the absence of ^{238}U , the delayed neutron fraction remains quite high. It is worth noting that 40-45% of the effect is due to delayed neutrons coming from ^{241}Pu , which has a delayed neutron fraction much more important than ^{239}Pu .

Rod array worth, rod faults, handling errors and shutdown margins

There needs to be an intermediate shutdown at mid-cycle conditions in order to meet shutdown requirements.

Inventories and plutonium consumption

The overall plutonium consumption amounts to 125.9 kg/TWhe from start to end of life conditions, and to 125.7 kg/TWhe if a subsequent three-year cooling period is assumed before reprocessing.

Transient analysis of the CAPRA PuN core

Transient analysis studies were carried out to investigate the behaviour of the PuN core. Three transients correspond to EFR design basis initiating events: unprotected slow loss of flow (USLOF), e.g. rundown of all the primary sodium pumps, unprotected fast loss of flow (UFLOF), e.g. a diagrid feed pipe failure, and unprotected slow transient over-power (USTOP), e.g. removal of all the CSD rods at the maximum possible rate allowed by the motor-drive system. Three unprotected fast transient over-power (UFTOP) variants corresponding to beyond basis events were also carried out: a reactivity step, e.g. for fuel relocation, a reactivity ramp, e.g. for core support failure, and a reactivity pulse, e.g. for a fast gas bubble passage.

Analysis was performed based on physics data calculated for the reference quality Pu and the assumed behaviour of the innovative fuel pin design (a very thin annular fuel disposition between two concentric claddings. Parametrical studies also involve three other Pu qualities (% by mass):

	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am
High	1.8	57.9	22.5	11.1	5.6	1.1
Good	5.6	39.1	26.7	13.0	14.3	1.3
Reference	3.4	32.6	39.8	8.0	15.4	0.8
Poor	2.8	16.9	50.4	8.3	20.8	0.8

It should be noted that there is little information currently available concerning the behaviour of PuN and about the innovative pin design. It was therefore necessary to make many assumptions and simplifications in these studies, and the results presented here should be considered as preliminary.

The analysis was carried out using the CRAFT core transient analysis code. This code is basically a three-dimensional S/A and fuel pin thermal hydraulics model combined with a point kinetics model. The CRAFT model of this core represents a full 120° sector of the core and contains a total of 245 channels. Two fuel pin sub-channels are used to represent each fuel S/A in the core: one for the pins in the bundle centre and one for the pins adjacent to the wrapper. One channel represents

the sodium flow contribution from the diluents absorbers, and first row of shield S/As. The core height is represented by 10 axial meshes. Linear reactivity coefficients are used, one for each node, to calculate the individual effects on the global reactivity.

No phase change was predicted for USLOF and UFLOF transients. Inner clad melting (11-19 sec. after initiation) and sodium voiding (34-48 sec. after initiation) were predicted for USTOP transients. The poorer the Pu grade, the later the phases changes occur. In all these design basis transients, the main mitigation against the effects of the transient is feedback from the expansion of the control rod support which causes the rods to be inserted further into the core.

For fast transient over-powers, the inner clad melting occurs very quickly (0.5-2.0 sec. after initiation) and is followed by sodium boiling in the ramp UFTOP transient. The low Doppler constant representative of the higher quality Pu cases results in more severe consequences than for the poorer cases. In the most severe case (pulse UFTOP) none of the trip systems could respond sufficiently quickly to be effective.

Conclusions

These studies are preliminary due to calculational uncertainties for uranium-free cores, and to the lack of experimental data for PuN fuel, and there remains considerable scope for further study (e.g. on the batch loading pattern and diluent management, or on transient assessment). Nevertheless, some trends can be drawn from the above results.

The PuN core exhibits a much larger reactivity loss per day than the CAPRA reference oxide core (+40%) and has a shorter cycle length (increased number of batches). The sodium void worth and the Doppler constant are reduced, although there is a large uncertainty on these values. The peak burn-up reaches 46% heavy atoms and the Pu burn-up rate is approximately 125 kg/TWhe. The fuel loading pattern has been optimised in order to reduce the differences in sodium outlet temperatures between adjacent subassemblies which can have very different burn-ups.

Six reactivity transients have been investigated for the core (USLOF, UFLOF, USTOP and three variants of UFTOP). The three first transients correspond to EFR transients with design basis initiating faults, while the three UFTOP variants correspond to beyond design faults. Due to the lack of experimental data about the behaviour of PuN fuel, the results should be considered as preliminary.

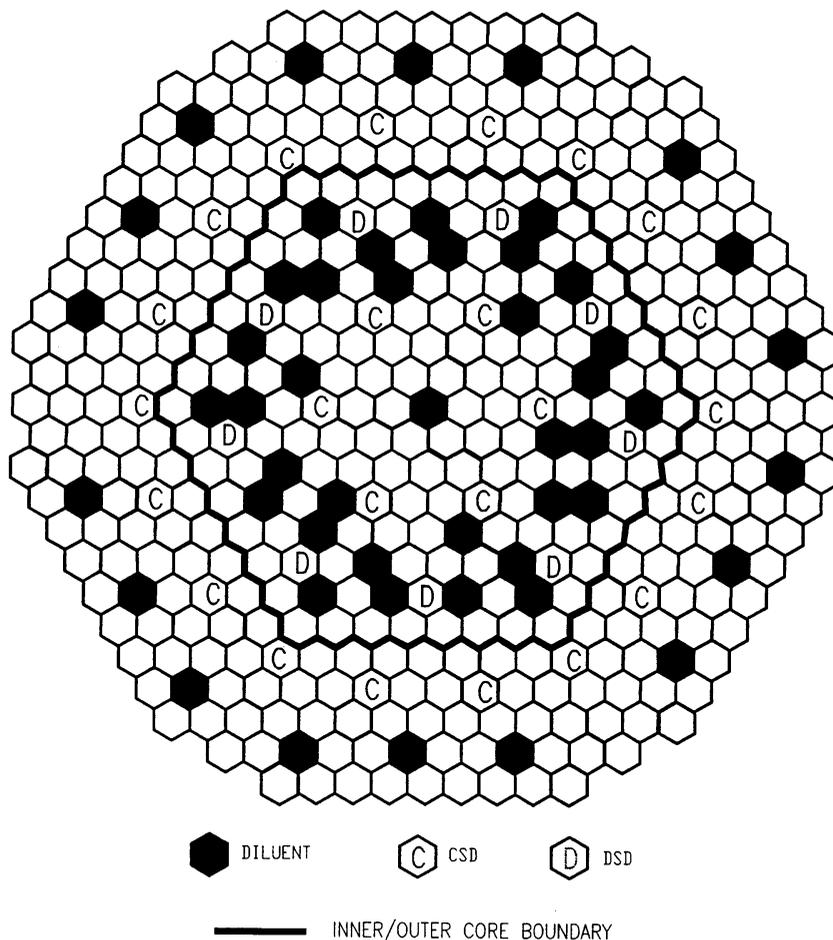
For design basis transients, the main mitigating effect is feedback from the expansion of the control rod support; no phase changes occur, except in USTOP transients (inner clad melting). For beyond design basis initiated transients, the first phase change is melting of the inner clad; in severe cases (pulse UFTOP variant) none of the trip systems respond sufficiently quickly to be effective.

Overall, these *neutronic* studies show that PuN cores may be feasible, however, their performance is limited relative to the standard oxide CAPRA core.

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Figure 1. 120° sector layout, showing the fuel batches



CONCEPTUAL CORE DESIGN FOR URANIUM METALLIC FUELLED LIQUID METAL REACTOR

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Abstract

A uranium metallic fuelled core with 150 MWe (392 MWt) capacity which uses only U-Zr binary fuel not in excess of 20 w/o enrichment is proposed in this study. The core design was justified throughout extensive studies for design parameters and fuel management schemes. As a result, an economic and safe equilibrium uranium metallic fuelled core design was established. Optimisation has been further achieved by adopting strategic loading schemes for initial and transition cycles to reach target equilibrium cycles early. An iterative method has been developed to find initial core enrichment splits. With non-uniform feed enrichments adopted at the initial core, this iterative method shows the core can reach uranium equilibrium cycles after just four reloads, keeping feed enrichment unchanged from the second cycle. The transition cycle analysis indicated that recycling of self-generated plutonium is not sufficient to make KALIMER a pure plutonium equilibrium core even after 56 reloads.

Introduction

A comparative study had been performed for various possible design alternatives to develop the conceptual uranium fuelled LMR design which could serve as a starting reference design for further design optimisation, with the projection of incurring a domestic technology development and building experiences in the nuclear industry infrastructure [1]. These efforts came up with a 150 MWe rated KALIMER liquid metal reactor (LMR) design with enhanced safety features. It is fuelled with U-Zr binary alloy fuel not exceeding 20 w/o enrichment to satisfy the Korean nuclear non-proliferation policy. The most demanding shortcomings in the uranium metallic fuelled core come from a reduced breeding and an increased active core volume, and thus result in an increased reactivity swing and a relatively low core average linear heat generation rate (LHGR) compared with typical plutonium metallic fuelled cores.

Intrigued by such an unusually low LHGR, many different design options and a variety of fuel management schemes were investigated in uranium equilibrium cycles for the purpose of either increasing LHGR or improving fuel cycle economics with the once-through cycle assumption for an uranium equilibrium core [2]. This paper will focus on the findings and discussions on the evolution of a safe and economic uranium metallic fuelled LMR core. Also covered are initial and transition cycle features from uranium fuelled initial cycle to uranium equilibrium cycles, together with transitions to a plutonium equilibrium core using the recycling of self-generated plutonium. As results of these works, this paper presents a number of transition cycle options to reach equilibrium cycle and the evolutions of heavy metallic fuel mass balances, power distributions, breeding ratios for projected KALIMER uranium initial, transition and equilibrium cores.

Reference core design description and analysis methodology

Reference core description

As a starting core configuration for an annually refuelled 392 MWt reactor, the two-enrichment zoned, homogeneous core shown in Figure 1 was taken. There is no axial blanket. The active core height is 100 cm and the height-to-diameter ratio (H/D) is 0.481. The core and fuel design parameters, and operating conditions are given in Table 1. The driver and blanket fuels have smeared densities of 75% and 85%, respectively. Refuelling occurs after 12 months of operation at 85% capacity factor, with one-third of the driver fuel and blanket assemblies being replaced during each outage. Following removal from the core, driver fuels and blankets are allowed to decay for one operating cycle in the in-vessel storage (IVS) positions.

Design analysis methodology

The core design and evaluation were performed with the calculation module packages in the K-CORE System [3]. The evaluation process was initiated by the generation of region-wise microscopic cross-sections, based on the self-shielding factor approach. The composition dependent, region-wise nin-group microscopic cross-sections were generated from the reprocessed 80-group neutron cross-section library version KAFAX [4] with the cross-section processing module.

Fuel cycle calculations were carried out with the neutron flux and burn-up calculation module consisting of DIF3D [5] and REBUS [6] codes. Flux solution calculations solved the nodal diffusion theory approach for hexagonal-z reactor representation. Especially for an equilibrium core, the fuel

Figure 1. KALIMER core layout including batch refuelling scheme (I5O5.5B7) for transition cycle analysis

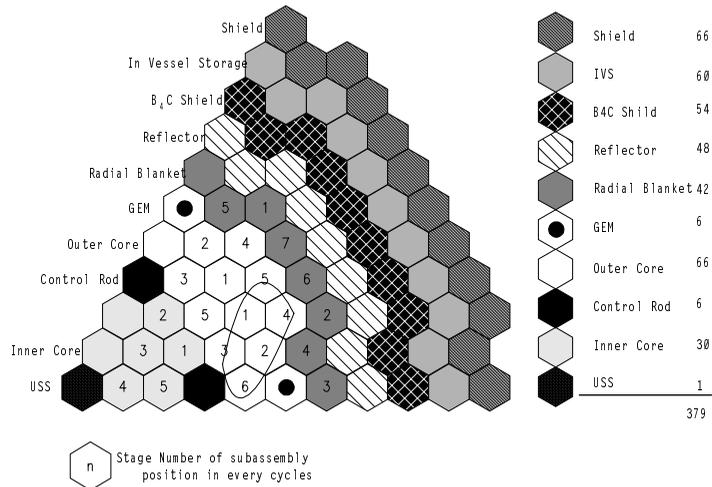


Table 1. KALIMER core design description

<u>Operating conditions</u>	
Core thermal power (MWt)	392
Core mixed mean inlet/outlet temp. (°C)	361.4/ 530.0
Feed fuel enrichments (IC/OC) (%)	16.2/20.0
Effective full power day (EFPD)	310.0
Number of batches	
Inner/outer driver fuel	3/3
Radial blanket	3
<u>Core and fuel design parameters</u>	
Active core height (cm)	100.0
Core diameter (cm)	344.3
Duct pitch (mm)	161.0
Pins per fuel assembly (driver/r. blanket)	271/127
Pin outer diameter (driver/r. blanket)(mm)	7.50/12.00

cycle computations for the operating interval under a fixed fuel management scheme were solved; this equilibrium cycle calculation approximates reactor characteristics after many cycles of operating subject to a fixed, repetitive fuel management strategy. These calculations were performed with all the control rods fully withdrawn.

Equilibrium cycle development

Design variations in equilibrium cycle

With the reference core design as given in Figure 1, design variation studies in uranium equilibrium cycles were carried out on this reference core configuration; design variations include

changes in core height, replacement of gas expansion module (GEM) with outer core fuel assembly or radial blanket assembly, increase in fuel slug diameter, axial blanket placement and variations of radial blanket batch numbers. For different core layout designs, the inner core feed enrichments were searched to meet the cycle energy extraction requirement, while the outer core feed fuels were assumed to be 20 w/o enriched U-10Zr binary form.

The nuclear performance parameters and related economics information per design are given in Table 2. In this study, the core wide three-dimensional power peaking factor was constrained to remain in the outer core region, which then would prevent subsequent sodium void propagation toward the inner core region where sodium void worth is slightly positive. Other typical ground rules applied to metallic fuelled LMR core designs do not play an important role of figure of merit, because the core average linear heat generation rate (LHGR) is so low that the other design values subject to the comparison are staying well below the typical design limits. A survey of Table 2 reveals that there are only three design alternatives – H110, FAT and GEMF – which locate the core wide peak power density in the outer core region. Observation of the natural uranium supply or the separate work requirement shows that the FAT fuel option in which the fuel slug outer diameter is slightly increased is the best among these three design alternatives. This is because the FAT fuel design with packed fuels hardens the neutron spectrum and reduces the leakage out of the core. In addition, two other designs are accompanied by the reduction of core average LHGRs and thus suffer too much from departures of the target LHGR of about 7 kW/ft.

Table 2. Core layout options and their performance characteristics in equilibrium cycles

	Reference	Core Height		Fuel Slug Diameter		GEM replaced by		Radial Blanket		Axial Blanket	
	H=100cm	H=90cm	H=110cm			OC Fuel Blanket		4 Batch	6 Batch	3 Batch	6 Batch
	H100	H90	H110	FAT	FAT-H90	GEMF	GEMX	BLK4	BLK6	XB L3	XB L6
Supplied U Enrichment [w/o]											
inner core	16.24	18.08	14.79	14.36	16.07	14.80	16.17	16.16	16.00	16.05	15.81
outer core	20.00	20.00	20.00	20.00	20.00	20.00	20.00	20.00	20.00	20.00	20.00
radial blanket	0.20	0.20	0.20	0.20	0.20	0.20	0.20	0.20	0.20	0.20	0.20
Natural U Supply [Kg]											
inner core	30449	30855	30212	28514	29059	27711	30312	30297	29985	30085	29627
outer core	82668	75153	90183	87739	79763	90183	82668	82668	82668	82668	82668
total	113117	106008	120395	116254	108822	117894	112980	112965	112653	112753	112295
Separate Work [SW U]											
inner core	35232	36089	34604	32548	33585	31742	35056	35037	34639	34767	34183
outer core	97602	88729	106474	103589	94172	106474	97602	97602	97602	97602	97602
total	132834	124817	141078	136137	127757	138216	132658	132638	132241	132369	131785
Discharge Burnup [MW D/Kg U]											
inner core average	44.50	52.97	37.96	38.03	45.74	38.58	44.35	44.10	43.31	43.42	42.23
outer core average	33.95	35.65	32.33	33.73	35.45	33.52	33.75	33.96	33.98	33.44	33.46
Peak LHGR [KW /ft]	6.9	8.2	6.1	6.6	7.3	6.2	6.8	6.8	6.6	6.7	6.5
Peak Position [x,y,s]	2,1,4	2,1,4	5,2,5	5,2,5	2,1,4	5,2,5	2,1,4	2,1,4	2,1,4	2,1,4	2,1,4
Breeding Ratio	0.66	0.62	0.68	0.68	0.65	0.67	0.68	0.65	0.65	0.78	0.78
Reactivity Swing [pcm]	1465	1858	1174	1159	1524	1224	1406	1448	1414	1295	1247

With the FAT fuel design selected as the best option, reactivity parameters such as sodium void worth, control rod worths and Doppler coefficients were evaluated to justify the new core layout (see Table 3). Only a sodium voiding in the inner core region gives rise to a positive sodium void worth. The gas expansion module (GEM) is extremely efficient, resulting in a negative reactivity of 970 pcm which is sufficient to make the core subcritical by 777 pcm at BOEC, upon combination with the sodium voiding in the inner core region. Toward EOEC, the power distribution rolls slightly into the inner region of the core, and thus the sodium void worth at EOEC tends to be more positive or less negative regardless of sodium voiding location, compared with those of BOEC.

Table 3. Reactivity effects in uranium equilibrium core

Reactivity parameter	BOEC	EOEC
Temperature coefficient [$\Delta k/k/K$]		
Doppler temperature coefficient	$-0.0816 T[K]^{-1.4}$	$-0.0886 T[K]^{-1.4}$
Structure temperature coefficient ($\times 10^{-4}$)	$-1.0928 T[K]^{0.85}$	$-1.1299 T[K]^{0.85}$
Sodium void worth (pcm)		
Inner core	232	315
Outer core	-735	-655
Inner and outer cores	-517	-335
Outer core and radial blanket	-935	-857
Inner core and GEM	-777	-698
GEM worth (pcm)	-972	-973
Control rod worth [% $\Delta k/k$]	17.86	18.01

While the determined core configuration assures safe power plant operations, it has too low a core average linear power (4.2 kW/ft) and eventual low discharge burn-ups. Since the reactor breeding ratio is just around 0.68 with 20 w/o enriched U-Zr binary fuel, the increase of core average linear power would not be plausible due to too high depletion rates of fissile nuclides, unless the fuel cycle length is shortened.

Based on the fixed core configuration, fuel management studies further proceeded by changing fuel management schemes to improve fuel cycle economics as well as to achieve a flatter power distribution. Concerning the fuel cycle economics, the fuel residence time in the core plays an important role in determining the fuel cycle cost. With the FAT fuel design adopted, various fuel management options which enable extended burn-ups were extensively evaluated. After the I4O4B3 refuelling scheme (i.e. 4, 4, 3 batches for inner core, outer core, radial blanket), refuelling schemes with next higher batch numbers began to violate the ground rule regarding the location of peak power density in the REBUS equilibrium cycle mode. However, even with this increased batch operation, the discharge burn-up is still far less than 100 MWd/kgU. In order to extend fuel residence time beyond four-batch operations of inner and outer core fuels, it was necessary to integrate design changes previously abandoned for the I3O3 refuelling scheme. It can be seen in Table 4 that with the FAT GM_I7O7 design (i.e. fat fuel used, GEM replaced by outer core fuels, and 7/7 batches for inner and outer core regions), both the natural uranium supply and separate work are reduced to 47% of those for the FAT I3O3 design.

Target equilibrium cycle for transition cycle development

An equilibrium cycle analysis by REBUS models a scatter-reloading in which explicit assembly positions are occupied by an artificial mixture of assembly compositions in a few stages. In order to account for a realistic loading of fresh fuel assemblies, an explicit cycle analysis needs to be done even for the equilibrium cycle core. Such an explicit cycle analysis could provide bounding physics data over the reactor lifetime in either an initial core or transition cycle cores. If the positioning of fresh fuels in the outermost ring of the outer core region can be avoided, there are some rooms that the core wide maximum power density still resides in the outer core. When the shuffling is not exercised, transition cycles will show cyclic feed enrichment variations, depending on the position

Table 4. Fuel management options and their performance characteristics in equilibrium cycles

	FAT Fuels are used								
	Batch Number/Variation			O.C. Fuels in GFM		H=110cm			
	M04	I505	I505.5B6	GM I606	GM I707	HT I606	HT I707	GM HT I606	
Supplied U Enrichment [w/o]									
inner core	14.96	15.64	15.49	14.66	15.39	14.61	15.27	12.88	
outer core	20.00	20.00	20.00	20.00	20.00	20.00	20.00	20.00	
radial blanket	0.20	0.20	0.20	0.20	0.20	0.20	0.20	0.20	
Natural U Supply [Kg]									
inner core	22305	18658	18478	14560	13114	15836	14188	13935	
outer core	65805	52644	47858	47858	41021	47858	41021	52209	
total	88110	71302	66336	62418	54135	63694	55209	66144	
Separate Work [SWU]									
inner core	25580	21502	21272	16659	15087	18113	16307	15701	
outer core	77692	62153	56503	56503	48431	56503	48431	61640	
total	103272	83655	77776	73162	63518	74616	64739	77341	
Discharge Burnup [MWD/KgU]									
inner core average	52.52	67.85	66.56	72.57	87.76	71.28	85.76	60.52	
outer core average	44.16	54.22	59.13	63.20	72.46	61.02	70.02	60.28	
Peak LHGR [KW /ft]	6.5	6.5	6.4	6.1	6.0	5.9	5.9	5.8	
Peak Position [x,y,s]	5,3,4	2,1,4	2,1,4	5,2,5	5,2,5	5,2,5	2,1,4	5,2,5	
Breeding Ratio	0.68	0.68	0.67	0.69	0.68	0.70	0.70	0.71	
Reactivity Swing [pcm]	1259	1347	1311	1179	1259	1125	1202	916	

where fresh feed assemblies are loaded. This cyclic behaviour of feed enrichment variations even after a number of cycles, can be avoided by assuming the fuel shuffling and thereby keeping fuel management paths invariant for every cycle. This possibility was taken into a detailed analysis for the transition cycle analysis.

Taking into account the fact that the outer core consists of 66 assemblies, a fuel management strategy for the outer core region can be given by either five or six batches. Therefore, an explicit cycle analysis was decided to be done in 5/5.5/7 batches for inner/outer/blanket assemblies. The increase in batch number for the outer core region tends to move the core wide maximum pin power peaking toward the inner region of the core. Although the equilibrium mode run of REBUS indicates the maximum pin power peaking in the inner region, this can be avoided as soon as a judicious assembly shuffling is assumed.

When the shuffling of inner/outer/blanket are assumed, there arises the necessity of developing the fuel shuffling scheme. From exploratory searches, a shuffling pattern with the minimum power peak in one of its non-equilibrium cycles was decided to be the equilibrium cycle shuffling pattern. The shuffling pattern determined in this manner is shown in Figure 1, which implies the I505.5B7 batch operation. Table 4 indicates that this operation with the fuel shuffling would allow the average discharge burn-ups by 67 and 59 MWD/kgU for inner and outer core fuels.

Transition cycle development

From the point of view of core neutronics and design, the final goals for the uranium initial (start-up) core option shall be to keep good fuel cycle economics throughout several reloads and to draw the least amount of operating license concerns related with performance characteristics for transition cycles. In this frame of work, various in-core fuel management strategies for the first several cycles after the uranium initial core were extensively evaluated.

In search of transition cycles to the equilibrium cycle, there can be two different scenarios of achieving equilibrium cycles. One may allow the cycle length as a free variable, and feed the same

enrichment once the equilibrium feed enrichment is already known. The other may fix the fuel cycle length, allowing the feed enrichment to change. The latter is known to usually take up more cycles to reach the equilibrium cycle. The former option is usually prohibited by the utility, because the cycle energy requirement comes from the utility. Therefore, the cycle length has been fixed with the feed enrichment left as a free variable in this analysis.

The outer core enrichment was decided to directly start from the maximum allowed 20 w/o in order to decrease the degree of freedom during the search process. Instead of a fixed outer core fuel enrichment, one can also assume the fixed enrichment split between the inner and outer core fuels. This approach will lead to an understanding of complicated figures and will help deal with small enrichment changes cycle by cycle. Therefore, only the inner core fuel enrichment was allowed to change to meet a cycle energy extraction of 310 EFPD, satisfying the EOC eigenvalue of 1.002 in the transition cycle analysis.

U-U transition cycles from initial core with uniform inner core fuel enrichment

With the shuffling pattern in Figure 1 maintained, cycle specific feed enrichments for transition cycles up to cycle 20 are given in Figure 2(a), in which whole inner core average enrichments as if they were all fresh, are also shown. The initial core starts with the uniform inner core feed enrichment of 13.45 w/o. Even after 19 reloads, a stabilised feed enrichment is not yet achieved. Accordingly the inner core power distribution also did not settle down, as shown in Figure 3(a). The projected equilibrium cycle inner core feed enrichment is 15.39 w/o. The feed enrichment variations show a cyclic mode behaviour, although the fixed fuel shuffling pattern is assumed. The source of this cyclic mode behaviour was identified to be the uniform feed enrichment for the initial inner core fuel assemblies. It is obvious from the fact the feed enrichment for the second cycle is 16.4 w/o, which means that discharged fuel assemblies at the end of the first cycle were too valuable to be thrown away.

Figure 2. Evolution of feed enrichment for KALIMER U-U transition cycles

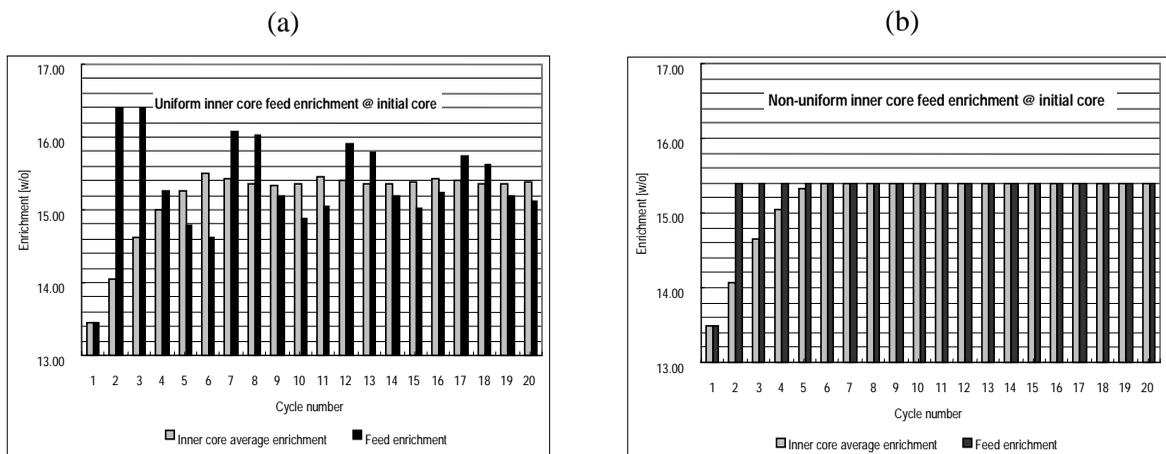
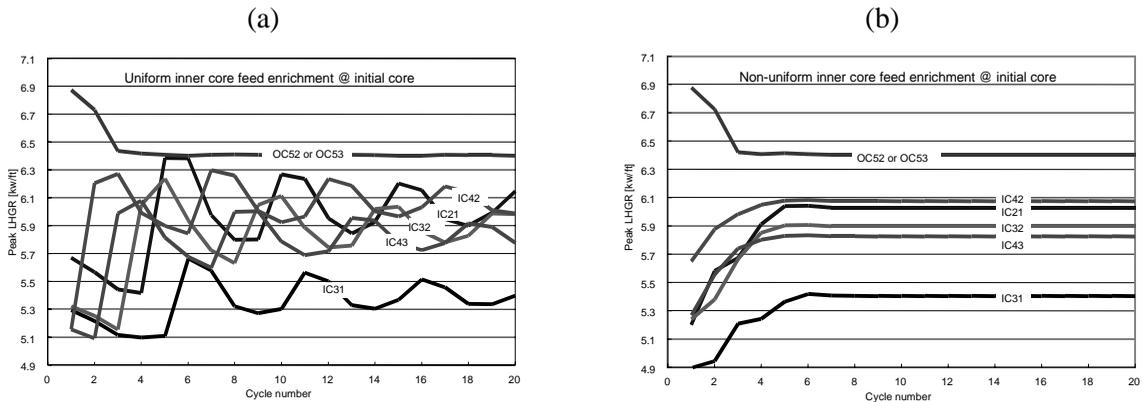


Figure 3. Evolution of inner core assembly peaks for KALIMER U-U transition cycles



U-U transition cycles from initial core with inner core enrichment split

Appropriate enrichment splits in the initial inner core were introduced to achieve the equilibrium cycle quickly. Having known the equilibrium cycle feed enrichment, an optimum initial cycle feed enrichment set for the inner core region fuels was evaluated by iterative calculations of transition cycles. For this iterative search procedure, the initial guess for the enrichment split between feed assemblies of near enrichment was decided to be 0.9 w/o, since this amount of enrichment split is approximately equal to the average reactivity loss per cycle.

Initial core feed enrichment splits were iteratively searched, with the fixed feed enrichment from the second cycle. After 13 iterations passed, the resulting initial core feed enrichments were 12.471/12.490/13.468/13.890/15.125 w/o for 5/4/3/2/1th burn-up stage fuels, respectively. Accounting for the manufacturing tolerance, these enrichments were rounded off to a single decimal digit such as 12.5/12.5/13.5/13.9/15.1 w/o. Finally, starting from the initial core loaded with assemblies of these enrichments, explicit transition cycle calculations proceeded up to cycle 20 with the feed enrichment for the second cycle and all subsequent cycles set to 15.4 w/o.

The cycle by cycle feed information is available in Figure 2(b). Figure 3(b) shows the inner core assembly power evolutions as cycles proceed. From the fifth cycle, the power distributions for the initial core with enrichment splits do not change, while those for the initial core with a uniform inner core enrichment fluctuates even after 20 cycles. The reason why the inner core power rises up at the fifth cycle comes from the fact that the outer core fuels are supplied with 20 w/o from the initial core. Once the non-uniform feed enrichments are additionally given to the outer core fuels, one would be able to get an essentially constant power distribution from the initial core throughout the reactor lifetime. In this manner, the number of transition cycles can be minimised. This feature will, then, allow to make reload licensing concerns easily manageable, since the equilibrium cycle is achieved after only a few cycles.

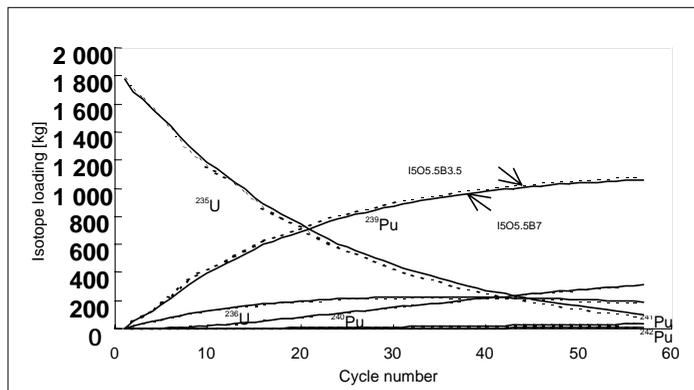
The average discharge burn-ups for inner and outer core fuels amount to 65.8 and 59.5 MWd/kgU, respectively. This is a remarkable improvement from the previous ones of 52.5 and 44.2 MWd/kgU obtained in the FAT I4O4 design [2]. These values are higher than the achievements in BN-350 and BN-600 uranium fuelled LMRs [7] and highlight the initial inner core enrichment split, because these burn-ups are achieved even with the maximum enrichment limit of 20 w/o.

Transition cycles for Pu recycled equilibrium core from initial core with inner core enrichment split

This is the case when self-generated (bred) plutonium is recycled into reload cores. The transition cycle analysis assumed one cycle cooling and one cycle fabrication. Namely, isotopes discharged at the end of cycle N are cooled and fabricated, and fed into cycle (N + 3). The first three cycles assumed the U-U transition cycles. The discharged fuel assemblies from the first cycle are cooled in the second cycle, reprocessed in the third cycle, and fed into the fourth cycle reload core. In this analysis, the first priority was given to the inner core fuel make-up. That is, reprocessed U and Pu fissiles are first fed to make up the inner core fuel for a desired inner core fuel enrichment to meet the cycle length of 310 EFPD. Excess fissiles are then fed to the outer core fuel, if they exist.

The evolution of ^{235}U , ^{236}U , ^{238}U and Pu isotopes are shown in Figure 4. Even after 57 cycles of operation, KALIMER is expected to be unable to achieve the pure Pu equilibrium core, in which ^{235}U is neither fed from the reprocessing nor from the external supply. The reactor breeding ratio at the 57th cycle is 0.97, while that of its Pu equilibrium counterpart is 1.03. The reactor breeding ratio increments per cycle suggest that KALIMER will require about 100 cycles to reach a Pu equilibrium core. This is because the Pu production is almost equal to the depletion, as the Pu enrichments approach around 12 w/o; it is true for the KALIMER core which has Pu enrichment of 11.3 w/o after 56 reloads. In order for the KALIMER core to overcome the increased Pu burnout, a massive production of Pu by blankets is required.

Figure 4. Isotope loading changes for recycled U-Pu transition cores



Conclusion

As an effort to evolve the core design, design variation studies in uranium equilibrium cycle were performed on the reference core configuration and have determined the fat fuel design option as the best among several design parameters and features adopted in the reference core design. With the adoption of the fat fuel design, fuel management studies have been further carried out by changing fuel management schemes to improve fuel cycle economics as well as to achieve a flatter power distribution. As a result, the I4O4B3 refuelling scheme was found out to be the maximum batch operation having its core wide one pin peak outside the inner core regions.

Further extended burn-ups for the discharged fuel assemblies have been achieved for the KALIMER uranium metallic fuelled core by employing the appropriate fuel shuffling with the 1505.5B7 refuelling scheme and the inner core fuel enrichment split for the initial core. Expected

average discharge burn-ups for KALIMER are higher than what have been achieved in any uranium fuelled LMRs. With the introduction of the inner core fuel enrichment split, the KALIMER core exhibits equilibrium power distribution after just four reloads, and requires a constant fuel feed enrichment from the second cycle. An early attainment of the equilibrium cycle would eliminate many licensing concerns which might arise during the transition cycles.

The transition cycle analysis with a self-generated, recycled plutonium, exhibited that the KALIMER core would need about 100 cycles to attain a plutonium equilibrium core. In the sense that the natural resource utilisation should be maximised to overcome the limits of God's gift, KALIMER needs to suffer drastic design changes.

Acknowledgements

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DYNAMIC BEHAVIOUR OF NITRIDE LMR CORE DURING UNPROTECTED TRANSIENTS

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Abstract

The dynamic behaviour of a nitride fuelled LMR core was analysed and compared to that of an oxide core in a large (3 600 MWt), pool-type, liquid-metal-cooled reactor (LMR). The study focused on three representative accident initiators with failure to scram: an unprotected loss-of-flow (ULOF), an unprotected transient overpower (UTOP), and an unprotected fast transient overpower (UFTOP). The margins to fuel melting and sodium boiling have been evaluated for these representative transients. The results show that there is an increase in safety margin with the nitride core which maintains the physical dimension of the oxide core.

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Introduction

Many countries in the world maintain the development of fast liquid-metal-cooled reactors (LMRs) as a long-term energy source and pursue R&D programmes for LMRs. In this context, the design of LMRs has been evolved and continues to do so. The safety enhancement is a reason for this evolution, particularly in core design. As regards design of LMR cores, two main options are considered. The first option is the choice of nuclear fuels: mixed oxide is the well-established reference, but mixed nitride is usually considered an attractive candidate. The second option is power levels. Large cores (1 500 MWe) are preferred in Europe whereas Russia and Japan work on medium sized cores (600 ~ 800 MWe), at least as an intermediate step. The USA developed modular cores based on small power level (~200 MWe). The criterion of choice is for both economy and safety.

From the economical point of view, high burn-up rate and high linear power (as well as high breeding gain if necessary for the original purpose of fast reactors) are needed. From the safety point of view, during core accidents, kinetics and reactivity effects are important, in particular sodium void (i.e. density change) reactivity and Doppler feedback effect.

The objective of this study is to evaluate and compare the safety potential of a nitride fast reactor core to that of an oxide core during unprotected transients. For this, it is necessary to simulate the core in the nuclear steam supply system and to analyse the dynamic behaviour of this whole system during accidental transients. These transient analyses therefore consider the reactor core, the pools, the pumps, the heat exchangers (IHXs), and other components of the cooling system that can have an impact on the progression of an event. It is essential to adequately represent the various reactivity feedback effect mechanisms including Doppler effect, sodium density changes, radial expansion of the grid plate, axial fuel expansion and control rod driveline expansion.

A comparative study with regard to safety enhancement was undertaken, involving the European Fast Reactor (EFR) [1] type core, which was used as the reference oxide core, and its variant with a nitride fuel, which has the same physical dimension as that of the reference core. In the EFR type reactor, the active core comprises 387 fuel assemblies with three different enrichment zones. Within the active core region there are 24 control and shutdown assemblies (CSD) and nine diverse shutdown assemblies (DSD). The active core is surrounded radially by one row of 78 blanket assemblies. The number of fuel pins per fuel assembly is 331. The maximum burn-up of fuel assemblies at the end of cycle – that is, after 5×340 EFPD (equivalent full power day) – reaches $170 \text{ GWd/t}_{\text{HM}}$. The thermal output of the core is 3 600 MW and the sodium inlet temperature is 395°C , with an average temperature increase across the core of 150°C .

The nitride core design derived from the EFR concept of replacing the mixed oxide with the mixed nitride. The geometry of the fuel pin was not changed, but the smear density became 85% of the theoretical density in order to accommodate the irradiation swelling of the nitride. The cycle length of assemblies in the reactor was conserved. The three enrichment zones and the enrichment ratios between the different zones were also conserved.

Out of a wide range of unprotected transients [2], three particularly significant accident initiators were considered: an unprotected loss-of-flow (ULOF), an unprotected transient over power (UTOP), and an unprotected fast transient over power (UFTOP). As is shown in the initiator names, a total failure is assumed for the plant protection system during accidents. Analyses were performed with a particular emphasis on changes in sodium and fuel temperatures so that margins to sodium boiling and fuel melting can be assessed.

Material properties of oxide and nitride fuels

Table 1 summarises the thermo-physical properties of the oxide and nitride fuels. The irradiation performance of oxide fuels has been well demonstrated up to 25 at.%. A high melting temperature compensates for a mediocre thermal conductivity which restricts its linear power to lower than 500 W/cm [3]. Therefore, the temperature gradient in the fuel pellet is important and the margin to melting is about 800°C. The neutron spectrum in an oxide core is softer than that in a metal fuelled core, which results in a large negative Doppler constant.

Table 1. Physical characteristics of oxide and nitride fuels

Fuel type	Oxide	Nitride
Nominal composition	(U,Pu _{0.25})O ₂	(U,Pu _{0.2})N
Theoretical density (g/cm ³)	10.5	14.3
Thermal conductivity at 1 000°C (W/m K)	2.62	14.66
Specific heat at 1 000°C (J/kg K)	322.6	232.5
Expansion coefficient at 1 000°C (10 ⁻⁶ /K)	9.72	8.78
Melting temperature (°C)	2 750	2 440

The nitride fuel [4] has a good thermal conductivity (almost six times the oxide fuel) and a relatively high melting temperature, which allows a high linear power with a large margin to melting. Stored heat in the fuel is considerably reduced due to a low fuel temperature during normal operation. Its high density and harder spectrum could allow a higher breeding gain than for oxide fuel. These properties of the nitride fuel seem to comply with safety requirements. That is, there is a possibility that the nitride fuel could avoid sodium boiling and fuel failure in various accidental situations.

Despite potential advantages of nitride fuels, they have not been thoroughly studied because of a lower breeding gain caused by the parasite absorption by ¹⁴N and potential problems caused by the formation of ¹⁴C. But recent studies [5,6] have reinforced the interest of nitride fuels. The enrichment in ¹⁵N is recommended in order to bring the production of ¹⁴C to an acceptable level, and preliminary evaluations indicated that it could be done at an acceptable cost. Moreover, from about 1 600°C, the nitride has a tendency to dissociate when the pressure of nitrogen in equilibrium is not sufficiently high [7]. This problem can lead to a formation of liquid metal uranium at that level of temperature and should be studied more precisely from the safety point of view.

Reactor modelling and neutronic performances

The necessary neutronic parameters were determined by a series of neutronic calculations using the European Reactor Analysis Optimised System (ERANOS) [8] computer code, and the thermal feedback coefficients were obtained by using the thermal calculation code COREA [9]. These serial calculations were performed by using an automated procedure developed during this study (Interface ERANOS-COREA) [10]. It produces neutronic data directly exploitable by COREA. The methodology used in the automated procedure for the feedback effect calculation is summarised below. For the geometrical modifications, we performed a direct calculation in diffusion theory after having increased +1% of the height or the radius of the core, with unchanged material densities. The reactivity variations due to an increase of +1% of the density of four materials (fuel, sodium, steel, and absorber) were obtained by a series of first order perturbation calculations based on unchanged

microscopic cross-sections compared to the reference case. The Doppler constants were determined by a first order perturbation calculation in recalculating modified microscopic cross-sections due to temperature change (P_N to 180°C). The modelling for COREA was limited to the active core region (fuel and blankets) and the core was represented in the form of parallel channels.

The core was assumed to be at the end of the equilibrium cycle (EOEC) and in full power operation at the time of a transient. In general, the situation at EOEC is the most unfavourable due to the increase of the sodium density reactivity with irradiation. The fuel pellet was assumed to be tied to the cladding inner surface. This assumption is generally valid after a few at.% of irradiation. The thermal exchange coefficient between the fuel and the cladding was considered constant and assumed to be equal across the core (5 000 W/m² °C for oxide fuel element and 7 000 W/m² °C for nitride fuel element [11]).

The static neutronic characteristics of the two cores are presented in Table 2.

Table 2. Neutronic characteristics of the cores

Cores	Oxide	Nitride
Reactivity loss/cycle (\$)	6.7	4.0
Control rods worth (\$)	23.8	20.8
Maximum burn-up rate (GWd/t _{HM})	175	155
Maximum damage rate (dpaNRT)	199	197
Doppler constant at EOEC (\$)		
Fissile	-1.9	-1.79
Fertile	-0.4	-0.3
Sodium void effect at EOEC (\$)	+6.57	+6.00
Neutron life time (sec)	4.30×10^{-7}	3.33×10^{-7}
Breeding gain at BOL		
Core	-0.36	-0.27
Blankets	+0.28	+0.28
Breeding gain at EOEC		
Core	-0.20	-0.15
Blanket	+0.241	+0.22
$\beta_{\text{eff}} (\Delta k/k)$	0.003402	0.003516

The transient analysis, which predicts the variation of the fuel, cladding and sodium temperatures as a function of time during normal operation or transients, was performed by using the nuclear plant dynamics code DYN2B [12]. This transient code was validated by specific experiments conducted for Phénix and Super-Phénix, and allows for the calculations of the fuel, cladding and sodium temperatures in every channel (group of subassemblies) and axial segment. In all cases, average values are calculated, whereas the safety criteria imply preferably maximum values. In DYN2B, these maximum values are reconstructed posteriorly with hot channel factors. For our comparative study, we have simply used the factors of the Super-Phénix core for both cores. The primary pump of the EFR type reactor was assumed to coastdown with a flow-halving time of 10 seconds.

With regard to the core modelling for DYN2B, the assemblies of similar power and flow rates were grouped, and for every grouping, only an average pin was modelled, using a two-dimensional (RZ) spatial representation. The reactivity was obtained from the detailed reactivity balance equation established with thermal feedback coefficients and the temperature variations in the core. The point reactor kinetics model was used.

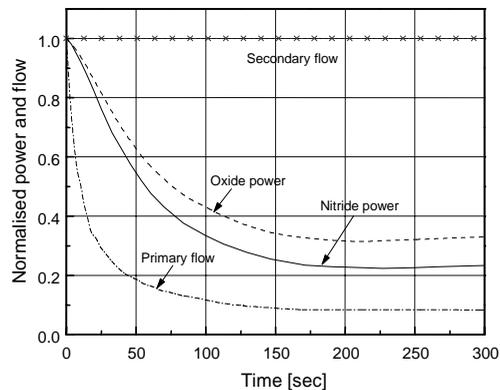
Transient analysis and discussions

Unprotected loss-of-flow (ULOF)

This transient is assumed to be initiated by the trip of primary pumps but the secondary pumps are assumed to be running at rated condition.

The power and core flow rate during the transient are shown in Figure 1. In this transient, the main effect is the sodium overheating associated with the positive sodium density reactivity. The sodium temperature increase degrades the cooling capability of the fuel, and thus the fuel is overheated leading to a negative Doppler feedback reactivity effect. After a few seconds, a negative reactivity due to the expansion of control rod drivelines intervenes and the net reactivity becomes negative. The power level falls and the fuel temperature decreases. The Doppler effect plays an unfavourable role when it begins to add a positive reactivity.

Figure 1. Power and flow variations during the ULOF

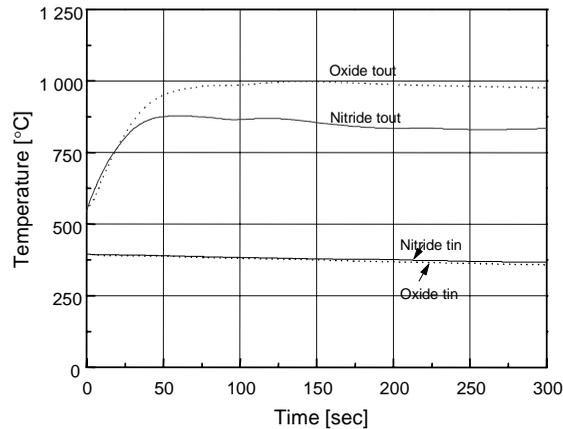


Finally, the positive (sodium density and Doppler) and the negative (expansion of the control rod drivelines) feedback effects equilibrate so that the net reactivity becomes zero. The reactor reaches an equilibrium state with a reduced power level and a cooling by natural convection.

Similar phenomena occur for both of the cores. However, the different magnitudes of feedback effects result in different safety margins during the transient. For the oxide core, the large positive sodium density feedback effect and the large negative Doppler constant play an unfavourable role: when the sodium temperature reaches its maximum value (140 seconds), these two effects add 210 pcm and 139 pcm of positive reactivity, respectively. The sodium reaches its boiling temperature at its maximum value (see Figure 2).

The nitride core exhibits more favourable behaviour even if its positive sodium density effect is as important as in the oxide core. In fact, a particular feature of the nitride fuel is the presence of a

Figure 2. Sodium temperature variations during the ULOF



Doppler constant equivalent to that of the oxide, but to be *cold* in the nominal regime. The latter characteristic, combined with the fact that the formula of the Doppler effect takes part in the initial fuel temperature, explains that the same increase of temperature has a much larger effect on reactivity in the case of the nitride. At the same time, the same decrease of power results in a reduced temperature variation which adds a much reduced positive Doppler reactivity, i.e. the nitride core is less opposed to the power reduction. These two situations are successively met in the ULOF transient.

At the beginning of the transient, the fuel temperature increases as the sodium temperature rises, and the nitride fuel can have a larger negative feedback reactivity. Afterwards, the fuel temperature decreases as a result of the power reduction. The magnitude of the decrease in fuel temperature is smaller in the case of nitride (140°C compared to 265°C in the case of oxide), and there is a reduced effect on reactivity, which is favourable at this moment. The gain on the sodium outlet temperature compared to the oxide fuelled core is about 125°C.

Unprotected transient over power (UTOP)

This transient represents an inadvertent withdrawal of all control rods with a speed of 1 mm/second (maximum value permitted for EFR). The ramp reactivity insertion rate is +8 pcm/second, which is anticipated for EFR at 10 cm of the control rod insertion from the top of the core.

The positive reactivity insertion results in an immediate increase in the power and fuel temperature as shown in Figures 3 and 4. The Doppler feedback effect gives a negative reactivity, which compensates the inserted reactivity. On the whole, the net reactivity attains a certain level (~ +10 pcm) and is maintained during the transient. Because of this constant positive reactivity, there is a linear increase in power and temperature. Calculation was terminated when fuel began to melt.

For the oxide core, fuel melting begins at 78 seconds. At this moment, the inserted reactivity is +625 pcm and the power increase rate is 70 MWt/second. On the other hand, the nitride core shows a more rapid increase in power (126 MWt/second). This is due to the thermal properties of the nitride fuel which provide a smaller Doppler feedback effect: for the same variation of power, less fuel temperature variation is associated and there is less Doppler feedback in spite of the equivalent Doppler constant. The total inserted reactivity at the moment of nitride fuel melting is +824 pcm

Figure 3. Power variations during the UTOP

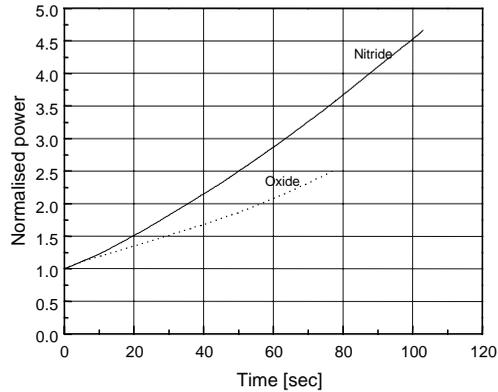
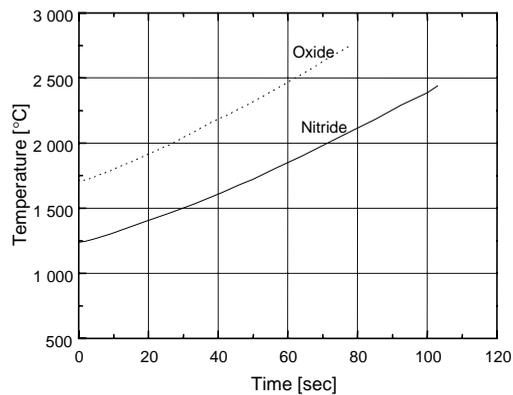


Figure 4. Peak fuel temperature variations during the UTOP



compared with +625 pcm for the oxide fuel, which allows for a delayed melting (103 seconds) compared with the oxide fuel (78 seconds), but the risk of sodium boiling and cladding failure is encountered earlier at about 80 seconds.

Indeed, in the case of nitride, the sodium and cladding temperatures are higher than for oxide. So for the nitride fuelled core, we can estimate a higher risk of sodium boiling for an unprotected transient but we have to consider this characteristic as having a more effective detection potential.

Unprotected fast transient over power (UFTOP)

This case has been chosen as an envelope case of eventual situations which could occur as a result of a gas passage in the core. The step reactivity insertion of 400 pcm was assumed to be maintained for 150 ms. This reactivity insertion rate corresponds to approximately 1/5 portion of sodium voiding in the EFR type core.

This transient is characterised by a fast insertion of reactivity, which is slightly higher than 1\$. The reactivity introduced by the initiators classified as design basis accidents is relatively low (<1\$). Thus this transient was studied only to compare dynamic behaviour of the two cores which have been subjected to a fast and large insertion of reactivity. The core will become prompt critical before

feedback effects play a role to decrease the total reactivity under 1\$. Therefore, as shown in Figure 5, the power increases very rapidly to a maximum value and then decreases, corresponding to a total positive reactivity of about 10 pcm which results for the most part from a large negative Doppler feedback reactivity. The average fuel temperatures are presented in Figure 6.

Figure 5. Power vibrations during the UFTOP

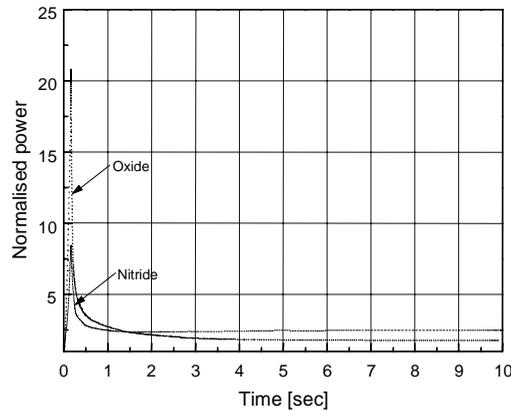
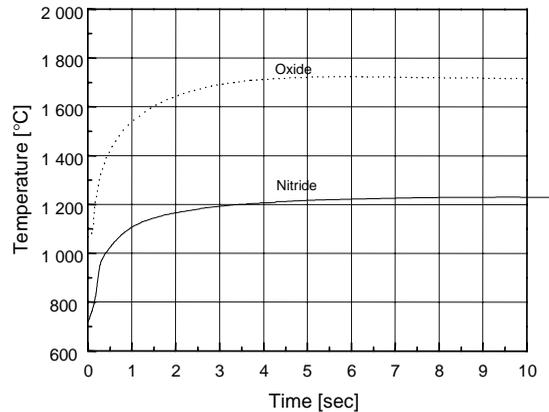


Figure 6. Average fuel temperature variations during the UFTOP



During the power excursion, the fuel becomes almost adiabatic. In this case, the Doppler effect is the only feedback effect which can intervene effectively. The fuel expansion is not effective because of the tied gap between the fuel and the cladding. The other parameters which intervene are the core kinetics parameters, i.e. β_{eff} and neutron lifetime, and the thermal inertia of the fuel.

For the oxide core, the power at peak is 21 times the nominal power at 0.16 seconds. The peak fuel temperature is 2 466°C at 6.4 seconds, and thus the margin to melting is particularly small.

For the nitride core, the power at peak is equivalent to 8.5 times the nominal power at 0.15 seconds. Compared with the oxide case, this small increase comes principally from the large Doppler feedback at the beginning of the transient. In fact, the thermal inertia of the nitride fuel is 15% less than that of the oxide fuel. This results in a more rapid fuel temperature increase in the nitride fuel than in the oxide fuel.

Moreover, effect on reactivity for a given variation of the fuel temperature is sensibly large for the nitride fuel because of the low operation temperature of the nitride fuel. These two effects are favourable for the Doppler feedback effect.

During the thermal equilibrium, high thermal conductivity of the nitride fuel tends to inverse the situation: the power is stabilised at 9 120 MWt for the nitride core compared with 7 000 MWt for the oxide core. In spite of a large power increase, the fuel temperature increase from the initial state of the nitride core is smaller than that from the oxide core (510°C compared with 656°C). Therefore, the nitride core is favourable in two respects: more efficient Doppler feedback effect during the transient; and a substantial margin to melting after the transient.

Summary and conclusions

In an accidental situation caused by an ULOF or an UTOP event, the two effects are systematically favourable: axial fuel expansion and differential expansion of the control rod drivelines. In general, the first effect is small (~ -0.3 pcm/°C), and the second is larger but delayed and variable because it depends on control rod insertion position which changes during the cycle. The Doppler effect, essentially favourable effect in UTOPs, is favourable at the beginning of ULOF but becomes unfavourable afterwards. Finally, the sodium density effect is always unfavourable, particularly in ULOFs. In general, if a core has a smaller sodium density effect and larger Doppler and expansion effects, the risk will be reduced.

In addition to feedback effects, the thermal properties of fuel play an important role in the dynamic behaviour of cores. This aspect is well illustrated in this comparative study on oxide/nitride fuels. The differences shown during the transients are directly linked to physical characteristics of the nitride fuel, even though the sodium void reactivity and the Doppler constant of the nitride core are very similar to those of the oxide fuelled reference core.

In the ULOF event, the nitride core is characterised by a larger Doppler feedback effect when it is necessary (at the beginning of the transient) and a smaller Doppler effect when it is unfavourable (after 10 seconds). Thus the margin to sodium boiling is larger for the nitride core.

For the slow UTOP event, the nitride fuel temperature increase is sensibly less pronounced than oxide fuel due to the high thermal conductivity. This means on the one hand a less effective Doppler feedback effect resulting in a rapid power increase, but on the other hand a high margin to melting. Consequently, we have in the design basis accidents (DBAs) domain margins to melting sensibly increased compared to the oxide fuelled core whereas in natural behaviour these margins can only compensate the reduced Doppler feedback effect permitting a situation equivalent to that of the oxide case (identical maximum admissible reactivities). Nevertheless, the cladding and the nitride fuel reach the domain where risk of simultaneous cladding failure and fuel dissociation exist. But dissociation phenomena require more experimental studies.

For the fast UTOP event, the fuel temperature increase becomes almost adiabatic. In consequence, a high thermal conductivity and a good fuel/cladding thermal exchange coefficient cannot intervene over a short time interval. On the other hand, a relatively low initial fuel temperature of nitride makes the Doppler feedback effect more efficient, and the slightly reduced thermal inertia of the nitride fuel permits its rapid heating, which reinforces the Doppler feedback effect.

In conclusion, the high thermal conductivity of nitride is an advantage in the DBA domain, in particular for early detection, in so far as the heat produced in the fuel is rapidly transferred to the system (cladding, sodium). The fuel temperature increase is small and margins to melting are gained. Use of a nitride fuel can enhance the safety of liquid-metal cooled reactor cores compared to oxide fuelled cores, but knowledge of thermal characteristics at high burn-up is needed to renew this transient analysis. Fuel dissociation phenomena also have to be studied experimentally as potential safety criteria, to be considered mainly in the context of special UTOP events.

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THERMAL DECOMPOSITION BEHAVIOUR OF UN AND (U_{0.8}Pu_{0.2})N

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Abstract

Thermal decomposition behaviour of UN and (U_{0.8}Pu_{0.2})N pellets was evaluated to better understand their preparation and their irradiation behaviour. Pellets of UN and (U_{0.8}Pu_{0.2})N were prepared by carbothermic reduction. The tests were carried out in three different atmospheres: He, Ar/5%H₂ and a vacuum of 2 Pa. The thermal decomposition occurs continuously over the surface of the pellet, spreading along grain boundaries near the pellet surface, and leading to a decrease in the pellet volume. The initial thermal decomposition temperature of UN and (U_{0.8}Pu_{0.2})N pellets is at least 1 800°C in He, Ar/5%H₂ and a vacuum of 2 Pa atmosphere. In the higher temperature range, thermal decomposition occurred with vaporisation of metal.

Introduction

Uranium and plutonium mixed nitride, which has NaCl structure, has been anticipated to be a high performance nuclear fuel for fast reactors because of its high fissile density, high thermal conductivity and excellent compatibility with coolant sodium as compared with those of oxide fuels [1-3]. It was reported that the nitride decomposed into liquid metal and nitrogen at high temperatures [3-6]. An understanding of the thermal decomposition behaviour of a nitride fuel is important for the improvement of preparation techniques and irradiation behaviour, because the thermal decomposition causes composition changes in nitride pellets.

The thermal decomposition behaviour of a nitride fuel was reviewed by Hj. Matzke [3,4]. The melting point of UN was measured in a nitrogen atmosphere of 2.5 kbar and was estimated to be 2 850°C. However, it has been reported that uranium nitride decomposed into U metal and nitrogen at a temperature lower than the melting point, at about 2 000°C, in a reduced nitrogen pressure atmosphere of 10 Pa. In the case of He bond nitride fuels, thermal stability in a He atmosphere is important to evaluate the operating temperature limits. The thermal decomposition of (U_{0.8}Pu_{0.2})N pellets were tested in N₂ and He atmospheres by K. Richer and C. Sari [5]. They reported that (U_{0.8}Pu_{0.2})N pellets were stable up to 1 723°C and decomposed with vaporisation of Pu in temperatures above 1 723°C. Measurements of vaporisation were also carried out to evaluate the stability of nitrides at high temperature. It was reported that vapour pressure of U, Pu and N₂ of (U_{0.8}Pu_{0.2})N pellets were measured in a vacuum [6-9], and the vapour pressure of Pu is much higher than that of U or N₂. The vaporisation of Pu causes composition changes due to loss of Pu at the pellet surface.

In general the sintering of nitride pellets has been carried out in Ar or a Ar/H₂ mixture [1-5]. It is important in the preparation to understand the influence of sintering condition, such as temperature and atmospheric nitrogen pressure on the density and composition of the pellets. The aim of this work was to evaluate nitride pellet stability under the different atmospheres (He, Ar/5%H₂ and vacuum) through observation of the thermal decomposition.

Experimental

Tablets of UN and (U_{0.8}Pu_{0.2})N were synthesised by carbothermic reduction of the oxide powders, UO₂ and U_{0.5}Pu_{0.5}O₂, with graphite in a N₂/5%H₂ mixed gas stream at 1 500°C for 8 hr. Fabrication equipment is installed in high purity Ar gas glove boxes (O₂ < 10 ppm, H₂O < 10 ppm) These two kinds of nitride tablets were ground for 48 hr by a ball mill. After grinding, nitride powder was palletised, and then sintered at 1 650°C for 3 hr and at 1 750°C for 3 hr in flowing Ar gas. The sintered pellets achieved theoretical density of 92.7%, and contained oxygen up to 6 000 ppm and carbon up to 2 000 ppm.

The thermal decomposition tests were carried out to estimate the initial decomposition temperature and to investigate the decomposition behaviour by two types of tests: Type I and Type II. The test conditions are shown in Table 1. In the Type I test, a tungsten crucible with UN pellet inside was heated from 1 500°C to 2 500°C with a tungsten-mesh resistance furnace at a constant heating rate of 10°C/min in the three kinds of atmosphere: He, Ar/5%H₂ and vacuum. The pellet surface and tungsten crucible temperatures were measured with two optical pyrometers, and the change of the surface morphology of the pellet was observed by a digital camera during the decomposition test.

Table 1. List of samples and the test condition for the thermal decomposition test

Sample no.	Composition	Test type	Atmosphere	Heating condition
1	UN	Type I	He	1 500-2 500°C, 10°C/min
2	UN	Type I	Ar-5% H_2	1 500-2 500°C, 10°C/min
3	UN	Type I	Vacuum of 2 Pa	1 500-2 500°C, 10°C/min
4	UN	Type I	He	1 500-2 500°C, 10°C/min + keeping for 110 min at 2 500°C
5	($U_{0.8}Pu_{0.2}$)N	Type II	Flowing He gas of 0.4 l/min	1 200-3 000°C, step 200°C
6	UN	Type II	Flowing He gas of 0.4 l/min	1 200-3 000°C, step 200°C

The crucible temperature was measured with an accuracy of $\pm 50^\circ\text{C}$ by a pyrometer sighted on a black-body well at the side of the crucible. Both He gas and Ar/5% H_2 gas containing 2 ppm and 20 ppm nitrogen were used in the test. The pressure inside the furnace was also measured during the test in a vacuum atmosphere. After each test, the pellet was cooled naturally to ambient conditions in about 15 minutes, and was analysed by optical microscopy and X-ray diffraction.

To estimate the initial decomposition temperature, a portion of UN and ($U_{0.8}Pu_{0.2}$)N pellets weighing about 30 mg were tested under Type II conditions. The sample was put into a graphite crucible which was heated up to 3 000°C with an impulse furnace in a flowing He gas of 0.4 l/min. The flowing He gas was directed to the sample, and nitrogen released from the sample was measured by a detector (TCD) which measured the thermal conductivity of the flowing gas.

Results and discussion

The results of the decomposition test for UN pellets in He atmosphere are shown in Figure 1. The temperature of the crucible increased up to 2 500°C at a constant rate of 10°C/min, but the temperature of the UN pellet was different from that of the crucible. The temperature of the crucible was measured more accurately than that of the pellet, because it was measured at a black-body well. To observe the temperature changes in detail, the variation of ΔT , which is the difference between the temperature of the crucible and the pellet, is shown in Figure 2 as a function of the crucible temperature. The change of ΔT ought to be monotonous, if the state of a pellet surface has no change. However, the temperature of the pellet surface began to change irregularly at about 2 250°C in He and Ar/5% H_2 , and 1 800°C in vacuum. Ceramographs of UN pellets in Type I tests are shown in Figure 3. The metallic phases were observed on the surface and on the grain boundaries near the surface of the pellets tested in each atmosphere. The metallic phase was identified as FCC-uranium metal by X-ray diffraction analysis. The field of the metallic phase was observed to spread along the grain boundaries in the UN pellet kept at 2 500°C for 110 min in He atmosphere and spherical grains of UN were observed in the field of the metallic phase (see Figure 3(d)). A mass of uranium metal was confirmed on the surface of a pellet tested in a vacuum atmosphere. Ceramographs of UN pellets tested in He atmosphere are shown in Figure 4. The decrease of the cross-sectional area and the deformation of the UN pellet shape kept at 2 500°C for 110 min were observed, and metallic phases existed in the deformed area. These experimental results indicate that the irregular changes of ΔT shown in Figure 2 are caused by thermal decomposition, and that thermal decomposition of UN pellet occurs continuously on the pellet surface and spreads along the grain boundaries near the pellet surface.

Figure 1. Crucible and UN pellet surface temperatures during the decomposition test for sample No. 1 in He atmosphere

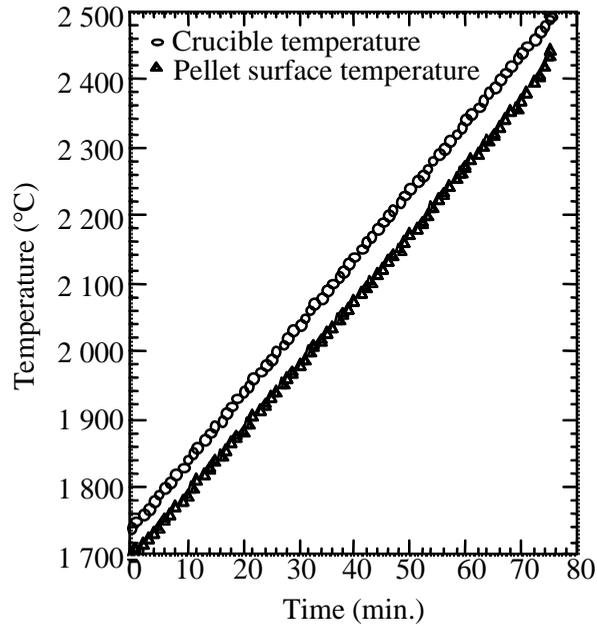


Figure 2. The crucible/pellet temperature difference (ΔT) on crucible temperature

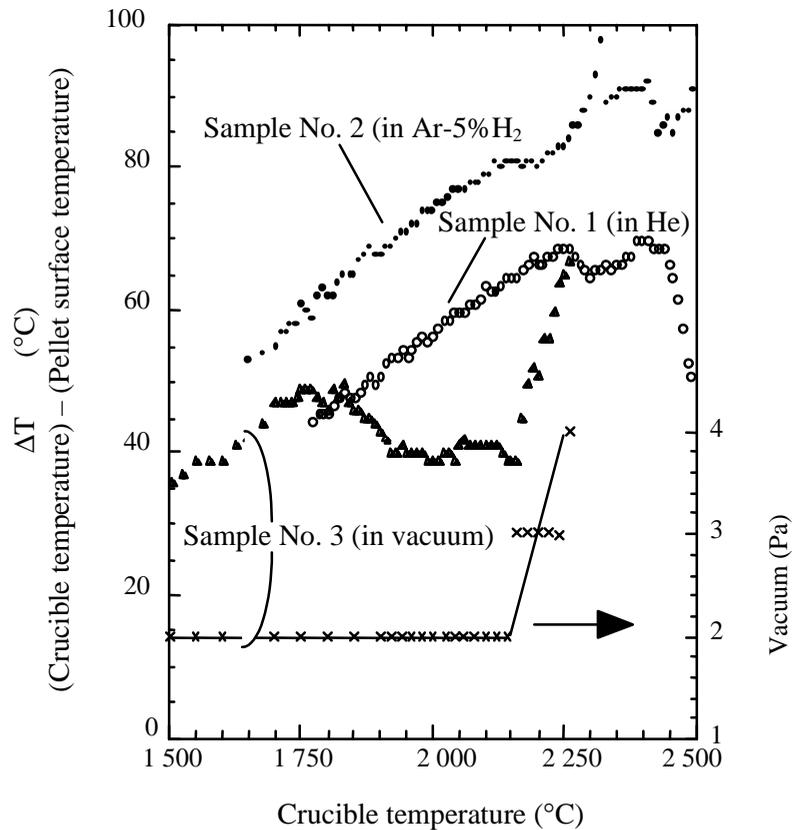


Figure 3. Typical microstructures of UN pellets after the decomposition test.
(a) Sample No. 1 tested in He. (b) Sample No. 2 tested in Ar/5% H_2 . (c) Sample No. 3 tested in vacuum. (d) Sample No. 4 heated at 2 500°C for 110 min in He.

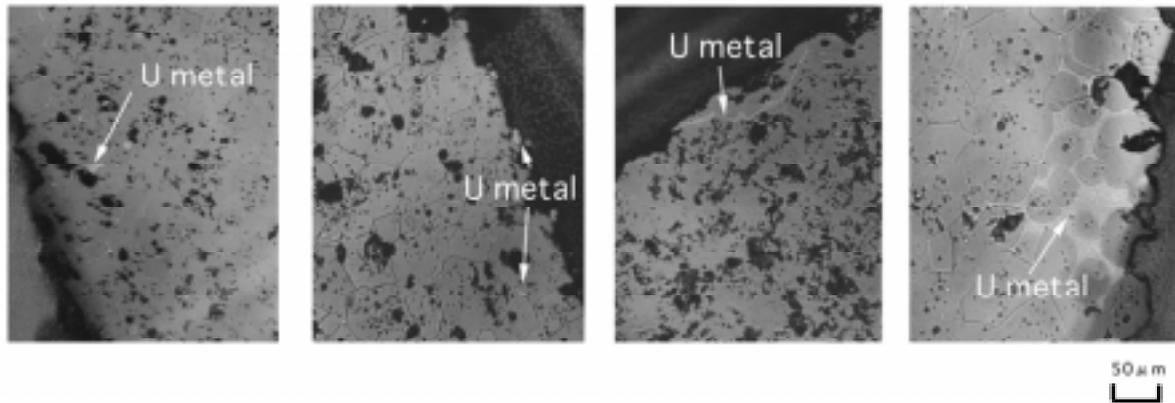
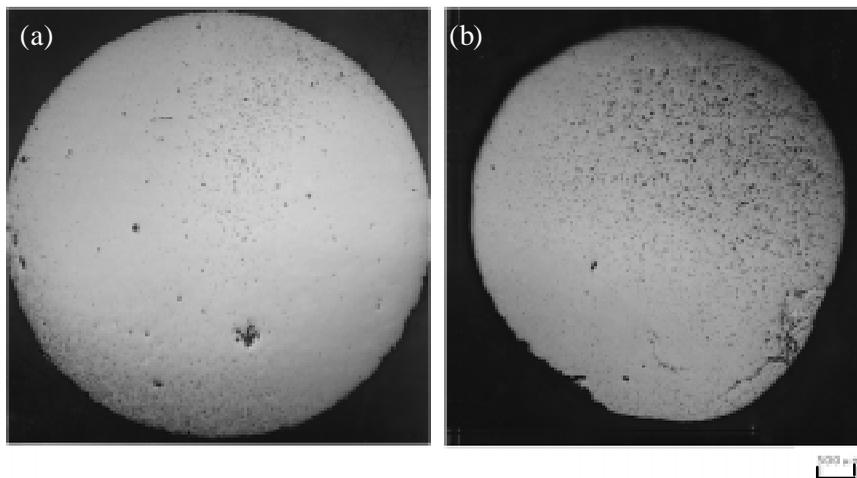
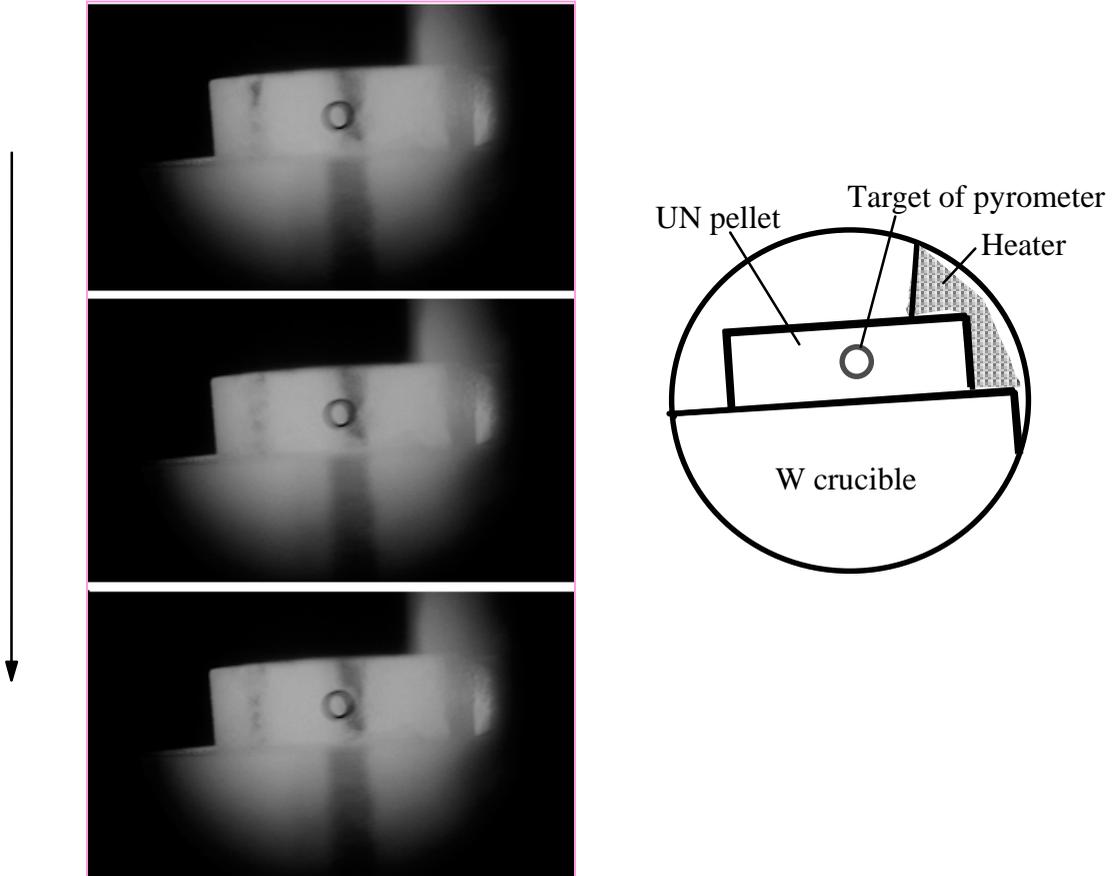


Figure 4. Typical ceramographs of UN pellets tested in He atmosphere.
(a) Sample No. 1 heated up to 2 500 °C and cooled to room temperature for about 15 min.
(b) Sample No. 4 heated at 2 500°C for 110 min.



For the test in vacuum atmosphere, the pressure in the furnace inside was also measured (see Figure 2). The pressure was initially 2 Pa, and the corresponding initial temperature was 1 500°C, and it began to increase at about 2 150°C. Additionally, vaporisation from the pellet surface was observed above 2 150°C by a camera. The sequential photographs of the UN pellet during the test are shown in Figure 5. The change in the surface morphology of the pellet is observed. The test in a vacuum atmosphere was stopped at about 2 300°C, because the crucible reacted with uranium metal. It was previously reported that the partial pressure of U metal was smaller than that of nitrogen with UN [7,8]. This report suggests that vaporisation of U metal occurs at a temperature higher than the initial decomposition temperature. In this work, the results of the test in a vacuum atmosphere indicate that thermal decomposition of UN begins slowly at 1 800°C and occurs rapidly with the vaporisation of uranium metal above 2 150°C.

**Figure 5. Sequential photographs of UN pellet during the test in a vacuum.
The change of surface morphology was observed above 2 150°C.**



The results of Type II tests are shown in Figure 6. Samples of UN and $(U_{0.8}Pu_{0.2})N$ were heated up to 3 000°C in a graphite crucible. The temperature of samples increased from 1 200°C to 3 000°C with a step of 200°C, and the release of nitrogen gas from both samples started at 1 800-2 000°C. The release of nitrogen gas indicates that thermal decomposition of the nitrides occurs, and that it initiates between 1 800 and 2 000°C for pellets of UN and $(U_{0.8}Pu_{0.2})N$. Significant releases of nitrogen gas were observed at around 2 400°C for $(U_{0.8}Pu_{0.2})N$ and 2 600°C for UN. The vaporisation of uranium metal was observed at the temperature of more than 2 150°C in the Type I test in a vacuum. The significant nitrogen release from UN and $(U_{0.8}Pu_{0.2})N$ in the Type II test may also be caused by thermal decomposition with vaporisation of uranium or plutonium metal.

Previous research indicates that the initial thermal decomposition temperature depends on the atmospheric nitrogen partial pressure [3,4]. The results from the current work are shown in Figure 7, which presents nitrogen partial pressure as a function of temperature. The nitrogen pressure in atmosphere in this work was calculated from impurities by MALT II, which is a materials-oriented little thermodynamic database [10]. The three different atmospheres (He, Ar/5% H_2 and vacuum) used in this work have the nitrogen pressure of about $\log(N_2)MPa = -5.7$. The starting temperatures of decomposition reported by Hj. Matzke are also shown in Figure 7 [3,4]. According to the report, the

Figure 6. Results of nitrogen release from the samples

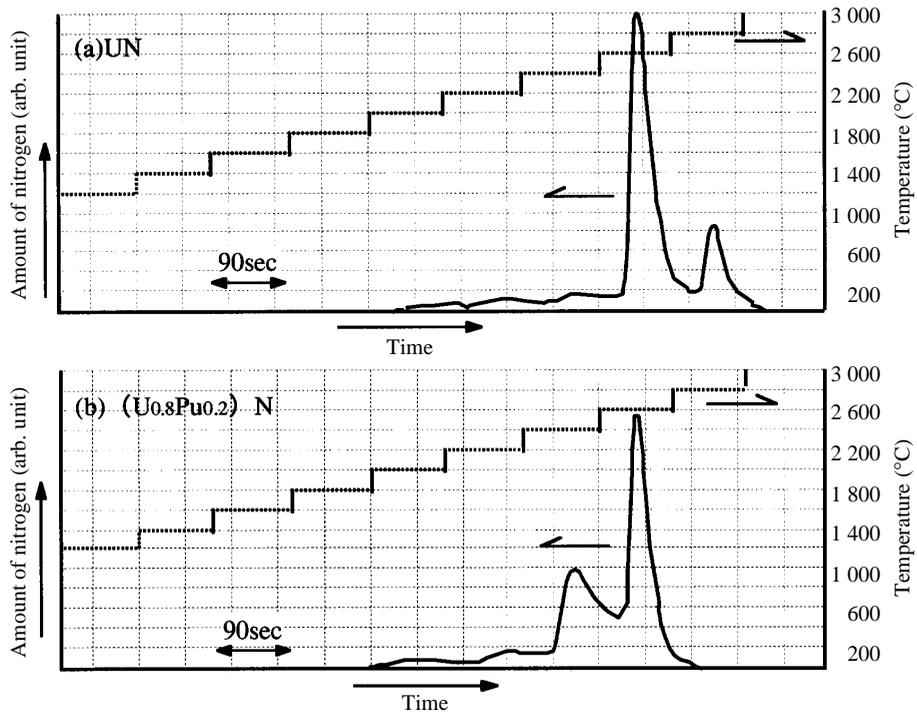
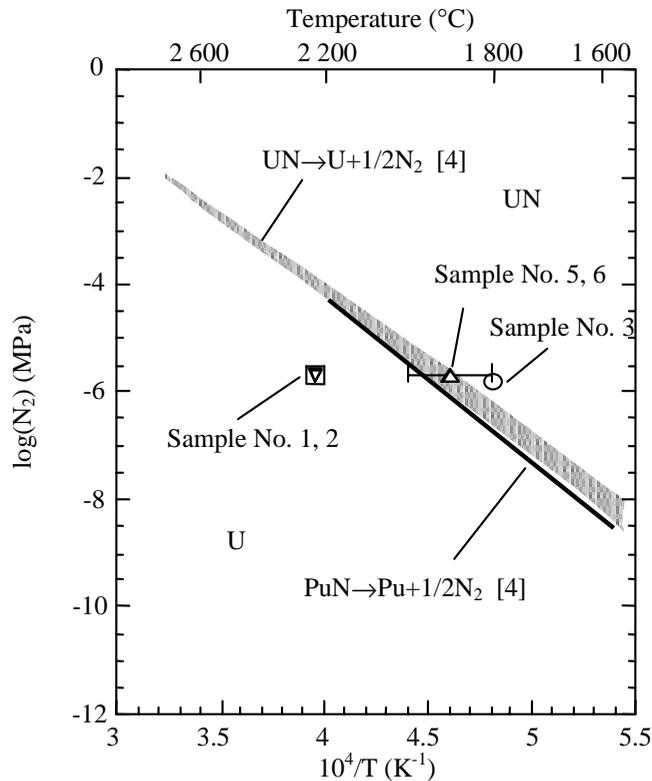


Figure 7. The decomposition pressure of mononitride in equilibrium with liquid



decomposition starting temperature is about 1 900°C as the nitrogen pressure is $\log(N_2)\text{MPa} = -5.7$ which is equivalent to that observed in this work. However, the decomposition temperatures in the results of Type I in He and Ar/5% H_2 atmospheres are higher than those reported in previous work. The initial decomposition at about 1 800°C in Type I tests in He and Ar/5% H_2 might be difficult to detect, because their decomposition rate is slower than in a vacuum and flowing gas due to the change of nitrogen pressure at the pellet surface during the test [9]. In the Type II test the thermal decomposition temperatures of both UN and $(U_{0.8}Pu_{0.2})N$ samples are estimated to be in the range of 1 800-2 000°C. It has also been reported that the decomposition temperature of UN is nearly equal to that of $(U_{0.8}Pu_{0.2})N$ [4,6]. K. Richer and C. Sari [5] reported that the thermal decomposition of $(U_{0.8}Pu_{0.2})N$ pellets started at 1 723°C in N_2 and He atmosphere. The differences in the initial decomposition temperature among various works may be caused by impurities like oxygen in the nitride samples or error of the estimation of the nitrogen pressure in the atmosphere. The thermal decomposition of UN and $(U_{0.8}Pu_{0.2})N$ pellets begins a minimum temperature of 1 800°C in He, Ar/5% H_2 and vacuum of 2 Pa atmosphere, because the three kinds of atmosphere have almost the same nitrogen pressure.

Conclusion

Thermal decomposition testing of UN pellets was carried out in He, Ar/5% H_2 and vacuum atmospheres, and thermal decomposition was observed at about 2 250°C in He and Ar/5% H_2 , and at about 2 150°C in a vacuum. In another decomposition test, the initial decomposition temperatures of UN and $(U_{0.8}Pu_{0.2})N$ were estimated to be in the range of 1 800-2 000°C by detection of the released nitrogen. The three atmospheres have almost the same nitrogen pressure according to the calculation results of MALT II. The thermal decomposition of UN and $(U_{0.8}Pu_{0.2})N$ pellets begins at a minimum of 1 800°C in He, Ar/5% H_2 and vacuum of 2 Pa atmospheres. In higher temperature ranges the thermal decomposition occurs with vaporisation of metal. The existence of uranium metal was observed at the pellet surface and in the grain boundaries near the surface by a ceramography and X-ray diffraction analysis. The thermal decomposition occurred continuously over the surface of the pellet and spread along grain boundaries near the pellet surface. Moreover the thermal decomposition caused a decrease in the cross-sectional area and deformation of the pellet's shape. From the results of this work, UN and $(U_{0.8}Pu_{0.2})N$ pellets should be used in an operating temperature of less than 1 800°C during their preparation and their irradiation to avoid thermal decomposition.

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REVIEW OF RUSSIAN MOLTEN SALT REACTOR TECHNOLOGY STUDIES

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Abstract

The objective of this paper is to discuss the potential advantages of innovative reactor technology based on nuclear fuel in the form of molten fluoride salts and integrated with non-aqueous processing methods as applied to the problem of Pu inventories reduction and radwaste elimination. The aims of this contribution are:

- To review the results of works performed in Russia relevant to the development of molten salt reactor technology.
- To evaluate the importance of remaining uncertainties and to identify the additional work needed for molten salt nuclear fuel concept implementation.

The restrictions on length allow to present only a general overview of these results. Fuel technology, container materials, radiation chemistry of the fuel, components, fuel clean-up and heat transfer with emphasis on experiments will be of priority. The main finding of experimental studies is that neither physical nor technical obstacles to the implementation MSR and related fuel cycle technologies in future nuclear power systems have been observed.

Introduction

Though the molten salt nuclear fuel concept has been proven through the successful operation experience of the MSRE experimental reactor [1], this approach has not been implemented in industry. Interest in the molten salt nuclear fuel technology still exists, however, and continues to grow, especially as applied to the problems of deep burning of Pu and transmutation of long-lived radwaste. It is thus to be expected that MSR technology has a prospect to be implemented in future nuclear power systems and, without a doubt, it is worth further study.

In Russia the molten salt reactor (MSR) programme was started in the second half of the 70s. The Russian Research Centre – “Kurchatov Institute” (at that time known as the Kurchatov Atomic Energy Institute) was a basic organisation under the supervision of which a collaboration of specialised institutions was formed and functioned. In this case, much of the development of Russian MSR studies was under the leadership of Prof. Vladimir M. Novikov.

A reduction of activity occurred after 1986 due to the Chernobyl accident, along with a general stagnation of nuclear power and nuclear industry. Toward the end of the 80s, however, there was an increase of conceptual studies as a result of the interest in creating inherently safe reactors of a new generation [2]. In 1994-1996 a feasibility study of molten salt fuel as applied to Pu burning and long-lived radwaste transmutation in accelerator-based system was supported by the International Science and Technology Centre (ISTC). This study was known as Project #17. A new concept concerning MSR fuelled by Pu and MA is currently the subject of a feasibility study supported by MINATOM.

The objectives of the Russian MSR programme were as follows:

- To identify potential advantages and intrinsic pitfalls of MSR concepts.
- To search for a means of providing a maximum of advantages and a minimum of vulnerability.
- To explore the feasibility of technical implementation of the main MSR design features.

The MSR study was organised around the following issues:

- Exploration of possible use and niches for MSR concepts.
- Reactor physics and reactor safety.
- Container materials for fuel and coolant salts.
- Heat transfer and hydraulics of fuel and coolant.
- Handling and circulation of fuel and coolant salts.
- Process and radiochemical bench tests of model installations.
- Radiation chemistry of molten fluoride fuel salt.

The first two issues constitute theoretical studies, while the others address both theoretical and experimental questions. An extensive review of MSR technology at the Russian Research Centre – “Kurchatov Institute” (RRC-KI) through 1989 is given in Refs. [3-7]).

The next sections will discuss MSR studies based on this publication and those that appeared afterwards.

Outline of the future fuel cycle [8]

For nuclear power, short- and long-term development of the problems concerning nuclides flows and neutron balance are of the greatest priority. Some neutron balance estimations demonstrate the necessity for multi-component structures which will permit long-term and wide scale nuclear power development. This concept would make it possible to decrease the total loss of nuclides to waste, to minimise its equilibrium quantities, to provide required neutron balance, and would allow for the possibility of eliminating the negative consequences with regard to nuclear power functioning.

In addition to traditional solid fuel thermal and fast reactor designs, in future multi-component nuclear power systems one can consider processes concerned with the final uses of the recycled fuels as fluorides, in the molten salt reactors:

- Discharged fuel goes through reprocessing plant, actinides are extracted from waste;
- Uranium after isotope correction and plutonium and, possibly part of Np are recycled as a fuel in thermal and fast solid fuel reactors. Purex reprocessing of spent MOX fuel becomes more difficult, as the content in PuO_2 increases.
- Part of the plutonium together with minor actinides (MA) and, if needed, some FPs are incinerated in a molten salt-burner reactor (MS-BR). Stable and short-lived FPs are removed from the burner reactor by the separation systems. These problems can be solved by chemical and physical methods in batch or on-line modes.
- Modified Purex, electrochemical and/or gas-fluoride volatility processes should be investigated for recycling options considered above.
- The stable and short-lived FPs are directed to an interim storage facility from where they are sent to final disposal.

The advantages of the MSR as a burner reactor follow not only from a possibility for its effective combination with the dry technique of fuel processing/separation, which potentially has low cost and produces a small volume of wastes, but also from its capability to use fuel of different nuclide compositions.

The MSRs have the flexibility to utilise different fissile fuel in continuous operation with no special modification of the core, as was demonstrated during MSRE operation for $^{233,235}\text{U}$ and Pu. Also, a good neutron balance allows to use MAs largely contaminated by lanthanides. This simplifies the lanthanide/MA separation process and could drastically decrease the loss of MAs to wastes. MS-BRs further require a minimum of special fuel preparation and can tolerate denaturing and dilution of the fuel.

The strategy of nuclear power development proposed in [8] is not an alternative to fossil-fired power, but represents its essential supplement. Different types of reactors do not compete with, but complement each other allowing the best manifestation of the advantages of every type of plant design.

Container materials studies [3-6]

An important part of Russian MSR programme dealt with the investigation of the container materials. General requirements for the materials were formulated as follows:

- They must be easily produced, i.e. easily subjected to treatment by pressure, cutting rolling, well weldable, and in such a way so as to provide the possibility of fabricating complex components of the reactor structure.
- Pressure on walls in a typical MSR fuel circuit is less than 2 MPa. Thus, guided by this limiting value and taking a ten-fold safety factor, the candidate material must have a limiting value for lasting strength bigger than 20 MPa (30 years of operation at temperature 750°C).
- Metal constructions in MSR are outside the peak neutron flux and neutron flunk is estimated as 10^{20} and 5×10^{21} n/cm² for fast (> 0.5 MeV) and thermal neutrons. It was demanded that the limiting value for bending relative deformation after gaining a full neutron fluency would be no less than 4%. (The initial value of this parameter, for a non-irradiated Hastelloy-N amounts to 10% at 650°C.)
- To keep the main sequences of corrosion at an acceptable level (heat exchanger wall thickness, molten salt content, mass transfer) the depth of corrosion should not exceed 10 µm/year and metal should not be subject to a local corrosion (pitting or intergranular cracking).

The development of domestic structural material for MSR was substantiated by available experience accumulated in the ORNL MSR programme. The alloy HN80MT was chosen as a base. Its composition (in wt.%) is Ni (base), Cr (6.9), C (0.02), Ti (1.6), Mo (12.2), Nb (2.6). Corrosion resistance of materials was studied using two methods. The first is the method of capsule static isothermal test of reference specimens in various molten-salt mixtures. Also, flibe, flinak and sodium fluoborate eutectic salts were circulated for thousands of hours in natural and forced convection loops constructed of stainless steel and nickel based alloy to obtain data on corrosion, mass transfer and material compatibility. The tabulation of the operating parameters of these loops is presented in Table 1.

Table 1. Thermal and forced convection loop experiments involving molten salts at RRC-KI

Loop	Type*	Salt composition, % mol	Container material	Maximum temperature, °C	ΔT, °C	Time, h
F1	I	71.7LiF-16BeF ₂ - 12ThF ₄ -0.3UF ₄	HN80MT	750	70	1 000
F2	I	71.7LiF-16BeF ₂ -12ThF ₄ -0.3UF ₄	HN80MTY	750	70	1 000
C3	L	92NaBF ₄ - 8NaF	12H18N10T	630	100	1 000
C4	I		EP-164			
C5	I		HN80MT			
Solaris	L	46.1LiF-11.9NaF-42KF	12H18N10T	620	20	3 500
M1	I	66LiF-33.9BeF ₂ -0.1UF ₄	12H18N10T	630	100	500
KURS2	I	66LiF-33.9BeF ₂ -0.1UF ₄	12H18N10T	750	250	750

* I – integral, L – loop arrangements

Not only normal, but also high oxidation conditions were present in the loop (e.g. moisture). The last circumstance made it possible to estimate the limiting corrosion characteristics of materials.

The information accumulated by ORNL on corrosion processes during thermal and forced convection loop tests allowed to compare them with data on our experiments carried out under similar conditions. Such a comparison enables in a number of cases to see new mechanisms of corrosion behaviours and predict an operability potential of presented materials. The development and optimisation of the HN80MT alloy was envisaged to be performed in two directions:

- Improvement of the alloy resistance to a selective chromium corrosion.
- Increase of the alloy resistance to tellurium intergranular corrosion and cracking.

About 70 differently alloyed specimens of the HN80MT were tested. Among alloying elements were W, Nb, Re, V, Al and Cu. The main finding is that alloying by aluminium at a decrease of titanium up to 0.5% revealed a significant improvement in both the corrosion and mechanical properties of the alloy. The chromium corrosion and intergranular corrosion have reached the minimum value at Al content in the alloy ~ 2.5%. The irradiation effect on a corrosion activity of fuels was also studied. It was shown that at least up to the power density 10 W/cm^3 in fuel composition LiF-BeF₂-ThF₄-UF₄, there is no radiation induced corrosion.

Then a radiation study of 13 alloy modifications was carried out. Specimens (in a nitrogen atmosphere) were exposed to the reactor neutron field up to the fluency of $3 \times 10^{20} \text{ n/cm}^2$. Experimental results of alloy mechanical properties at temperatures of 20, 400 and 650°C for non-irradiated and irradiated specimens permits to rest only four modifications. The alloys modified by Ti, Al and V have shown the best post-radiation properties.

Finally, corrosion under the stressed condition was studied. It is known that tensile strain promotes an opening of intergrain boundaries and thus boosts intergranular corrosion and create prerequisites for an intergranular cracking. The studies did not reveal any dependence of intergranular corrosion on the stress up to the value 240 MPa, that is 0.8 of a tensile yield of the material and five times higher than typical stresses in MSR designs.

The results of combined investigations of mechanical, corrosion and radiation properties of various alloys of HN80MT permitted to suggest the Ti- and Al-modified alloy as an optimum structural material for MSR. This alloy, known as HN80MTY (or EK-50), has the following composition (in wt.%): Ni (base), Fe (1.5), Al (0.8-1.2), Ti (0.5-1), Mo (11-12), Cr (5-7), P (0.015), Mn (0.5), Si (<0.15), C (<0.04). The comparison of our corrosion data with that obtained at ORNL for Hastelloy indicates that corrosion resistance of HN80MTY is higher and its maximum working temperature could be up to 750°C. Still, the weldability of the alloy requires improvement.

Radiation stability of fluoride salts [3]

In-reactor loop tests regarding the radiation stability of fluoride molten-salt fuels were carried out. The radiolysis products (first of all, F₂) were detected in the gas phase over the melts. It was determined that:

- The radiolytic evolution of fluorine from molten fluoride compositions when irradiating in a nuclear reactor is insignificant.

- The measured values of radiation chemical yield $G(F_2)$ (the number of F_2 molecules, evolving per 100 eV of absorbed energy) are in the range of 10^3 - 10^6 , therefore the fluoride fluid fuel may be attributed to the category of radiation resistant materials in the temperature range up to 1 200°C.
- The frozen fuel salt at 50°C has a $G(F_2)$ value of about 10^2 , which is much better as compared to water, which is used as the primary coolant in LWRs.

The high radiation resistance of molten fluorides allow them to be recommended as the fuel for high temperature reactors.

The heat transfer experience [2,4]

The design of MSR requires detailed information on transport properties of the proposed fuel and coolant fluoride melts.

At RRC-KI heat transfer studies with different fuel and coolant salts (Pr modules range from 2 to 35) flowing in forced and natural convection loops were made in a wide range of parameters typical for MSR designs.

Forced convection runs covered a Reynolds module range from 5 000 to 20 000 and a heat flux q_s range up to 10^2 kW/m². The criterion dependencies generalising the heat transfer data were obtained for natural convection heat exchangers at separate or joint actions of surface and volume (nuclear) heat sources in the fluid up to $q_s = 30$ kW/m² and $q_v = 7$ MW/m³.

No evidence of influence of corrosion and irradiation products on heat transfer was found (at temperatures up to 750°C, neutron fluences up to $2 \cdot 10^{20}$ cm⁻² and concentration of Cr, Fe, Ni metal impurities less than 10^{-1} by mass.).

The conclusion is that the use of accurate physical property data with correlations for normal fluids ($Pr > 1$) is adequate for heat transfer for design with fluoride salts with respect to forced and natural convection.

Pump experience [2,5]

At RRC-KI we know how to make reliable short-shaft centrifugal pumps for molten salt test loops, having built and operated pumps with capacities up to 2 000 rpm.

Although it may take some years to produce the larger pumps for the demonstration of a MSR prototype, the problems are well understood.

We do not have experience with the long shaft pump configuration for molten salt systems. However, in Russia it has been used in lead-bismuth and sodium systems developed for the fast reactor programme.

We expect that results of the long shaft development programme for liquid metal pumps will have a direct application to salt pumps. At the same time, we understand that the requirements for lead-bismuth (or sodium) and salt pumps are sufficiently different that in other important respects they are unique.

Actinide /lanthanide separations [9-12]

The MS-BR would manage the noble gas and the semi-noble and noble metal in the same manner as other MSR concepts [1]. Also, in order for an MS-BR to operate efficiently, means must be provided for the removal of certain fission products, i.e. rare earth elements of atomic numbers 57-62 of the lanthanide series which generally account for more than 50% of the parasitic neutronic absorption. Also, TRUF₃ solubility is maximum in pure fuel salt and decreases with the addition of trivalent rare earths. The MS-BR fuelled by Pu and MAs must contain a system that provides for removal of TRU from the fuel salt and their reintroduction in the purified solvent.

Important parameters for reductive extraction and electrorefining are electrochemical potential/free energy of formation $\Delta G_f^\circ(T)$ of substances and separation factors of elements between molten salt and liquid metal θ . These separation factors for different elements with the same valency (for example actinides and lanthanides) could be given by:

$$\ln\theta = \{\Delta G_f^\circ(Ln, T) - \Delta G_f^\circ(An, T)\}/(RT)$$

where T is temperature (K), and R is the molar gas constant ($J \cdot mol^{-1} \cdot K^{-1}$).

Thermodynamic considerations point to the need for more efforts in separating Am and Cm from Ce and La in a chloride system as compared to a fluoride system (see Table 2). The calculated separation factors of various elements in fluoride and chloride systems suggest that fluorides possess a better separation ability between actinide and lanthanide groups, but are less effective within groups.

Table 2. Partial molar Gibbs energy of some trivalent halides, $\Delta G_f^\circ(T)$, kcal/mol

Nuclide	Chloride	Fluoride
	($T = 800$ K)	($T = 1\ 000$ K)
U	164	305
Np	174	310
Pu	187	320
Am	195-200	325
Ce	205	345
La	209	348

Design and construction of engineering scale tests of several parts of chemical processing MSR concepts in the US and Russia were underway when national programmes were discontinued. In these programmes the main technology development efforts were directed toward reductive extraction. A rare earth removal unit based on a Bi-Li reductive extraction flowsheet developed by ORNL for an LiF-BeF₂ solvent system could provide negligible losses of TRU ($\approx 10^{-4}$) through the use of several counter current stages.

For fluoride based systems, other methods of TRU/lanthanides separation – along with reductive extraction – could be considered. Particularly of interest is electrochemical deposition of elements as metals on liquid or solid electrodes in electrochemical cells, filled with fuel salt and bismuth or lead. It is found that the recovery of 99.9% of TRU could be attained by a combination of electrorefining and counter current extraction in the LiCl-KCl/Bi system by using Li as reductant while satisfying the separation ratio between TRU and rare earths of over 1.0.

The applicability of the given method for fluoride based systems is defined by the fact that the fuel salt at a temperature of 800-1100 K is a good electrolyte, the conductivity of which is comparable to the conductivity of liquid electrolytes. In a fluoride system based on LiF-BeF₂ or LiF-NaF-KF the standard potentials for salt components are higher than those for actinide and lanthanide trifluorides.

Because the difference in potentials of deposition of Am and Ce in fluoride systems reaches 0.2 V (see Table 2), it is possible to assert that at electrolysis of the fuel salt there will be a selective separation of these elements. The degree of separation will depend on the value of an over-voltage on electrodes. As the experience of industrial production of a number of alkaline and alkaline-earth elements by electrolysis shows, at a difference of potentials of deposition of elements more than 0.1 V, a degree of their separation is more than 100 (at nominal currents about 1 A/cm²). Thus it is possible to expect a degree of separation of actinides and lanthanides of more than 100, and at several stages of electrochemical clean-up the contents of actinides in lanthanides can be reduced up to 10⁴ times. Further investigations would be necessary to investigate the feasibility of the fluoride system and the efficiency of the electrowinning and electrorefining processes.

ISTC supported R&D

Some steps on the development of MS-BR technology were performed in 1994-1996 in the framework of the ISTC-supported Project #17. The experimental and engineering work programme of this project included the following main items [13]:

- Chemical compatibility of a PuF₃ fuelled system with a container material in ampoule conditions [14-15].
- Preliminary design of natural and forced convection loops for the study of problems associated with handling and circulation of molten salt fuelled PuF₃.
- Conceptual study of the blanket for Pu burning.
- Proposal on MSR-DEMO, a facility construction at Chelyabinsk-70 [15].

Experiments have demonstrated that plutonium addition did not worsen the corrosion situation. This means that plutonium deep incineration in an MSR system seems to be quite possible.

Prime R&D needs for new MS-BR concept development and implementation

Though the molten salt nuclear fuel concept has a solid background, its specific application for deep burning of Pu and radwaste transmutation requires additional studies:

- Chemistry involving Pu, Am, Cm and Np as fuel components requires confirmation of TRUF₃ solubility and needs further study, especially in testing loops, for Redox potential, corrosion and FP behaviour control.
- Development and testing of some critical plant components (e.g. steam generator, heat exchanger, fuel pumps).
- Molten salt fuel processing technique is an important area for development.

To solve some of these as well as other essential issues, Russian institutes, including VNIITF (Chelyabinsk-70), the RRC-Kurchatov Institute, the Institute of Chemical Technology (Moscow) and the Institute of High Temperature Electrochemistry (Ekaterinburg), have submitted Project #698, *Experimental mock-up of the molten salt loop of accelerator-based facility for transmutation of radwaste and conversion of the military plutonium*, to the ISTC.

The following issues for fluoride based system as applied to new burner concepts could receive experimental attention in the framework of ISTC Project #698:

- Creation of a suitable database on MSR technology to prevent the ultimate loss of years of valuable experience emerging from previous studies.
- Reliable handling and circulation of PuF₃ fuelled salt and testing critical components.
- Verification of the individual and collective solubilities of trivalent actinide/lanthanides fluorides in different salts.
- Compatibility of alloys and graphite with fuel salt containing actinides.
- Monitor and control of the Redox potential of molten salt fuel.
- Heat transfer and hydraulics study of fuel.
- Tritium control technology.
- Verification of the fuel processing techniques.
- Draft design of a molten salt burner including its cost estimations, safety and non-proliferation assessments and other general issues.

Conclusion

The MSR programme performed in Russia covered a wide range of theoretical and experimental studies. As a result, a unique experience has been accumulated which could be applied for implementation of molten salt technology for effective Pu burning and radwaste transmutation. Application of this experience and reorientation of the facilities of Russian nuclear weapons centre Chelyabinsk-70 to this peaceful function are objectives of the ISTC Project #698. This project is currently under consideration and has received a project development grant for the project workplan improvement and co-ordination of efforts with foreign collaborators. One of the aims of this report is to attract the attention of potential foreign partners to the ISTC Project #698 and to invite them to take part in a collaboration in terms of the project.

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CONCEPT OF NUCLEAR INCINERATION OF PWR SPENT FUEL IN A TRANSMUTER WITH LIQUID FUEL

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Abstract

The principle drawbacks of any kind of solid nuclear fuel are listed and briefly analysed in the first part of the paper. One of the primary results of the analyses performed shows that the solid fuel concept, which was to a certain degree advantageous in the first periods of nuclear reactor development and operation, has guided the branch of utilisation of atomic nucleus energy to a dead end (not having been able to solve principle problems of the corresponding fuel cycle in an acceptable way). On the basis of this, the liquid fuel concept and its benefits are introduced and briefly described in the following parts of the paper, allowing to develop new reactor systems for nuclear incineration of spent fuel from conventional reactors and a new clean source of energy.

As one of the first realistic attempts to utilise the advantages of liquid fuel, the reactor/blanket system with molten fluoride salts in the simultaneous role of fuel and coolant, as incorporated in the accelerator-driven transmutation technology (ADTT) being proposed in [1], has been proposed for deeper theoretical and experimental studies in [2]. A preliminary design concept for the experimental LA-0 assembly that is currently in preparation in the Czech Republic is also briefly introduced [3].

Introduction

There are principle drawbacks to any kind of solid nuclear fuel listed and analysed in the first part of this paper. One of the primary results of the analyses performed shows that the solid fuel concept, which was to certain degree advantageous in the first periods of nuclear reactor development and operation, has guided this branch of atomic nuclear energy utilisation to a dead end (not having been able to solve principle problems of the corresponding fuel cycle in an acceptable way). On the basis of this, the liquid fuel concept and its benefits are introduced and briefly described in the following part of the paper.

As one of the first realistic attempts to utilise the advantages of liquid fuel, the reactor/blanket system with molten fluoride salts in the role of fuel and coolant simultaneously, as incorporated in the accelerator-driven transmutation technology (ADTT) being proposed in [1], has been studied both theoretically and experimentally. A preliminary design concept of an experimental assembly LA-0 is briefly introduced in the following paragraph, and this project is currently under preparation in the Czech Republic.

Finally, another very promising concept [4,5] of a small low power ADTT system is introduced, characterised by a high level of safety and economical efficiency. This subcritical system with liquid fuel driven by a linear electron accelerator represents an additional element – nuclear incinerator – to the nuclear power complex (based upon thermal and partly even fast critical power reactors) making the whole complex acceptable and simultaneously giving an alternative and very highly acceptable nuclear source of energy and even other products (e.g. radionuclides, etc.). In conclusion, the overall survey of principal benefits, which may be expected by introducing liquid nuclear fuel in nuclear power and research reactor systems, is given and critically analysed. Other comparably important principles (e.g. the general subcriticality of reactor systems principle) are mentioned which, being applied in the near future, may form a basis for an absolutely new nuclear reactor concept and a new nuclear power era.

Solid nuclear fuel concept drawbacks

In spite of the fact that all of the following is well known, it seems to be worth evoking the circumstances of nuclear power at the end of the 20th century at the beginning of the search for new nuclear energy systems and fuel cycle options for the 21st century. Since the discovery of the reaction of atomic nucleus fission, the main goal of all efforts was to utilise it for energy generation. As one of the most important conditions for an efficient achievement of this goal self-sustainability of the fission chain reaction was demanded in an assembly containing fissionable nuclei of nuclear fuel without an external source of neutrons. If this condition was met, the assembly was defined as being critical. Let us note that it was by definition (theoretically) critical on prompt neutrons released, immediately, from fission reactions only. Very early, it was observed experimentally that the assembly reaching criticality is in fact very slightly subcritical on prompt neutrons and that there is a not a very strong natural source of delayed neutrons originated from radioactive decay of some of the fission products always added (which, fortunately, allowed easier control of the system).

At the early stages of nuclear development, reaching criticality was one of the most difficult tasks, and all effort and ideas were devoted to this aim. The reason for this was that there were only small amounts of fissionable materials available at that time in the form of the low (0.7%) content of ²³⁵U in natural uranium. Therefore, solid phase metallic uranium with the highest possible density was used in the form of blocks with a specifically defined size arranged in a heterogeneous lattice filled in

by a solid (graphite) or liquid (heavy water) moderator with a certain pitch determined by optimal neutronic conditions. This arrangement has remained fairly exclusive, and is used even in latter systems with fuels enriched by ^{235}U content up to much higher levels than the content of the natural composition of uranium. The reasons were of a different nature, however, the designs have mostly started from what became already an approved conventional principle – solid fuel blocks in a heterogeneous lattice – which has been kept even in the case of pure or high enriched fuel in a fast neutron system without moderator.

One of the consequences following the adoption of the solid fuel concept has been a type of control system which has been mostly applied for short term control of nuclear reactors – the concept of solid absorbers – and what is more, the concept of a negative neutron source (neutron poison) at all. This, and a number of other consequences, can be traced to start all from the initial tension in neutron economy when the principle of a self-sustaining fission chain reaction and consequently the concept of a critical reactor were adopted in nuclear reactor technology. They all begin to form a magic circle of convention in which short term and finally even long term operational behaviour of nuclear, namely power, reactors became imprisoned and limited in their ability to develop in a positive and broadly accepted manner. This thesis is explained in the following with some illustrative examples.

The adoption of the solid fuel concept leads to the principal necessity of keeping the fuel blocks at a certain position in the reactor core for a shorter or longer period of time. This in-core residential time is especially long in power systems where at least a quasi-continuous exchange of fuel would be very complicated and expensive. Therefore, the following very inconvenient consequence arises: the whole time the block of solid fuel remains at a certain position in the reactor core, there are fission fragments and by neutron capture induced radionuclides (let us call them altogether products) being accumulated in the volume of the fuel block. There are several secondary consequences caused by this fact which contribute to the formation of the above-mentioned “magic circle”:

- 1) Reactivity margin for a short term as well as long term negative influence of the increasingly accumulated products has to be applied which has to be compensated by another artificial negative source of neutrons. It has, in principle, another consequence in the greater amount of fuel being present in the core than is really necessary for the demanded power, resulting in the generation of more products, including actinides, in the system.
- 2) The original fuel is finally so heavily poisoned by the products that it can no longer maintain the self-sustained fission chain reaction, and further operation of the reactor under original conditions is impossible. A principle change in the operation and structure of the reactor becomes unavoidable, which means an outage and exchange of at least a part of the original fuel charge.
- 3) The most controversial problem – what to do with spent solid fuel – arises and a vicious circle has been closed or a solid fuel concept “trap” snapped.

The briefly described solid fuel concept thus displays its most important and sensitive drawbacks: 1) continuous accumulation of products during the whole residential time of fuel blocks in the core, 2) following necessity to stop the operation, discharge spent fuel and store it for a necessary period of time (in order of magnitude of years until it reaches a desirably low level of radioactivity) in a specific storage, 3) the last and the most difficult drawback is the need of an optimal decision regarding the final destiny of spent fuel.

Up to now, the only two possible solutions were developed either to reprocess (chemically) the spent fuel and to prepare the next generation of solid fuel (which implies basically the same class of drawbacks) or to dispose it in a depository of a corresponding quality (which sometimes is called a repository because a possible reuse of the disposed product is supposed). In the former case, mostly chemical methods and processes are applied. In the latter, a lot of branches are involved; however, nearly all of them are of a classical (non-nuclear) nature. The only nuclear process employed is natural radioactive decay.

This fact contains one very controversial principle or, better yet, a violation of a basic principle which can be described as follows: The energy generation in nuclear reactors utilises enforced nuclear processes which are simultaneously producing products or nuclear waste (including secondary raw materials, e.g. actinides). The treatment of the products needs to apply an adequate technology on an adequate scale. Up to now, this principle has not been applied and fulfilled in systems developed and designed for spent solid fuel management. There is an adequate technology which only one can utilise nuclear processes and which can transfer the high level and long-lived radionuclides towards short-lived or even stable nuclides – the transmutation technology performed in a suitable nuclear reactor device and combined with an at least quasi-continuous separation of certain components of its core or reprocessing of the reactor fuel as to avoid the consequent induction of radioactivity by neutron irradiation of stable and short-lived nuclides. One of the principle concepts allowing the exploration of such a technology on an industrial scale is the concept of liquid nuclear fuel.

The liquid fuel concept for neutron source-driven transmutation technology

Molten fluoride salt fuel for neutron source-driven transmutation technology

The concept of a neutron source-driven subcritical blanket for incineration of nuclear waste has become well known over the past several years [1] (see Figure 1). Let us briefly recall the main features of the most recently developed version of this concept and let us show a part of a proposed research programme to approve its ability for an efficient realisation on an industrial scale.

The fuel material is in the form of the fluoride salt AcF_4 which is dissolved in a molten salt carrier whose composition is a mixture of ${}^7\text{LiF}$ and ${}^9\text{BeF}_2$. The carrier's melting point and operating temperature are about 500°C and 650°C , respectively. The molten salt flows over either the outside of a close-packed set of cylindrical high-purity graphite blocks or inside cylindrical channels coaxial situated in, e.g. hexagonal graphite blocks (Figures 2 and 3).

The experimental research system designed by the author pre-conceptually in [3], should be developed and realised in the Nuclear Research Institute Řež plc in the Czech Republic. The final purpose of the system would be an experimental testing of a given type of transmuter reactor/blanket core neutronics and possibly also other physical and technological characteristics and properties including time behaviour. For the very first stage, the following scheme can be applied which will allow reaching the first results very cheaply and relatively soon. There can be an elementary, though sufficiently representative, sample of the investigated reactor blanket lattice inserted into an existing experimental reactor core serving as a driver and the basic set of its characteristics can be experimentally measured and verified. A suitable experimental reactor could be, e.g. the LR-0 experimental reactor (a full-scale VVER type core modelling zero power reactor operated at the Nuclear Research Institute Řež) or VR-1 (a training reactor operated at the Faculty of Nuclear Science and Physical Engineering of the Czech Technical University in Prague) which have been successfully operated for core analyses of thermal reactors since 1982 and 1990, respectively.

Low power ADTT system

Molten salt reactors (MSRs) with continuous control of nuclide composition almost do not require an initial reactivity margin. In such reactors, subcriticality may be reduced up to the minimum value β , where β is the effective delayed neutron fraction. However, with such a small subcriticality and in view of available uncertainties in nuclear data and nuclide concentrations, the difference between subcritical and critical MSR disappears to a great extent: in both cases nuclear safety is ensured by the large negative temperature reactivity effect. The deeper subcriticality is of course substantiated by the fact that under such conditions we exclude the necessity to control a reactor burner in a dynamic mode, that is a bit difficult and poorly known.

In this case, the accelerator-driven positive source performs only one of the usual functions – the function of a reactor control system without inertia, an alternative to (up to now usually used as reactor control organs) negative sources like, e.g. absorbers or decreasing of the dimensions of the system, etc. The high level parameter proton accelerator with all its disadvantages (e.g. like the length ~ 1 km, the investments \sim US\$ 1 billion, etc.) having been applied e.g., in the Los Alamos concept is not necessary more in the system and a low level parameter accelerator can be employed.

The blanket concept of a transmuter for PWR spent fuel incineration

There has been a convenient blanket concept for an efficient nuclear incineration of PWR spent fuel developed as a combination of the two ideas described in the paragraphs above. The concept is illustrated by the Figure 4. Where two zones are indicated, one under-moderated (dark fuel) and thus better equipped for actinides burning and the second well-moderated (light fuel channels in graphite blocks) and thus more convenient for fission products incineration.

Experience in fluorine technology and application of molten fluoride liquid fuel in ADTT

There has been a technological process worked-out for the separation of uranium and plutonium from the spent fuel by a dry (fluorine) method in the NRI Řež in close collaboration with the Russian Kurchatov Institute in Moscow during the 80s. The whole process was upgraded to a pilot plant scale with a capacity of 1-3 kg of processed fuel/hour. Part of the technological equipment was built and verified on an inactive scale at the NRI Řež. The whole technological process was then realised at the Institute of Atomic Reactors at Dimitrovgrad, Russia (Figure 5). All equipment including fittings, measuring instruments and accessories were built in the former Czechoslovakia, and the plutonium part of the pilot plant was built in the former USSR.

A certain experience has also been obtained on the uranium isotope separation through ultra-centrifugation and electrochemical processes.

The experience gained in the course of the research is going to be applied in developing fluoride based liquid fuel as well as fluoride chemistry separation processes for the use in accelerator driven transmutation technology (ADTT)

Conclusion

The analyses of spent fuel management from PWRs as well as all other nuclear reactors employing the solid fuel concept have showed the principal drawbacks of that concept, causing a

series of consequences leading to a crucial issue currently facing the use of nuclear nuclear – spent solid fuel with accumulated actinides and long-lived fission products (without regard to whether the open nuclear fuel cycle or a multiple reprocessing is applied). The necessity of an employment of nuclear processes and an adequate nuclear technology (nuclear incineration) in an efficient solution of that problem is definitely evident. The national R&D programme which was very briefly introduced in this paper is closely connected with the world-wide effort of research teams in leading countries of nuclear power such as France, USA, Russia and Japan, and forms a contribution to the common effort of finding a solution to this global issue. The overall co-operation in this field gives a real chance to make nuclear power acceptable as a clean source of energy for the 21st century.

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Figure 1. Principle scheme of a transmuter system

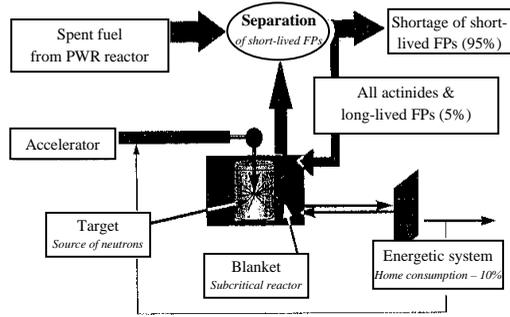
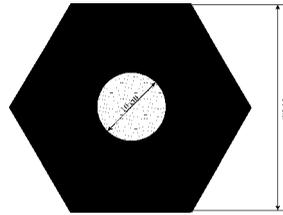
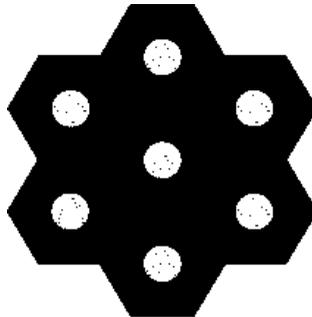


Figure 2. Single module of a transmuter blanket



graphite
 fuel (LiF 70% + BeF₂ 20% + UF₃ 10%)

Figure 3. The seven-module core of a transmuter blanket



graphite
 fuel (LiF 70% + BeF₂ 20% + UF₃ 10%)

Figure 4. The blanket concept of a transmuter

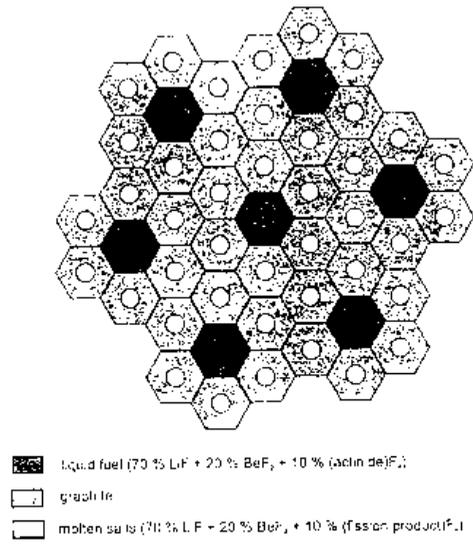


Figure 5. The fluorine technology reprocessing line Fregat



**STUDY OF AN ACCELERATOR DRIVEN SYSTEM OPERATING
IN EPITHERMAL SPECTRUM WITH A CONSTANT K_{eff}**

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Abstract

In any Pu burner operating with a thermal spectrum the fission/capture ratio is biased towards capture; therefore, instead of burning ²³⁹Pu and ²⁴¹Pu through fission, their conversion into ²⁴⁰Pu and ²⁴²Pu prevails.

One way to operate a system of this sort is to use very great proton currents; another is to add a neutron poison that is removed or burned as the concentration of the even Pu isotopes grows. An example of the removal of the neutron poison is the addition of ⁹⁹Tc, while Er may be considered an example of burnable poison. The ideal solution in this case, however, is provided by Th which initially works as a poison absorbing the Pu's excess of reactivity and subsequently, due to the production of ²³³U, compensates for the poisoning caused by the Pu's even isotopes.

There arises the problem of utilising the ²³³U that is obtained.

A possible solution of the problem is to consider an accelerator driven system (ADS) that operates at constant k_{eff} , adding continuously growing quantities of Pu from LWRs to compensate for the gradual poisoning due to the build-up of ²⁴⁰Pu and ²⁴²Pu.

Calculation method

The calculations have been carried out by means of MCNP4B with the cross-sections from ENDF/B-VI.

Calculational model

A system similar to LANL's ABS has been considered, though excluding graphite from the core, because preliminary calculations showed that the power density in a thermal system is 150 kW/kg above that of fast reactor fuel; therefore the graphite power density would be much greater than that in any reactor that employs graphite. Furthermore, while there is no more graphite in the core, it still remains in the reflector and consequently the ADS works with an epithermal spectrum and therefore with a fission/capture ratio better than that of a thermal system.

The proof that the system operates with an epithermal spectrum is given by the fact that small additions of Th cause a large decrease in the values of k_{eff} . Thus, the fuel considered in this case is 65.99 LiF - 33.99 BeF₂ - 0.02 PuF₄.

Evaluation of the initial inventory of Pu

Due to the operational mode of this ADS the inventory and the initial concentration of fissile material will be evaluated by means of the condition:

$$dk_{\text{eff}}/dc = 0$$

Thereby the system should operate at the concentration that corresponds to the maximum possible k_{eff} , that would never be excluded even in case of incidents.

To make this evaluation the course of the curve $k_{\text{eff}} = f(c)$ with the Pu concentration between 1.0 and 3.0 g/l has been determined and its maximum has been found corresponding to $c = 2.1$ g/l.

With the data the initial inventory of Pu has been evaluated to be 250.7 kg of Pu.

Validation of Rief-Takahashi formula

This formula is employed to determine the proton current corresponding to each calculation phase, assuming the ADS power constant; this verifies that in each phase the accelerator can satisfy the needs of the subcritical system.

The Rief-Takahashi formula is validated by comparing the power values corresponding to various fissile material concentrations evaluated employing the formula with those obtained with the Monte Carlo method. The fissile concentration values are those considered in the previous calculation phase. Furthermore the proton current has been considered to be 15 mA and the proton energy 1.0 GeV.

The Rief-Takahashi formula, from Ref. [1], relates the reactor power P , the particle beam current I_b , and yield of neutrons per proton in the target y :

$$P = y a I_b E_f k_{eff} / (\nu(1 - k_{eff})C)$$

In this equation a represents the importance of the target's position (a can be greater than 1 for the target in a centred position), E_f the energy released per fission, ν the number of neutrons per fission and C the charge of the proton.

Given for a certain material $I_b y$ (characteristics of the accelerator and of the spallation quality of the material) and E_f and ν (characteristics of the fission properties of the material) the knowledge of k_{eff} and of P allows the evaluation of the parameter a . The 15 mA 1 GeV proton current generates 23.5 neutron per proton (y).

Table 1 shows the influence of the Pu concentration on the parameter a ; in any case to perform the burn-up study of the present work a will be considered a constant calculated taking into account the average value of Table 1; with this value Table 2 was prepared, where it is possible to see that the maximum discrepancy between the power values calculated with Rief-Takahashi formula and Monte Carlo method is lower than 13.5%

Table 1. Validation of the Rief-Takahashi formula for liquid fuel epithermal ADS; influence of Pu concentration on the constant a

Pu concentration (g/litre)	a
1.0	1.0093
1.3	1.0251
1.8	1.1323
2.1	1.3132
2.4	1.2572
2.7	1.1357
3.0	1.1325
Average	1.1436

Table 2. Validation of the Rief-Takahashi formula for liquid fuel epithermal ADS

Pu concentration (%)	k_{eff}	Neutron power Monte Carlo calculations (MW)	Neutron power Rief-Takahashi formula calculations (MW)	Relative differences (%)
1.0	0.91180±0.0007	243.93	276.40	13.31
1.3	0.95547±0.0006	514.22	573.67	11.56
1.8	0.98035±0.0006	1320.71	1333.89	0.99
2.1	0.98248±0.0007	1721.61	1499.30	12.91
2.4	0.98021±0.0007	1455.82	1324.26	9.04
2.7	0.97689±0.0008	1122.39	1130.18	0.69
3.0	0.96866±0.0007	818.30	826.37	0.99

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INTERNATIONAL ACTIVITIES

Chair: K. Hesketh

THORIUM FUEL CYCLE OPTIONS FOR ADVANCED REACTORS: OVERVIEW OF IAEA ACTIVITIES

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Abstract

Within the framework of IAEA activities, the agency has maintained an interest in the thorium fuel cycle and its utilisation world-wide. Its periodic reviews have assessed the current status of this fuel cycle, its applications world-wide, its economic benefits and its perceived advantages *vis-à-vis* other nuclear fuel cycles. Activities on the use of thorium as nuclear fuel are carried out by the IAEA upon the recommendation of the International Working Groups on Advanced Nuclear Power Reactors. To implement these recommendations, several activities were planned, among them a series of two Advisory Group Meetings, Co-ordinated Research Projects (CRPs) and Technical Committee Meetings (TCMs). These Th-based nuclear fuel cycle activities are presented in this paper.

Introduction

Within the framework of IAEA activities, the IAEA has maintained an interest in global and national perspectives for developing advanced fuel cycle options, including the thorium-²³³U option. Th/U fuel cycles and thorium-fuelled reactor programmes have been implemented in several countries since the early 1950s. Reduction of radiotoxicity of discharged spent fuel has provided additional incentives for the utilisation of the thorium fuel cycle option. There are six major reasons for the introduction of thorium based nuclear fuel cycles:

- The enlargement of fissile resources by breeding ²³³U.
- Large thorium deposits in some countries, coupled with a lack of uranium deposits in those countries.
- Potential reduction in fuel cycle cost.
- Reduction in ²³⁵U enrichment requirements and so on.
- Safer reactor operation due to lower core excess reactivity requirements.
- Safer and more reliable operation ThO₂ fuel than UO₂ fuel at high burn-up due to its higher irradiation and corrosion resistance.

Several disadvantages in the thorium fuel cycle, as compared to the uranium fuel cycle, were recognised from the beginning of thorium related activities:

- The thorium-²³³U fuel cycle is more strongly radioactive than the uranium-plutonium cycle, and thus more care is needed during handling in fabrication.
- Nuclear reactions and decay schemes for producing fissile nuclides from thorium by neutron absorption are complicated, and spent fuel requires more time in water storage due to higher decay of residual heat.
- Potential difficulties can also be seen in downstream spent fuel reprocessing.

International studies

The International Fuel Cycle Evaluation (INFCE) Study, undertaken from 1978 to 1980, summarised thorium fuel cycle activities world-wide within the framework of its Working Group 8 (Advanced Fuel Cycle and Reactor Concepts). Nuclear fuel cycle concepts, technical status, economics and non-proliferation aspects were taken into consideration.

The study concluded that for all power reactor types, the technology for the utilisation of both Th-bearing fuels and the recycle of ²³³U had been less developed than for the U/Pu cycle. Major research and development was required in the areas of neutron physics, core calculation, remote technologies for fuel fabrication, reprocessing and waste treatment. Estimates indicated that over a range of cost parameters, Th cycles might be competitive with U/Pu cycles once the complete fuel cycle has been implemented.

The IAEA Symposium on Nuclear Fuel Cycle and Reactor Strategies: Adjusting to New Realities, convened by the IAEA in Vienna in June 1997, defined the “new realities” which necessitate a further review of thorium-based fuels in terms of their potential role in assisting the reduction of current plutonium stockpiles and attractive characteristics with regard to proliferation protection. In addition, the high conversion ratio of thorium fuel in thermal reactors offers added flexibility in fuel cycles during the start-up and introduction phase of LMFR.

Current status and motivation for thorium introduction

The present knowledge of world-wide thorium resources remains poor as a result of inadequate exploitation efforts. The total of reasonably assured resources (RAR) and estimated additional resources (EAR) of thorium world-wide are roughly about 6 022 000 t, while those of uranium amount to 4 488 000 t, costing less than or equal to \$130/kg U. Brazil, China, CIS, Egypt, India and the USA possess approximately 75% of world thorium resources.

In Germany and the USA, fuel fabrication technology for Th-based fuel has been developed under the HTR programme to a well-tested industrial-scale process. Indian fabrication technology for Th-based fuel has been adapted to the specific requirements of the HWR. Several tonnes of Th-based fuel has been manufactured for Switzerland’s LOTUS test facility, as well as for the test reactor/power reactors CIRUS and KAPS units 1 and 2. A similar fuel fabrication technology for Th-based fuel for HTRs and LWRs has been developed in Japan and has reached pilot plant stage. Recently new Th-based fuels, such as thorium hydride fuel alloyed with Ti and/or Zr, are being investigated. Irradiation tests of such Th-based fuels have been performed by Japan Material Test Reactor (JMTR). Russia has developed two fabrication technologies for WWERs and FBRs for Th-based fuel: $(\text{Th,U})\text{O}_2$ and $(\text{Th,U})\text{O}_2 + (\text{ZrC})$.

At present, the most important reasons for the utilisation of Th-based fuel cycles can be summarised as follows:

- Very low production of Pu and actinides.
- Higher fuel burn-ups than uranium-based fuel cycles.
- Possibility of acceleration of burning Pu (nuclear-grade or weapons-grade) without the need for recycle, i.e. faster reduction of the existing Pu stockpiles.
- Significantly lower production of toxic fuel waste or long-lived radiotoxic waste.
- Improved reactor operating safety due to the significantly higher melting point and better heat conductivity of the Th-based fuel.
- Proliferation resistant characteristics.
- Possibility to combine the Th-based fuel cycle with the accelerator driven systems and hybrid fusion driven systems for breeding ^{233}U and incinerating the long-lived radiotoxic isotopes.

Strategies for thorium use

Strategies for the use of Th-based fuel cycles will be selected depending on the required tasks and objectives, as well as on the underlying marginal economic advantages. These include a pure thorium scenario, a Th/U scenario, and a combination with Pu or U/Pu. Different options may be considered for each fuel cycle, such as the self-sustaining equilibrium thorium cycle (SSET) with reprocessing/ prefabrication; the high burn-up, high conversion ratio cycle (no reprocessing); or the once-through thorium cycle (OTT).

Regardless of the fuel cycle strategy chosen, Th-based fuel can be used in all proven reactor types including PWRs, WWERs, BWRs, HWRs, FBRs, HTRs/HTGRs; as well as for other reactor concepts such as molten salt reactor or aqueous homogenous suspension reactors. The type of Th-based fuel may also be selected and adapted in a very flexible way.

Role of the IAEA

The IAEA is a unique global inter-governmental organisation comprising 128 Member states that deals with a variety of nuclear energy matters. Aside from its function to provide safeguards and thus ensure safety, the IAEA has a promotional purpose “to encourage and assist research on, and development and practical application of, atomic energy for peaceful uses throughout the world.” In this context, the IAEA can provide a forum for the review and discussion of programmes, projects and topics in the development and introduction of nuclear energy, which also includes the thorium fuel cycle.

In the field of Th utilisation, such development may include:

- The encouragement and stimulation of thorium fuel cycle related research and development, including the exploration and evaluation of thorium resources in Member states and international organisations.
- Supporting international efforts to organise demonstrations and experiments on thorium fuel cycle feasibility.
- Continuing nuclear data research co-ordination activity by extending thorium fuel cycle research into reaction and energy areas.
- Performing assessments of possible synergetic nuclear energy systems based on different scenarios, such as LWR-FBR-ADS, LWR-ADS.

IAEA activities

Activities in the 1980s

Following the INFCE study, the IAEA continued Th fuel cycle related activities. At that time, major emphasis was placed on the analysis and collection of knowledge on conventional Th/U and Th/Pu fuel cycle technologies in fabrication, reprocessing and refabrication. These technologies were considered during the Advisory Group Meeting (AGM) on Advanced Fuel Technology Performance

in 1984 and 1989, and the Technical Committee Meeting (TCM) and Thorium Based Nuclear Fuel: Current Status and Perspectives in 1985. Evaluation of thorium resources and their respective technologies was considered at the IAEA TCMs in 1985, 1991 and 1995.

Technical committees in the 1990s

Proceeding the above-mentioned changes in tendencies towards the end of the 1980s and beginning of the 1990s, the IAEA placed major emphasis on waste and ecological aspects of Th/U and Th/Pu fuel cycles and Pu burning issues (Pu transmutation/disposition through Th/Pu fuel cycles) in different reactor system concepts, as well as new types of Th/Pu or Th/U fuels (cermets, cerchers). These aspects were examined at the TCM on Unconventional Options for Plutonium Disposition (1994) and the TCM on Fuel Technology and Performance of Non-Water Cooled Reactors (1994). Fuels and fuel cycles with reduced actinide generation, including Th fuel and fuel cycles, were the focus of the TCM held in 1995. Both power reactors of existing (HTRs, MSRs, FBRs, LWRs) and accelerator driven systems were considered for this purpose.

Technical Committee Meeting on “Fuel cycle options for LWRs and HWRs”, Victoria, Canada, 28 April to 3 May 1998

This TCM was jointly conducted within the framework 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 a forum to identify, review and exchange information on international developments in fuel cycle options for light water reactors and heavy water reactors.

The scope of the meeting included global and national fuel cycle strategies and development activities for high burn-up fuel, MOX fuel, slightly enriched uranium fuel, thorium fuel cycles, recycle uranium fuel, LWR and HWR synergistic fuel options such as DUPIC fuels, and the impact on uranium utilisation, fuel cycle and power generation costs, plant performance and waste management.

The TCM was attended by 47 people representing 12 countries and the OECD NEA. Thirty-five papers were presented, including contributions from Canada and India. With respect to thorium fuel cycles, these presentations indicated that the once-through thorium (OTT) cycle in heavy water reactors may provide a potential bridge between future thorium recycle options and current uranium based fuel cycles.

Co-ordinated Research Project (CPR) on “The potential of Th-based fuel cycles to constrain Pu and to reduce long-term waste toxicities”

This CRP commenced in 1996 with the participation of China, Germany, India, Israel, Japan, Republic of Korea, Netherlands, Russia and the USA with a purpose to examine the different fuel cycle options in which Pu can be recycled with Th to burn Pu, or replace Pu with materials that are less conducive to proliferation.

Two benchmark exercises were performed during the first stage. Benchmark 1 dealt with “Calculation of the isotopic composition, cross-sections and fluxes for a typical PWR-cell loaded with (Pu-Th)O₂ fuel, as a function of the fuel burn-up”. Benchmark 2 dealt with the same calculations,

but for PWR lattice only. Results of the benchmark exercises were discussed at the first Research Co-ordination Meeting (RCM) held in October 1996. Generally, the benchmark results of the participating countries were in agreement regarding the destruction of Pu and the isotopic compositions of the remaining Pu.

During the second stage of the CRP, three types of reactors – LWR, HTR and MSR – were investigated in terms of their potential to burn plutonium, each by one of three groups of countries. This procedure allowed a comparison of the computed results among several participants investigating the same type of reactors, and thus would also serve as an additional benchmark exercise. The results obtained during the second stage of the CRP: “Calculations on burning of plutonium in LWR, HTR and MRS”, were presented and analysed at the RCM held in Tel Aviv, Israel from 23-25 June 1998. Fuelling strategies on the basis of current and near-future technologies were the main topics of discussion. The third stage of the CRP comprised a Pu-generating unit combined with a corresponding Pu-burning unit that was selected to ensure complete burning of the generated plutonium. A complete Pu-flow cycle was considered. On the basis of these data, overall radiotoxicities were compared for the U-fuelled PWR with the combined system of PWRs and Pu-burners. The third RCM is scheduled for April 1999.

CRP on “The use of Th-based fuel cycle in accelerator driven systems (ADS) to incinerate Pu and to reduce long-term waste toxicities as part of the IAEA benchmark on ADS”

The purpose of this benchmark was to assess the uncertainties of the calculated neutronic parameters of a simple model of Th- or U-fuelled accelerator driven system in order to reach a consensus on the calculational methods and associated nuclear data. Participants identified a number of issues to be discussed in order to achieve a better understanding of the ADS.

The first RCM was held in March 1997. The purpose of this meeting was to review the results of the first stage of accelerator driven systems (ADS) neutronic benchmark and to elaborate the programme for the second stage of the benchmark exercise. Results of the RCM were reported to the IAEA TCM on Feasibility and Motivation for Hybrid Concepts for Nuclear Energy Generation and Transmutation held in Madrid, Spain from 17-19 September 1997.

Advisory group meetings

The AGM on Thorium Fuel Cycle Perspectives held in Vienna, Austria from 16-18 April 1997 was attended by 38 representatives and observers from 17 countries and three international organisations. The aim of the meeting was to assess the advantages and disadvantages of the thorium fuel cycle and to discuss the physics of thorium fuel. In subsequent discussions, the benefits of a once-through cycle (e.g. the Radkowsky seed-blanket concept for PWRs) were highlighted, in addition to the Indian programme on thorium recycle. The meeting concluded that a re-examination of this fuel cycle is necessary so that its advantages, problems and possible solutions can be realistically assessed in current contexts. International co-operation in making an assessment of the thorium cycle was recommended.

The AGM on Thorium Fuel Utilisation: Options and Trends, was held in Vienna from 28-30 September 1998 to discuss technological aspects of thorium fuel utilisation and other issues. The meeting was attended by representatives and observers from 18 nations and three international organisations. Papers presented and statements of delegates indicated that interest in investigations

into Th fuel is growing in the IAEA Member states. A recommendation was made concerning the usefulness of improving and elaborating more precise estimations in the future relating to the development of the Th fuel cycle application and the corresponding technology development and employment in nuclear power programmes. Participants appreciated the co-ordinated effort of the IAEA in the field of Th fuel cycle and the results of the Co-ordinated Research Project (CRP) on the Potential of Th-based Fuel Cycles to Constrain Pu and to Reduce Long-Term Waste Toxicities. Particular interest in co-operation was expressed by a number of other Member states not yet involved in this project.

The AGM recommended that the IAEA conduct a new Co-ordinated Research Project (CRP) on Thorium Fuel Cycle Options assessment for three to five years in late 1999. Interest was also shown in the development of a benchmark programme including unit cell calculations, critical benchmarks, fuel assemblies, reactor simulations and fuel cycle calculations. The IAEA was requested to develop and maintain a “library”/database of all available information on relevant thorium fuel cycles.

Conclusions

The primary reason for the revival of interest in thorium based fuel is that studies, test reactor experiments and power reactor experiments have all demonstrated that thorium based fuel, whether in combination with U, Pu or U/Pu has many attractive features including the possibility of reduction of long-term radiotoxicity of discharged spent fuel, breeding of fissile material (^{233}U), burning of Pu, transmutation of minor actinides, and improvement of the proliferation resistance aspects.

Advantages, problems and possible options for thorium fuel must be realistically assessed under current conditions. Such assessment could be better implemented through an international project.

**TRANSMUTATION AND FUTURE SYSTEMS:
OVERVIEW OF THE ACTIVITIES SUPPORTED BY THE EUROPEAN COMMISSION**

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Abstract

This paper gives an overview of the research activities supported by the European Commission in the field of transmutation of radioactive waste under the Fourth Framework Programme (1994-1998). They are dealt with by five strategy studies, which are evaluating the capabilities of various burners and fuel cycles to limit the production and even destroy the stock of actinides (plutonium and minor actinides). In addition, two experimental projects are aiming at investigating the transmutation of americium and long-lived fission products. The objectives of these studies are described together with the main results already obtained. The European Union should adopt the Fifth Framework Programme (1998-2002) at the end of 1998. The content of the Fifth Framework Programme and the broad lines of the activities foreseen in partitioning and transmutation and future systems are briefly presented.

Introduction

The specific programme on Nuclear Fission Safety in the Framework Programme for the European Atomic Energy Community (1994-1998) has five main activity areas: Exploring Innovative Approaches, Reactor Safety, Radioactive Waste Management and Disposal and Decommissioning, Radiological Impact on Man and Environment and Management of the Consequences of the Chernobyl Accident. The area "Exploring Innovative Approaches" is covering research activities on reactor safety features as well as on fuel cycle concepts. The work on fuel cycle concepts is dealing with partitioning and transmutation (P&T) and includes three tasks: strategy studies, partitioning techniques and transmutation techniques. Nine research projects on P&T are partly funded by the EC with five in the field of strategy studies, two in partitioning techniques and two in transmutation techniques. As the chemical separation of long-lived radionuclides from high level waste is not in the scope of the workshop, the objectives and the main results obtained so far are reported only for the seven research projects dealing with strategy studies and transmutation techniques.

The European Union is now finalising the Fifth Framework Programme (1998-2002) which should be adopted before the end of 1998. The specific programme for research and training in the field of nuclear energy includes activities on both controlled thermonuclear fusion and nuclear fission. In the field of nuclear fission, it is foreseen to increase the support to the research activities devoted to partitioning and transmutation and to future systems. The future systems considered in the programme are in particular innovative or revisited concepts for reactors, fuels and fuel cycles. The content of the Fifth Framework Programme and the broad lines of the activities on P&T and future systems are briefly presented in the paper.

The research activities on transmutation under the Fourth Euratom Framework Programme

The objectives of the five strategy studies on P&T (four shared-cost actions and one concerted action) and of the two experimental projects on transmutation techniques [1] are described together with the main results obtained so far [2,3]. The strategy studies are aiming at assessing the capabilities of different burners and fuel cycles to limit the production or even destroy the stock of actinides (plutonium and minor actinides). In this section, three projects related to transmutation in reactors are first presented, followed by a study dealing with the thorium fuel cycle and finally three projects on accelerator driven systems (ADS).

Evaluation of possible P&T strategies and of associated means to perform them

This project is co-ordinated by CEA Marcoule and involves ten other European laboratories (FZK Karlsruhe, AEA Technology Harwell, BNFL Risley, Belgonucléaire Brussels, SCK.CEN Mol, ECN Petten, JRC-ITU Karlsruhe, GRS Köln, ENEA Roma, Univ. "Politecnico" Milano).

The aim of this study is to give indications on what can be expected from partitioning and transmutation (P&T) strategies. The project includes five work packages: (WP1) global evaluation of different P&T scenarios and assessment of (WP2) partitioning techniques, (WP3) the feasibility of transmutation, (WP4) advanced fuel or target fabrication and (WP5) geological barrier efficiency.

The five scenarios investigated in WP1 are: (1) "once-through" cycle; (2) implementation of P&T with highly moderated pressurised water reactors (HM-PWRs); (3) implementation of P&T with CAPRA fast neutron reactors (FNRs); when equilibrium is reached for scenario (3), it can be

followed by either (4) a progressive introduction of EFR FNRs or (5) a gradual decrease of nuclear power. Recycling of plutonium (Pu) and minor actinides is implemented after a 40-year period during which Pu is only recycled once in current MOX-fuelled PWRs. The total installed electric power remains fixed at 120 GWe.

The first simulation of Scenario 2 showed an impossibility to recycle both plutonium and minor actinides with only HM-PWRs. A new simulation, with a partial recycling of minor actinides is in progress. In the case of CAPRA burners (Scenario 3), the envisaged recycling of actinides seems quite feasible from the reactor standpoint. However, the possibility of handling and reprocessing fuels with Pu contents near 50% is being checked in WP2 and WP4. The scenario evaluation confirmed that mining and milling and reactor operation were the main sources of short-term risk to the public in the fuel cycle. The risk is smaller for Scenario 3 than for Scenario 1, as the additional risk due to reprocessing operations is largely compensated by the decrease of mining activities.

In WP5, three geological host formations in which the waste resulting from the scenarios would be disposed of are evaluated on the basis of specific sites (Mol for clay, Gorleben for salt) and a generic site (granite). Normal evolution and human intrusion scenarios are considered. Partial results have been obtained, which need to be refined and homogenised. In the case of repositories in salt, the prominent contribution of medium-level waste to the long-term risk has been outlined.

Preliminary assessments of PUREX-based and pyro-metallurgical reprocessing techniques applied to CAPRA fuels were performed (WP2). For the PUREX process, the main issue is the dissolution of fuel containing high plutonium concentration in nitric acid for which experimental data are expected. Concerning the pyro-metallurgical process, the oxide-to-metal conversion required for its implementation seems possible, but the influence of the accumulation of fission products in molten salt during recycling needs further assessments.

Concerning transmutation (WP3), most of the work deals with HM-PWRs and CAPRA FNRs. The HM-PWRs cores could be beneficial with respect to the minor actinide build-up. In this case, significant plutonium consumption appears possible with moderator-to-fuel ratio around 4, but the safety of such cores seems doubtful, when considering the present requirements. A high burn-up (210 GWd/t) CAPRA core was devised, which showed a great improvement of the fuel residence time and of the reactivity loss, in comparison with the standard CAPRA core. This improvement was compensated by a slight decrease in the specific plutonium consumption and a need for additional $^{11}\text{B}_4\text{C}$ moderator.

Three processes for fabrication of fuels and targets containing minor actinides have been investigated in WP4. The MIMAS process (powder blending) could be used without significant difficulties for this purpose. The sol-gel precipitation process could be safer (it produces less dust) and better for recycling (more homogeneous U-Pu mixture). Impregnation techniques tested in the framework of the EFTTRA programme proved to be suitable for target fabrication.

Supporting nuclear data for advanced MOX fuels

Belgonucléaire, Brussels, co-ordinates this study, which involves five other European partners (ECN Petten, SCK.CEN Mol, CEA Cadarache, JRC-ITU Karlsruhe, ENEA Bologna).

Its objective is to provide more accurate nuclear data for the P&T scenarios aiming at reducing the waste toxicity investigated in the strategy study described above. The use of advanced MOX fuels, either in PWRs or in FNRs, is studied. The necessary nuclear data working libraries are updated with new information from basic data evaluations and available integral experiments. The accuracy of the strategy studies to minimise wastes will be assessed at the end of the study.

Since the beginning of the project, a comprehensive compilation of irradiation results for high burn-up fuels has been carried out and experiments have been recalculated with modern computer codes and data sets.

The analysis of the composition of MOX fuels after irradiation at high burn-up (80 GWd/t in the overmoderated BR3 reactor in Mol and up to 46 GWd/t in the Saint Laurent B1 reactor) provides the experimental basis for PWRs. The values measured for the isotopic masses in the discharged fuel are being compared with the recalculated ones. Fairly good agreement is being found. In addition, the assessment of modelling errors has been completed.

In the case of the analysis of irradiation in FNRs, work has been progressing well. The comparison of computed and measured values for PHENIX irradiations (PROFIL experiment) has been completed. Concerning the irradiations in KNK II in Karlsruhe, the chemical analysis of the fuel pins has been also completed and that of the single nuclide samples has started.

Finally, the re-evaluation of the ^{241}Am , ^{240}Pu and ^{242}Pu data files is nearly finished.

Joint EFTTRA experiment on americium transmutation

The objective of the project is to study the transmutation of ^{241}Am embedded in an inert matrix (spinel) in an irradiation experiment in the high flux reactor (HFR) at Petten. It includes the fabrication of a target pin, the execution of irradiation, post-irradiation examinations (non-destructive and destructive) and the interpretation of the results.

The work is being performed by the EFTTRA group, a collaboration of laboratories and research institutes in Europe (CEA and EDF in France, FZK in Germany, ECN in the Netherlands and ITU and IAM of the JRC of the European Commission).

The project is progressing according to plan. The irradiation of the target in HFR and the non-destructive post-irradiation examinations have been completed. The destructive examinations will be carried out by ITU soon.

The target contained 10 pellets of spinel (aluminium-magnesium oxide) with 11.5% in weight of americium oxide (about 0.7g of americium in total) in a titanium steel alloy cladding. It has been irradiated for 359 days in HFR, which represents a fluence of $1.7 \times 10^{26} \text{m}^{-2}$ for neutrons with an energy above 100 keV.

The first non-destructive post-irradiation examinations indicate that:

- The distribution of americium is not homogeneous inside the pellets; there is more americium at the edge of the pellet than inside; this is due to the impregnation technique used to fabricate the pellet.

- The pellets have swollen in volume by 15 to 18%, which is too much compared to a swelling of about 3% for uranium oxide; the swelling is caused by the alteration of the spinel matrix by the fission products.

The results of the calculations of the radial and axial transmutation effects can be summarised as follows:

- The flux profile is flat in the vertical direction.
- 92.3% of americium has been transmuted.
- The total content of actinides in the sample has decreased by 32%.
- The shielding effect leads to a higher transmutation rate for americium at the outer region of the pellets than at the inner region.

Thorium cycles as a nuclear waste management option

This project is co-ordinated by ECN Petten. Six other European laboratories are participating in this study: CNRS Paris, Belgonucléaire Brussels, CEA Cadarache, FZJ Jülich, JRC-ITU Karlsruhe, ENEA Roma.

The goal of the project is an assessment of thorium cycles with a view to limit nuclear waste production and to burn waste, keeping in mind that thorium will be, in any case, a long-term option. The major fuel cycle steps have to be reviewed, focused on the European situation with thorium fuelled PWRs and FNRs as candidate reactors. The case of one thorium fuelled hybrid system is also investigated.

The six work packages of the project are covering the different steps of the thorium (Th) fuel cycle. The results obtained so far can be summarised as follows.

From the study of “Mining” (1), it appears that the thorium cycle leads to less mining waste production than the uranium cycle and that the waste could be disposed of in geological formations, which is not possible in practice for uranium waste. The final extracted thorium oxide product, however, has a higher gamma ray, alpha dose and radiotoxicity than uranium oxide.

“Fabrication of fresh thorium fuels” (2) does not lead to dose rates higher than those of conventional MOX fuel.

The study of “PWR (once-through) scenarios with Th/U fuel” (3.1) indicates that, compared to the reference low-enriched uranium scenario, the radiotoxicity is strongly reduced for the waste in all cases up to about 20 000 years, with best results for Th with highly enriched uranium at high burn-up. For longer storage times, decay products of ^{233}U dominate. Recycling will be studied in the next phase. “Th-assisted (once-through) Pu-burning in PWRs” (3.2) is a very good option, since such cores can consume twice more TRU compared to U/Pu MOX. Recycling will be studied in the next phase. “Fast reactors with Th and full recycling of actinides” (3.3) offer excellent possibilities to burn Pu and to breed ^{233}U simultaneously. A self-sustaining core with Th/ ^{233}U is possible with very small void coefficient. Important waste radiotoxicity reductions are possible below 30 000 years. “Accelerator

driven fast reactors” (3.4) loaded with Th/²³³U are being studied; computational methods have been validated and nuclear data problems have been identified. Waste radiotoxicity with recycling of actinides will be studied in the next phase.

“Reprocessing” (4) using the single-cycle THOREX process can lead to U, Pu losses of 0.1% and to protactinium (Pa) losses of 1-2%. Also high fission product decontamination factors are possible. The pyrochemical method seems promising for Th fuel as well.

The “Residual risks of geological disposal” (5) were evaluated for once-through PWR scenarios with Th/²³³U and Th/Pu fuels. The total dose of the actinides (most important ²³¹Pa) turned out to be rather small. In fact, long-lived fission products (¹²⁹I, ¹³⁵Cs) gave the highest contribution, followed by activation products (⁵⁹Ni, ⁹⁴Nb). There was not much difference with U-based fuels.

Finally, a systematic approach has been developed to assess the “Technical aspects of non-proliferation” (6). Some concerns with respect to possible proliferation of ²³³U in case of recycling and the misuse of accelerators were formulated.

Impact of the accelerator-based technologies on nuclear fission safety (IABAT)

KTH Stockholm is co-ordinating this project, in which participate nine other European research institutions (CEA Cadarache, ECN Petten, FZJ Jülich, ENEA Casaccia, FZK Karlsruhe, JRC-ITU Karlsruhe, AEA Harwell, Univ. Uppsala, Univ. Chalmers Göteborg).

The overall objective of the IABAT project is to make a European assessment of the possibilities of accelerator driven hybrid reactor systems from the point of view of safe energy production, minimum waste production and transmutation capabilities. This study has four work packages. The progress achieved in each of them is given as follows.

The first work package, “System studies on an accelerator driven hybrid”, includes eight research topics. Specific ADS systems have been chosen for future analysis. These systems cover a wide range of different ADS parameters like: neutron spectra varying from thermal to fast neutrons, fuel form from solid through suspension in liquid lead to molten salt solutions. ADS studied in this project involve different fuel cycles: thorium-based plutonium burners, LWR waste incinerators and minor actinide incinerators. A number of code systems to simulate, validate and benchmark different ADS concepts have been set up.

Concerning the “Assessment of the technology and cost of linear and circular accelerators” in the second work package, the basic requirements for the accelerator power and performance have been preliminary formulated.

There are four research topics in the third work package, “Basic nuclear and material data”. First experiments on ²³²Th fast fission yields and ²³³U fission yields have been performed at the OSIRIS mass-separator facility in Sweden. It has been agreed in the project that the optimal way of performing ADS calculation in the future will be the creation of an intermediate energy range (20-150 MeV) cross-section library for neutrons and protons and extending the existing reactor cross-section libraries to energy up to 150 MeV. This approach is complementary to the one chosen by the CERN group, which puts its efforts in the development of high energy transport codes to improve the simulation of high energy particle transport without creating new libraries.

In the fourth work package, “Studies of the fuel cycle for ADS”, which includes four research topics, calculations of the radiotoxicity of the thorium-based ADS fuel cycle have been performed.

Finally, it is worth mentioning that:

- The analysis of different conceptual designs points in the direction of fast spectrum and liquid lead (or lead/bismuth) cooling for ADS.
- An international collaboration on ADS is being established between different countries, continents and international organisations (EC, OECD-NEA, IAEA, ISTC).

Neutron driven nuclear transmutation by adiabatic resonance crossing (TARC)

This project is carried out by CERN as co-ordinator, Universidad Autónoma de Madrid, CNRS Grenoble and the Universities of Athens and Thessaloniki.

The main aim of this project is to develop both theoretically and experimentally a new method, the adiabatic resonance crossing (ARC), which enables to strongly enhance the capture rate of neutrons by the radionuclides to be incinerated. The peak cross-sections for neutron energies corresponding to the resonance region are much larger than for the other neutron energies. For instance, the cross-section of ^{99}Tc is 4 000 barn at the peak of a resonance at 5 eV, but it is only 20 barn at thermal energy. Access to the resonance region can be achieved by using a transparent medium like lead where the neutrons lose their energy in very small decrements. Then, the neutron energy decreases slowly through the resonances, leading to very large probability of capture during crossing. An experimental test has been carried out on ^{99}Tc with a neutron spallation source driven by the CERN proton synchrotron.

The project has six work packages:

- 1) Setting-up of the lead assembly and preparation and instrumentation of the beam line.
- 2) Experiments on ARC including timing experiments (CeF_3 counters) and activation experiments (delayed γ counting).
- 3) Study of advanced neutronics in lead by electronic experiments, activation measurements and temperature measurements.
- 4) Development of an appropriate formalism and computational tools for ARC.
- 5) Conceptual design of an incinerating device based on ARC.
- 6) Other applications of ARC.

The project has been completed by carrying out the following tasks. The detailed analysis of the data (work packages 2 and 3) has concentrated on the following items: (i) measurement of ^{99}Tc transmutation rate and of the ^{99}Tc neutron capture cross-section as a function of neutron energy with the CeF_3 detector; (ii) measurement of the production rate of ^{233}U from ^{232}Th and ^{239}Pu from ^{238}U using delayed γ spectroscopy; (iii) neutron flux measurements over the lead capture resonance region with

³He chambers working in the ionisation mode; (iv) measurements of the high neutron energy flux (≥ 1.4 MeV) with fission detectors; and (v) thermoluminescence measurements of the neutron flux below 0.5 eV. The computational tools that are indispensable for the interpretation of these data (work package 4) have been developed in parallel. The various detectors have been simulated and the results of simulation have been compared with experimental data in a systematic way. The present assessment of the experimental data confirms clearly the possibility of destroying in an effective fashion ⁹⁹Tc and ¹²⁹I by using the ARC method, as well as extending this concept to other applications.

Physical aspects of lead as a neutron producing target for the accelerator transmutation devices

The University of Louvain-la-Neuve is co-ordinating this concerted action, which involves twelve other European research institutions (Univ. Uppsala, Univ. Hannover, PTB Braunschweig, CEA Saclay, CNRS Nantes, Univ. Groningen, ECN Petten, GSI Darmstadt, FZJ Jülich, CNRS Caen, CNRS Orsay, Univ. Liège).

This concerted action started in the second half of 1998 for a two year duration. Its aim is to evaluate lead as a spallation target for the production of high intensity neutrons. The available experimental data are being compiled and compared to predictions from different theoretical models as well as the new measurements, which will be obtained in the EU laboratories in the next two years. The concerted action will be concluded by recommendations on possible additional measurements on lead and on the best codes to evaluate the reaction mechanisms.

The work programme includes the following items:

- Reactions induced by protons (30-70 MeV) and by neutrons (30-65 MeV).
- Production of residual nuclei by protons and by neutrons.
- Elastic scattering of 100 MeV neutrons on lead.
- Fission in proton induced reactions at 190 MeV on lead.
- Spallation induced by protons and deuterons at 0.8, 1.2 and 1.6 GeV.
- Fragmentation of ²⁰⁸Pb ions of 1 GeV.A on hydrogen.
- Particle multiplicities on lead targets induced by protons from 0.8 to 2.5 GeV.

Transmutation and future systems under the Fifth EURATOM Framework Programme

The Commission adopted its proposal for the Fifth Framework Programme (1998 to 2002) in April 1997 (content) and in July 1997 (budget). After a first reading by the Parliament in December 1997, the proposal was amended by the Commission in January 1998 (see Table 1). The Council reached unanimous political agreement on a common position concerning the Fifth Framework Programme (FP5) at its meeting on 12 February with an overall budget of 14 000 ECU, of which 1 260 ECU is allocated to the Euratom part of FP5. Because of the difference between the proposal of the Council and that of the Parliament (16 300 MECU as the Commission) for the overall

Table 1. Fifth Framework Programme – proposal by the Commission

• Quality of life and management of living resources	2 635 MECU
• User-friendly information society	3 925 MECU
• Competitive and sustainable growth	2 895 MECU
• Preserving the ecosystem	2 320 MECU
• Confirming the international role of Community research	491 MECU
• Innovation and encouragement of participation of SMEs	350 MECU
• Human research potential and socio-economic knowledge base	1 402 MECU
• Activities to be carried out by means of direct actions (JRC)	815 MECU
• Euratom Framework Programme	1 467 MECU
Total	16 300 MECU

budget of FP5, the conciliation procedure between both institutions started at the end of September in view of reaching an agreement on the budget by November. A good compromise would be around 15 000 MECU in the end. The adoption of FP5 is expected for December 1998.

The specific programmes implementing the Fifth Framework Programme were proposed by the Commission in May 1998. The Council reached provisional agreements on the content of the specific programmes at its meetings on 22 June and 13 October. The final decision should be taken at the end of 1998. The work programmes are being prepared by the Commission. Targeted calls for proposals should be launched from the beginning of 1999.

There are two specific programmes implementing the Euratom part of FP5 [4]. The first one, named “Nuclear Energy”, has two key actions, one on controlled thermonuclear fusion and one on nuclear fission. In addition, there are generic activities on radiation protection and support for research infrastructure. The second specific programme is concerning the direct actions to be implemented by the Joint Research Centre. Table 2 shows the breakdown proposed by the Commission for the budget of the Euratom part of FP5. Note that the total amount of 1 467 MECU proposed by the Commission has to be compared to the proposal of the Council of 1 260 MECU.

Table 2. Euratom Fifth Framework Programme – proposal by Commission

• Key action thermonuclear fusion	920 MECU
• Key action nuclear fission	176 MECU
• Generic research radiation protection	35 MECU
• Support for infrastructures	10 MECU
• Joint Research Centre	326 MECU
Total	1 467 MECU

The overall objectives of the key action on nuclear fission are to enhance the safety of Europe’s nuclear facilities and to improve the competitiveness of Europe’s nuclear industry. The more detailed

aims are to ensure the protection of workers and the public from radiation, to assure a safe and effective management and disposal of radioactive waste, to explore more innovative concepts and to contribute to the maintenance of a high level of expertise in nuclear technology.

The key action on nuclear fission comprises four chapters: (i) operational safety of existing installations (ageing effects, severe accidents); (ii) safety of the fuel cycle (waste management and disposal, decommissioning, partitioning and transmutation); (iii) safety and efficiency of future systems; and (iv) radiation protection.

In the area of partitioning and transmutation, strategy studies are foreseen to investigate its benefits and compare different methods such as critical and subcritical systems taking into account the whole fuel cycle. New efficient and selective processes will be developed for the separation of the critical long-lived radionuclides from high level and medium level waste (e.g. hydro-metallurgical processes, chromatography and pyro-chemical processes). Basic nuclear data essential for transmutation and the development of ADS will be measured and computed. The radiation damage induced by spallation reactions in materials will be investigated. It is foreseen to develop and test fuels and targets for actinide and long-lived fission product incineration. The preliminary study of an ADS is also considered in the programme with supporting research work on subcritical mock-ups, safety, coolants (liquid metal or gas), the confinement of the accelerator/reactor window and high power accelerators. Finally, new specific matrices could be also developed for the conditioning of long-lived radionuclides, which cannot be transmuted.

The future systems foreseen in the programme are on one hand evolutionary safety concepts and on the other innovative or revisited concepts. This last item deals in particular with the assessment of new or previously discarded reactor concepts that would be potentially cheaper, safer, more sustainable, producing less waste and reducing the risk of diversion (e.g. gas-cooled fast neutron reactors, high temperature reactors). Small reactors will be evaluated, identifying those of potential interest to the market beyond the next decade. The commercial potential of special applications of nuclear energy, such as combined heat and electricity production, desalination, hydrogen and methanol production will be investigated. Innovative fuels will be studied with advantages in terms of use of fissile materials, robustness to severe accidents and long-term storage. New fuel cycle concepts will be evaluated and developed (e.g. thorium fuel cycle and integral reactor with on-line reprocessing and fuel fabrication).

Conclusion

The research activities on transmutation under the Fourth Framework Programme are assessing the possibilities of destroying harmful radionuclides such as actinides and long-lived fission products in different burners (e.g. reactors, accelerator driven systems). The thorium fuel cycle is also being evaluated as an option to limit the production of actinides and even to incinerate them. These studies involve the participation of about 50 European research laboratories. Most of the projects will be completed early in 1999. Significant results have been already obtained, which can be used as an input for the Fifth Framework Programme.

The Fifth Framework Programme should be adopted by the European Union at the end of 1998. It is foreseen to make targeted calls for proposals from the beginning of 1999. In the key action on nuclear fission, a substantial part of the research work should be devoted to partitioning and transmutation and future systems.

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RECENT PROGRESS OF THE EFTTRA RESEARCH ON FUELS AND TARGETS FOR TRANSMUTATION OF ACTINIDES AND FISSION PRODUCTS

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Abstract

The progress of the irradiation programme of EFTTRA is described. Firstly, the results of the recent transmutation test for technetium in the HFR are discussed. In the so-called T2 test, a target of metallic technetium was transmuted to an extent of about 16%. Post-irradiation examination of the target revealed an excellent in-pile behaviour. Secondly, the investigations of concepts for inert matrix fuel (IMF) are discussed. This activity is ongoing and only preliminary results on the radiation stability of candidate inert matrix materials are available. Thirdly, the results of the T4 test, the transmutation of americium in a spinel-based fuel (microdispersed), are presented. It is shown that extensive swelling of the fuel took place, probably due to the damage of the matrix by fission product and/or accumulation of helium as a result of alpha decay. A more promising IMF-concept for transmutation of americium, based on a macrodispersion of a host phase in the inert matrix is discussed.

Introduction

EFTTRA, which is the acronym for Experimental Feasibility of Targets for Transmutation, is a network of research organisations in France, Germany and the Netherlands formed in 1992 [1,2]. The goal of EFTTRA is the study of transmutation of americium as well as of the long-lived fission products technetium (^{99}Tc) and iodine (^{129}I). The work of the partners of the EFTTRA group is focused on the development and testing of targets and fuels, taking into account the scenarios developed in Europe for possible P&T strategies. To that purpose fabrication routes are being investigated, irradiation tests are performed, and post-irradiation examinations are made in the various facilities of the EFTTRA partners. In Table 1 an overview is given of the EFTTRA irradiation experiments that are completed or ongoing at present.

Table 1. EFTTRA irradiation experiments

Name ^a	Reactor	Description	State of the art	Refs.
T1	HFR	Technetium and iodine	Completed	3-6
T2	HFR	Technetium	Completed	7
		Neutron damage in inert matrices	Completed	8
T2bis	HFR	Neutron damage in inert matrices	Completed	
T3	HFR	Neutron damage in inert matrices	PIE to be started	
		Dispersion inert matrix fuel using enriched UO_2	PIE to be started	
T4	HFR	Americium in spinel	PIE ongoing	9,10
T4bis	HFR	Americium in spinel	Irradiation ongoing	
T4ter	HFR	Central fuel temperature of spinel/ UO_2	Irradiation ongoing	11
F1	Phénix	Neutron damage in inert matrices	Continued	12
		Dispersion inert matrix fuel using enriched UO_2	PIE ongoing	12
F1A	Phénix	Neutron damage in inert matrices	Irradiation ongoing	
		Dispersion inert matrix fuel using enriched UO_2	Irradiation ongoing	
F2	Phénix	Technetium and iodine	Start pending	

^a Some of the experiments are also known under different names such as RAS or MATINA.

In the present paper the progress of the EFTTRA research activities is described. The paper will focus on results of the most recent irradiation tests of targets for transmutation of technetium and of inert matrix fuels for transmutation of americium. The latter subject will be dealt with in general, discussing the results of experiments on inert matrices, in some cases mixed with enriched uranium oxide to simulate americium, and very specifically by presenting the first results of the T4 experiment in which a target of spinel containing 11 wt% americium was irradiated in a high thermal flux.

Transmutation of technetium (T1 & T2)

The study of the transmutation of the technetium was started in 1992 when irradiation tests of targets of ^{99}Tc metal were planned for the high flux reactor (HFR) at Petten as well as the Phénix fast reactor at Cadarache (see Table 1). The fabrication of the targets for these tests was studied at ITU where a technique was developed for the fabrication of cylinders/pellets by casting of liquid technetium metal in a water-cooled copper mould. The targets that were fabricated for the irradiation tests each contained two such cylinders.

The results of the T1 test, which was completed in 1995, have been reported extensively [3-6]. They showed good behaviour of the technetium metal at an extent of transmutation of about 6%. To study the behaviour at an higher extent of transmutation, one of the targets (two cylinders) of the T1 test was re-encapsulated and irradiated again in the HFR. The T2 test was successfully completed in 1997, and the results of the post-irradiation examinations have recently become available and will be summarised shortly below.

The cumulative irradiation time for the T2 experiments was 24 reactor cycles (579.3 full power days) during which the total neutron fluence accumulated to about $5.8 \times 10^{26} \text{ m}^{-2}$. The post-irradiation examination (PIE) revealed an excellent in-pile behaviour of the metallic technetium rods. Metallographic examinations showed no changes in the microstructure compared to the unirradiated material. An example of a typical micrograph is given in Figure 1. Electron probe microanalysis (EPMA) of the radial distribution of ruthenium, the product of the transmutation process, showed an increase from about 15-16% in the centre to 30-40% near the rim of the pellets, giving a pellet average Ru concentration of about 18% (Figure 2). Mass spectrometric analysis of the pellet average Ru concentration showed that the extent of transmutation is about 16%.

Figure 1. Microscopic cross-section of irradiated Tc target from the T2 experiment showing oriented crystal growth

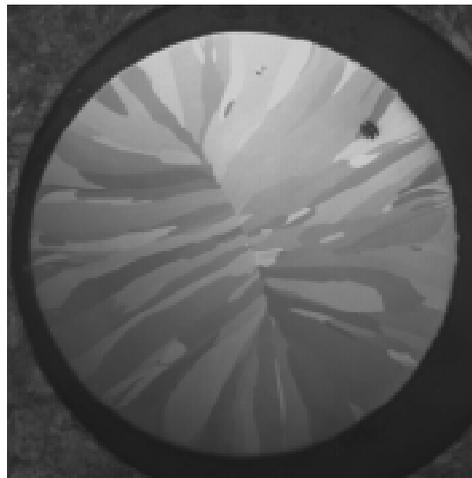
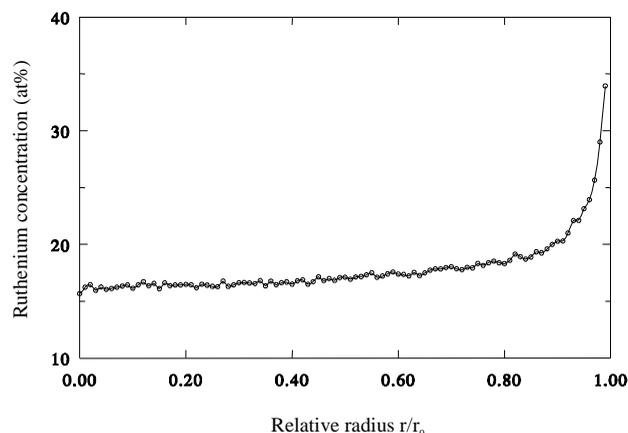


Figure 2. A representative EPMA curve of the ruthenium concentration in technetium from the T2 experiment



From the results of the T2 test it is concluded that there are no technical limitations to the use of metallic technetium as a target for transmutation. No further irradiation EFTTRA experiments in the HFR are foreseen, but an irradiation test of the same material in the Phénix reactor (F2) is in preparation, as mentioned before.

Inert matrix fuels (T2, T3, F1 and F1A)

The inert matrix fuel (IMF) concept is being investigated by EFTTRA for the transmutation of minor actinides. In the IMF concept an actinide compound is dispersed in a matrix of a non-activating material that has a good thermal conductivity and keeps a good mechanical integrity. A number of suitable inert matrix materials has been selected on the basis of literature survey or laboratory experiments [13]. These include oxides, metals and nitrides.

The radiation damage to the inert matrix is considered to be one of the major items to be investigated. The radiation damage is caused by neutrons, alpha decay (producing ~5 MeV alpha particles and ~100 keV recoil atoms) and fission products (light and heavy ions of about 70-100 MeV) and might result in unfavourable behaviour during irradiation, such as void formation, swelling or decrease of thermal conductivity.

In the EFTTRA programme the radiation stability of the most interesting inert matrices is tested by irradiations in the HFR at Petten as well as in Phénix at Cadarache (see Table 1). In these tests the radiation effects of neutrons as well as of fission fragments are studied by irradiation of pure matrices as well as their mixtures with UO₂ enriched in ²³⁵U, respectively. At present most attention is given to oxide matrices as MgO, MgAl₂O₄ (spinel) and Y₃Al₅O₁₂ (YAG). The following results have been obtained up to now.

- *T2 and T2bis.* The objective of these experiments is the investigation of the effect of neutron irradiation on the properties of inert matrix materials (Al₂O₃, MgAl₂O₄, Y₃Al₅O₁₂). Both experiments have been completed and the neutron fluences reached $2 \times 10^{26} \text{ m}^{-2}$ (T2bis) and $6 \times 10^{26} \text{ m}^{-2}$ ($E > 0.1 \text{ MeV}$). The temperature was about 815 K. Post irradiation examinations revealed that MgAl₂O₄ and Y₃Al₅O₁₂ showed negligible neutron induced swelling; Al₂O₃, which was also included in the test but which has already been discarded as inert matrix material, swelled considerably, as expected. These results extend and confirm the existing data on these materials.
- *T3.* In this experiment inert matrices mixed with UO₂, 20% enriched in ²³⁵U, have been irradiated to investigate the radiation effect of fission products. For comparison, the pure materials were also included. The inert matrices tested are MgO, CeO₂, Y₂O₃, MgAl₂O₄, and Y₃Al₅O₁₂. Different techniques have been used for the fabrication of the pellets (co-precipitation and powder blending). The burn-up reached in this experiment was about 18% FIMA. Neutron radiography of the sample holder did not reveal any irregularities of the samples. Non-destructive and destructive post-irradiation examinations are being performed at this moment.
- *F1.* In this experiments inert matrices have been and are being tested for their stability to fast neutrons and fission products. The materials included are oxides (MgO, Al₂O₃, MgAl₂O₄, and Y₃Al₅O₁₂), nitrides (TiN) and metals (W,Nb,V,Cr). The first irradiation (F1, also known as MATINA-1) was performed in 1995 and reached a fluence of $2 \times 10^{26} \text{ m}^{-2}$ ($E > 0.1 \text{ MeV}$) and a burn-up of 1.3% FIMA in the targets containing UO₂. The next experiments are aimed at

higher fluences up to $20 \times 10^{26} \text{ m}^{-2}$. After the F1 experiments all 19 pins were examined by neutron radiography. No elongation of the column of pellets was observed, except for Al_2O_3 for which the column length increased by about 1.3%. Two pins, $\text{MgO}+\text{UO}_2$ and $\text{MgAl}_2\text{O}_4+\text{UO}_2$, containing 40w% UO_2 , were examined destructively and the results of dimensional control, ceramography and EPMA revealed a good behaviour of these two composite fuels.

Transmutation of americium in an inert matrix (T4 and T4bis)

In 1995 the T4 experiment was initiated by the EFTTRA partners to study the feasibility of americium transmutation in a so-called once-through mode. In this scenario the extent of fissioning should be very high (>90%). The T4 experiment was performed as a shared cost action project in the cluster “P&T strategy studies and transmutation experiments” of the Fourth Framework Programme of the CEC (contract no. FI4I-CT94-0007).

For the T4 experiment pellets of magnesium aluminate spinel (MgAl_2O_4) containing about 10-12 wt% ^{241}Am were fabricated by the newly developed infiltration method (INRAM) at ITU [9,14]. This methods yields a relatively fine distribution of the americium in the pellet [9,14]. However, it was observed that this distribution is not uniform and that the americium, intended to be present as an oxide, formed a compound during sintering, probably AmAlO_3 . Two irradiation capsules were prepared.

The irradiation of the T4 target in the HFR at Petten was performed from August 1996 to January 1998 (358.4 full power days), during which a burn-up of 32% FIMA (value obtained from post-test burn-up calculation) was achieved. During the irradiation a neutron radiograph was made. After the irradiation the fuel was examined non-destructively in the hot cell laboratories in Petten, of which the following results are now available [10]:

- Neutron radiography of the sample holder at mid-of-life shows no visible damage of the fuel pin and an increase of the length of the pellet stack (Table 2).
- Gamma spectrometry and gamma tomography of the fuel pin shows that the distribution of the fission products is correlated to the initial americium concentration of the pellets. An example (^{141}Ce) is shown in Figure 3; profiles for other nuclides are similar.
- Gamma spectrometry and gamma tomography of the fuel pin show an increase in length of the pellet stack and in the diameter of the pellets (Table 2).
- Profilometry of the fuel pin showed an increase of the diameter of the cladding from 6.55 mm originally to 6.75 mm at maximum after irradiation (Figure 4).

Table 2. Dimensional changes of the pellets of the T4 test derived indirectly from non-destructive examinations

	Length (%)	Diameter (%)
Neutron radiography (MOL ^a)	+5.2	
Gamma spectrometry	+4.5	+6.7
Gamma tomography		+6.7

^a MOL = mid-of-life (231.4 full power days)

Figure 3: Axial distribution of the fission product ^{141}Ce along the pellet stack as obtained by gamma spectrometry (O) and the initial americium concentration in the pellets (solid line)

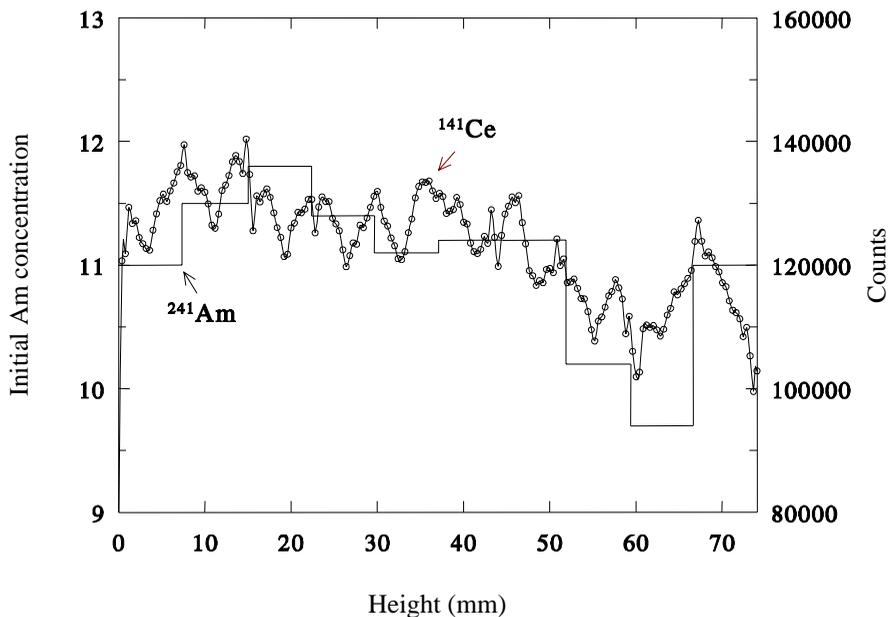
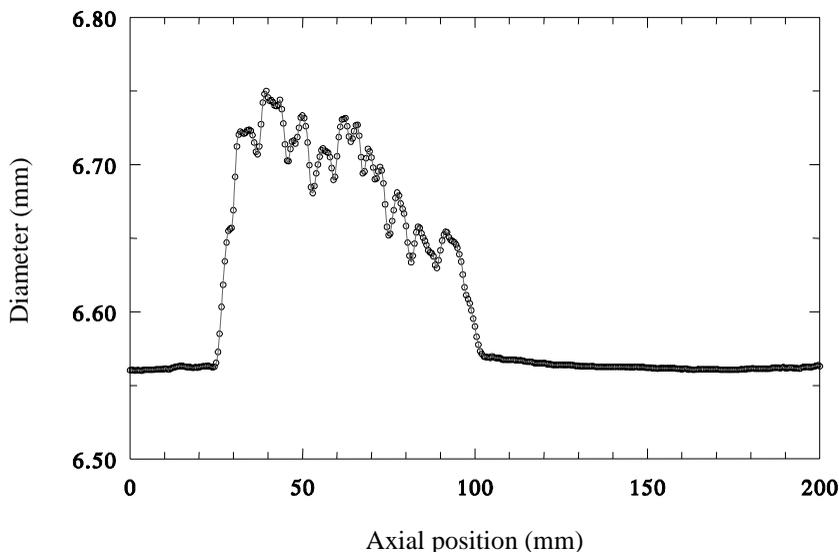
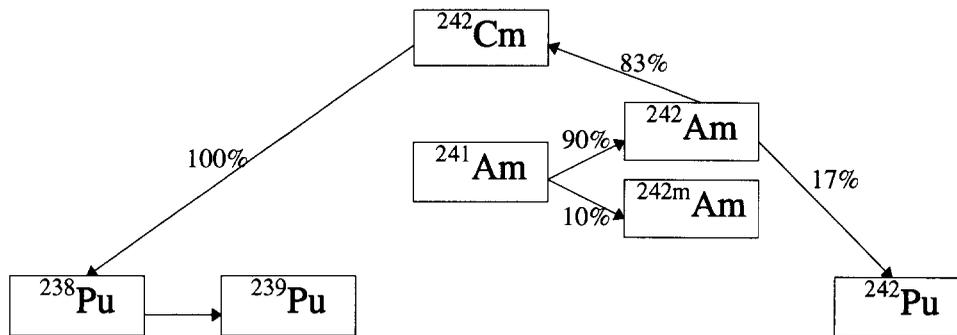


Figure 4. The diameter of the cladding of the irradiated T4 rod (original diameter 6.55 mm)



From the results it is clear that considerable swelling of the fuel occurred. From the data in Table 2 it can be calculated that the increase in volume is about 18%. This swelling can be caused by two different processes: (i) the damage of the spinel matrix by fission fragments, which can be extensive in this type of fuel due to the relatively small size of the americium dispersions; and (ii) the formation of gas bubbles containing helium that was produced by the alpha decay of ^{242}Cm , one of the isotopes in the transmutation chain of ^{241}Am (Figure 5).

Figure 5. Transmutation scheme for ^{241}Am in a thermal flux. For the present context two points are of relevance: (i) because of the very short half life of ^{242}Am ($t_{1/2} = 16\text{ h}$) most of it will decay to ^{242}Cm , though its fission cross-section is high ($\sigma_f = 2\,900\text{ barn}$); and (ii) the decay product ^{242}Cm has a low fission cross-section ($\sigma_f < 5\text{ barn}$) and decays with a short half life of 163 days, producing an alpha particle.



In 1997 the irradiation of an identical capsule was started, the accumulated irradiation time at the moment of the writing of this paper being 275.1 full power days. It is the intention to reach a significant higher extend of fission ($> 70\%$ FIMA) if the fuel does not fail due the swelling noted in T4.

The “hybrid” fuel concept

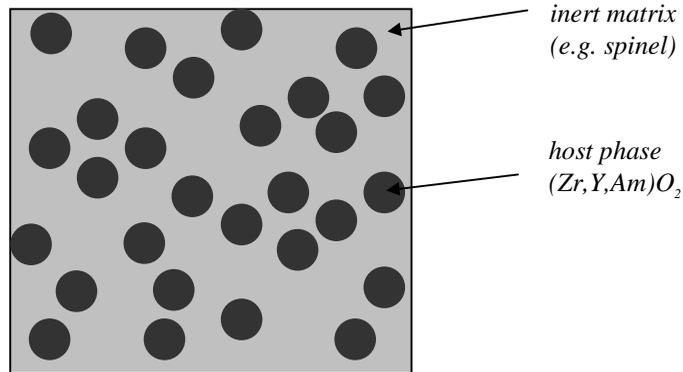
The future plans of the EFTTRA group are directed towards the improvement of the inert matrix fuel concept for americium targets. The results of the T4 test, combined with results of theoretical studies and separate effect experiments, are being used to modify the fuel design as reported in a separate study [15]. It is concluded that in principle a dispersion of the americium is to be preferred to a solid solution in order to localise the damage to the matrix, caused by fission fragments, in a small volume. Such a dispersion is composed of spherical inclusions (between 50 and 300 μm) of a host phase, which contains the americium, in a dense inert matrix (Figure 6). For the host phase the cubic solid solution $(\text{Zr},\text{Am})\text{O}_2$, possibly stabilised by yttrium, for the inert matrix MgO (fast reactors) and MgAl_2O_4 (thermal reactors) are the most likely candidates. Because this concept combines the advantages of dispersion and solid solution fuels, it is called “hybrid” fuel.

In order to optimise the hybrid concept the following research activities are required:

1. *Fundamental properties of the $\text{ZrO}_2\text{-Y}_2\text{O}_3\text{-AmO}_{2-x}$ host phase.* The proper composition of the cubic solid solution has to be established from phase diagram studies and the physical properties (e.g. thermal conductivity, oxygen pressure, etc.) have to be measured.
2. *Development of fabrication techniques for hybrid fuels containing americium.* In the laboratories of the partners of the EFTTRA collaboration, the fabrication of hybrid fuels is being studied at this moment. Mixing of spheres, prepared, for example, by a sol-gel method, with the powder of the matrix seems to be the most likely route because it allows control of sphere size and form of the spheres.

3. *Irradiation tests of the in-pile behaviour of fuel components (inert matrix, host phase) and fuel.* To test the hybrid fuel concept a number of new tests are being defined: the T5 irradiation experiment in the HFR and the COCHIX and CAMIX experiments in Phénix.

Figure 6. Schematic representation of the “hybrid” fuel concept of a heterogeneous fuel for transmutation of americium



Conclusions

Based on the results of the work of the EFTTRA group presented in this paper we can conclude the following:

- Technetium metal is shown to be a good material for targets for transmutation of ⁹⁹Tc. However, the feasibility of a transmutation scenario for technetium is not established since the transmutation rate in a high neutron flux position of the HFR is only about 10% per full power year.
- Data on the radiation stability (neutron, fission products) of various inert matrices have been and are being generated but the results are not yet complete. A synthesis of all data, taking into account specific irradiation conditions (temperature, fast fluence, fission rate) and sample characteristics (microstructure) will be made in the near future and should allow a further selection of candidate matrices.
- The results for the IMF concept tested in the T4 experiment, a microdispersion of Am in MgAl₂O₄, were negative. A reaction between americium and MgAl₂O₄ occurred during sintering and the pellets swelled by about 18% during irradiation.
- A so-called “hybrid” fuel concept, consisting of a macrodispersion of an americium-containing host phase in an inert matrix, is proposed for the future tests to be performed.

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* *Regrets not having been able to attend.*

Annex 2
TECHNICAL PROGRAMME

Wednesday, 21 October 1998

1. Opening session

Chair: W. Kröger (PSI)

- 9:15 Welcome introduction
W. Kröger (PSI)
- 9:45 The viewpoint of Swiss nuclear utilities
H. Fuchs (ATEL), H. Mouney (EdF)
- 10:45 Scope and goals of the Workshop
P. Wydler (PSI)

2.1. Plenary Session I: Advanced U/Pu oxide-based reactors (invited papers)

Chair: A. Zaetta (CEA)

- 11:00 Pu utilisation in PWR and FR
*S. Pillon, J. Tommasi, M. Delpech (CEA), H.M. Beaumont (NNC),
T. Newton (AEA Technology)*
- 11:30 Plutonium multirecycling in a 100% MOX core with a high moderation ratio
G. Youinou, M. Delpech (CEA)
- 12:00 Aspects of uranium recycle in light water reactors
K.W. Hesketh, R. Hagger (BNFL)
- 14:00 The concept of a breeding PWR
H. Tochihara, Y. Komano, M. Ishida (MHI)
- 14:30 Study on fast spectrum BWR core with breeding characteristics
T. Yokoyama, R. Yoshioka, Y. Tsuboi, Y. Sakashita, S. Matsuyama (Toshiba)
- 15:00 Conceptual core design of a resource-renewable BWR and long-term energy supply
R. Takeda, M. Aoyama, J. Miwa (Hitachi)
- 15:30 Process development for DUPIC FUEL fabrication
K. Bae, H. Choi, J. Lee, M.S. Yang, H. Park (KAERI)

2.2 Plenary Session II: Uranium-free reactors (invited papers)

Chair: R.J.M. Konings (ECN)

- 16:30 U-free Pu fuels for LWRs: CEA/DRN strategy
J. Porta, M. Delpech, A. Puill (CEA)
- 17:00 Radiation stability of inert matrix fuels
H. Matzke (JRC ITU)

Thursday, 22 October 1998

2.2 Plenary Session II: Uranium-free reactors, *cont.* (invited papers)

Chair: R.J.M. Konings (ECN)

- 8:00 Disposition of excess plutonium by the ROX-LWR system
H. Akie, T. Yamashita (JAERI)
- 8:30 Reactors with Th/Pu based fuels
J. Tommasi, A. Puill, Y.K. Lee (CEA)

2.3 Plenary Session III: Reactors with non-oxide fuels (invited papers)

Chair: M. Nakagawa (JAERI)

- 9:00 Metallic fuel for fast reactors
L.C. Walters, G.L. Hofman, T.H. Bauer, D.C. Wade (ANL)
- 9:30 Fuel cycle systems with nitride fuel for transmutation
T. Osugi (JAERI)
- 10:00 A fast spectrum Pu burner reactor with nitride fuel
J. Tommasi (CEA), H.M. Beaumont (NNC), T.D. Newton (AEA Technology)

3. International activities (invited papers)

Chair: K. Hesketh (BNFL/NEANSC)

- 10:45 Introduction, NEA standpoint
K. Hesketh (BNFL/NEANSC)
- 11:00 Thorium fuel cycle options for advanced reactors: Overview of IAEA activities
P.E. Juhn (IAEA)
- 11:30 Transmutation and future systems: Overview of the activities supported by the European Commission
M. Hugon (European Commission DG XII)
- 12:00 Recent progress of the EFTTRA research on fuels and targets for transmutation of actinides and fission products
J.N.C. van Geel (JRC ITU), R. Comrad (JRC Petten), R.J.M. Konings (ECN), G. Muehling (FZK), J. Rouaoult (CEA), G. Vambenepe (EDF SEPTEN)

4. Parallel Sessions (contributed papers)

Parallel Session 1 – Advanced U/Pu Oxide Based Reactors

Thursday, 22 October 1998

Chair: A. Zaetta (CEA)

- 14:00 The LR-0 reactor possibilities for MOX type fuel pins research
C. Svoboda (Rez)
- 14:20 First criticality of LWR-PROTEUS: A new programme of integral experiments for advanced and innovative LWR fuels
T. Williams, P. Grimm, R. Seiler, S. Pelloni, A. Stanculescu, R. Chawla (PSI)
- 14:40 Conceptual designing of water-cooled reactors with increased or reduced moderation
T. Okubo, T. Kugo, T. Shirakawa, S. Shimada, M. Ochiai (JAERI)
- 15:00 Study of advanced LWR cores for effective use of plutonium
T. Yamamoto (NUPEC)
- 15:40 Investigation of the fuel temperature coefficient of innovative fuel types
J.L. Kloosterman (Univ. Delft)
- 16:00 Role of fast reactors in solving power problems in future
A.P. Ivanov, V.M. Poplavski, A.M. Tsiboulia (Obninsk)
- 16:20 Some issues of ²³³U fuelled SNPS reactor nuclear safety
E. Ivanov, V. Tsvirko, Y. Tsarenko (Obninsk)

Parallel Session 2 – Uranium-free reactors

Thursday, 22 October 1998

Chair: R. J. M. Konings (ECN)

- 14:00 Preliminary evaluation of a BWR with CERMET fuel core loading
L. Zanotti, G. Rouviere, S. Baldi, J. Porta (CEA)
- 14:20 Reactor physics analysis of plutonium annihilation and actinide burning in CANDU reactors
P.S.W. Chan, M.J.N. Gagnon, P.G. Boczar (AECL)
- 14:40 Neutronics of inert matrix Pu fuel rods in a UO₂ PWR environment
S. Pelloni, J-M. Paratte, A. Stanculescu, R. Chawla (PSI)
- 15:00 Some neutronic properties of an inert matrix for the definition of a 100% IMF core
J. Porta, S. Baldi, B. Guigon (CEA)
- 15:20 Inert matrix and thorium fuels for plutonium burning in LWRs
*S. Coelli, C. Lombardi, A. Mazzola, E. Padovani, M. Ricotti (POLIMI),
E. Marmo (FN), T. La Torretta, F. Vettrano, G. Zappa (ENEA)*
- 16:00 Concepts and first fabrication studies of inert matrix fuel for the incineration of Pu
*M. Burghartz, G. Ledergerber, F. Ingold, T. Xie, F. Botta (PSI),
K. Idemitsu (Fac. of Eng. Kyushu Univ., Japan)*

- 16:20 Uranium-free burner reactor dedicated to minor actinides transmutation
N. Messaoudi, J. Tommasi, M. Delpech (CEA)
- 16:40 Silicon carbide as an inert matrix fuel for CANDU reactors
R. Verrall, H.R. Andrews, I.M. George, P.J. Hayward, P.G. Lucuta, S. Sunder, M.D. Vljajic, V.D. Krstic (AECL)
- 17:00 Plutonium rock-like fuel behaviour under RIA conditions
T. Nakamura, H. Akie, K. Okonogi, M. Yoshinaga, K. Ishijima, H. Takano (JAERI)
- 17:20 Irradiation damage in inert matrix of uranium-free fuels
M. Beauvy, C. Dodane, P. Raison (CEA), S. Bouffard (GANIL)

Parallel Session 3 – Reactors with non-oxide fuels

Room OSGA/UG5

Thursday, 22 October 1998

Chair: T. Osugi (JAERI)

- 14:00 Conceptual core design for uranium metallic fuelled liquid metal reactor
S.J. Kim, Y.I. Kim, Y.J. Kim, C.K. Park (KAERI)
- 14:20 Dynamic behaviour of nitride LMR core during unprotected transients
B.C. Na, J-C. Garnier, P. Lo Pinto, M. Delpech (CEA)
- 14:40 Thermal decomposition behaviour of UN and U_{0.8}Pu_{0.2}N
M. Kato, T. Hiyama, J. Kurakami (PNC)
- 15:20 Review of the Russian molten salt reactor technology studies
V.V. Ignatiev (Kurchatov), K.F. Grebenkine (RFNC)
- 15:40 The concept of nuclear incineration of PWR spent fuel in a transmuter with liquid fuel
M. Hron (Rez)
- 16:00 Study of an accelerator driven system operating in an epithermal spectrum with a constant k_{eff}
P.A. Landeyro, M. Guidotti (ENEA), P. Neuhold (ANSALDO)

Friday, 23 October 1998

5. Closing session

Chair: W. Kröger (PSI)

- 8:30 Future perspectives
W. Kröger, P. Wydler (PSI)
- Session summaries
Session Chairpersons

Panel discussion on international co-operation

H. Fuchs (ATEL, CH), *P.E. Juhn* (IAEA), *H. Mouney* (EdF, F),
M. Nakagawa (JAERI, J), *M. Salvatores* (CEA, F), *Ph. Savelli* (OECD/NEA),
L.C. Walters (ANL, USA)

Introduction to PSI's large facilities

E. Steiner (PSI)

6. Visit to PSI's large facilities

Accelerator complex – *T. Sigg*

PROTEUS critical facility – *T. Williams*

SINQ spallation neutron source – *G. Bauer*

Hot laboratory – *G. Ledergerber*

Note: Preceding the ARWIF'98 Workshop the partners of the "Initiative for Inert Matrix Fuel" held the Annual Inert Matrix Fuel Workshop on Reactor Physics and Materials (PSI, 19-20 October 1998).

Annex 3

QUESTIONS ADDRESSED BY THE WORKSHOP ON ADVANCED REACTORS WITH INNOVATIVE FUELS

(answers in italic)

1. What are the principal issues regarding:

- Plutonium utilisation, uranium resources, and waste management strategy:
 - *Enhancing UO₂ and MOX fuel utilisation in current LWRs to reduce costs.*
 - *Reducing separated plutonium stocks by means of evolutionary and innovative concepts (e.g. highly moderated MOX loaded LWRs, Inert Matrix Fuels (IMF)).*
 - *Preserving fissile inventories as an energy resource for the future (e.g. tight hexagonal PWR lattices, thorium fuels).*
 - *Minimising waste by effectively burning MAs and transmuting long-lived FPs.*
 - *Ensuring energy security for the long-term.*
- Optimum use of facilities and resources:
 - *R&D necessary to keep nuclear option alive.*
 - *International co-ordination of R&D programmes.*
 - *Enhancing co-operation and sharing of resources among highly developed and developing countries.*
- Reactor and fuel technology:
 - *Enhanced safety and proliferation resistance.*
 - *Simplified fuel cycles.*
 - *Material problems of non-oxide fuels for plutonium burning and transmutation.*
 - *Long licensing procedures for new fuels to be taken into account.*

2. Which of the technical issues are:

- Adequately dealt with by industry:
 - *Safety and economics issues relating to operation of existing plants.*
 - *Cost reduction in next-generation plants.*
 - *Fuel behaviour and safety of evolutionary concepts.*

- In the focus of current R&D programmes:
 - *Very high burn-up and other concepts to improve fuel utilisation.*
 - *Advanced oxide-fuel based designs to improve TRU burning capability.*
 - *Non-oxide fuels for plutonium burning and transmutation, enhancing inherent safety and improving public acceptance.*
 - *Resource-efficient, innovative LWR concepts.*
 - *Thorium based concepts.*
 - Still not tackled:
 - *Common strategy for IMF concepts;*
 - *Material problems related to new fuels (e.g. low thermal conductivity of IMF).*
3. Can the existing zero-power reactors and irradiation facilities cover short- and long-term needs? What is the remaining lifetime of the facilities? Do they have to be replaced and/or do we need new facilities?
- *Maintenance and lifetime extension of R&D facilities to be addressed.*
 - *Needs to be clarified in survey report (co-ordination with SESAR group).*
 - *New irradiation facilities needed (by ~2010 all fast spectrum irradiation facilities might be phased out).*
4. How can the available resources best be utilised taking current circumstances into account?
- *Effort to make diverse perspective a pre-requisite for stronger co-ordination of projects .*
 - *Opening and sharing of facilities and experiments.*
 - *Improved cross fertilisation e.g. between reactor physics and material science .*
 - *Strengthening of analytical work in parallel with and as a complement to experimental fuel and materials studies.*
 - *Exploiting knowledge from experimental databases.*
5. How can the dialogue between the R&D community, decision makers and the public be improved?
- *Through the role of nuclear energy in sustainable development (especially vis-à-vis greenhouse gas production), including comparative assessment of energy systems (with emphasis on “renewable”).*
 - *Dialogue on options to improve safety (goal: inherent safety) and waste management.*
 - *Dialogue on energy resources for the long-term future .*
 - *Dialogue on environmental criteria for energy production systems.*

6. What is the role of international organisations:

- In clarifying the issues and fostering consensus:
 - *Co-ordinate and stimulate R&D activities.*
 - *Provide respected source of authoritative advice.*
 - *Harmonisation of views (i.e. taking into account diverse perspectives).*
- As hubs for the exchange of basic nuclear data and computer codes:
 - *Act as central repository of knowledge and expertise.*
 - *Provide the infrastructure for activities such as nuclear data evaluations where individual countries' resources are no longer sufficient.*
- In safekeeping/validating/improving nuclear data and computer codes:
 - *Act as central repository of knowledge and expertise.*
 - *Maintain the state-of-the-art of computer codes, databases and benchmarks.*
 - *Help to establish continuity of expertise across generations.*
- In securing and providing for sharing human and financial resources:
 - *Through collaborative projects.*
- In utilising existing/fostering construction of new facilities:
 - *Monitor status of facilities.*
 - *Facilitate maintenance and construction through co-operative projects.*

7. Which specific issues in the nuclear science domain would benefit from international studies or projects to be co-ordinated by the NEA or another international body?

- *Co-operation on R&D on IMF for transmutation of plutonium and MAs.*
- *Workshop on very-high conversion water-cooled reactors research.*
- *Database for innovative fuels (basic properties and irradiation experiments).*

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