Unconventional options for plutonium disposition

Proceedings of a Technical Committee meeting held in Obninsk, Russian Federation, 7-11 November 1994
The IAEA does not normally maintain stocks of reports in this series. However, microfiche copies of these reports can be obtained from

INIS Clearinghouse
International Atomic Energy Agency
Wagramerstrasse 5
P.O. Box 100
A-1400 Vienna, Austria

Orders should be accompanied by prepayment of Austrian Schillings 100,— in the form of a cheque or in the form of IAEA microfiche service coupons which may be ordered separately from the INIS Clearinghouse.
This publication summarizes discussions and presents selected papers from a Technical Committee meeting that the IAEA convened in Obninsk, near Moscow, Russia, 7-11 November 1994 at the invitation of the Ministry of the Russian Federation on Atomic Energy, and which was hosted by the Institute of Physics and Power Engineering. The meeting focused on the disposition of plutonium produced from the operation of nuclear power plants in areas related to the nuclear fuel cycle. Plutonium is formed in all existing nuclear power plants and the unconsumed part remaining in spent fuel is a generic by-product of nuclear power generation. Over the next 15 to 20 years, a significant amount of plutonium will be produced in nuclear power stations worldwide, adding to amounts already in storage. Additionally, the world’s plutonium stocks are being affected by decisions concerning the management and utilization of plutonium recovered from nuclear weapons which are being dismantled. In this context, national strategies are directed at reducing the stockpiles of separated plutonium worldwide, and in further developing technologies capable of safely and securely using and handling plutonium. The purpose of the IAEA’s Technical Committee meeting was to consider unconventional approaches for plutonium disposition, both from the points of view of the fuel cycle as a whole and the specific types of nuclear fuel being used. The aims were to obtain technical descriptions of these approaches, engineering judgements on their technological status and development, and reports on national experience in this field. The meeting’s results and conclusions are providing valuable guidance for future activities in this subject area.
EDITORIAL NOTE

In preparing this publication for press, staff of the IAEA have made up the pages from the original manuscripts as submitted by the authors. The views expressed do not necessarily reflect those of the governments of the nominating Member States or of the nominating organizations.

Throughout the text names of Member States are retained as they were when the text was compiled.

The use of particular designations of countries or territories does not imply any judgement by the publisher, the IAEA, as to the legal status of such countries or territories, of their authorities and institutions or of the delimitation of their boundaries.

The mention of names of specific companies or products (whether or not indicated as registered) does not imply any intention to infringe proprietary rights, nor should it be construed as an endorsement or recommendation on the part of the IAEA.

The authors are responsible for having obtained the necessary permission for the IAEA to reproduce, translate or use material from sources already protected by copyrights.
CONTENTS

Summary of the Technical Committee Meeting .............................. 7

GENERAL OVERVIEW (Session 1)

Management of plutonium in Russia ..................................... 17
   N.N. Egorov, V.M. Murogov, V.S. Kagramanian, M.F. Troyanov,
   V.M. Poplavski, V.I. Matveev
An overview of the activities of the OECD nuclear energy agency on plutonium ........ 29
   N. Zaribas
Waste managements aspects of (Th, Pu) O
6 fuels ............................ 35
Rational Pu-transmutation for 233U ...................................... 49
   A. Lecocq, K. Furukawa

GAS COOLED REACTORS AND THORIUM ASPECTS (Session 2)

Plutonium destruction with pebble bed type HTGRs using Pu burner balls and
breeder balls .................................................... 63
   K. Yamashita, K. Tokuhara, R. Shindo
The plutonium consumption modular helium reactor (PC-MHR) ..................... 69
   D. Alberstein, A.M. Baxter, W.A. Simon
Advantages and limitations of thorium fuelled energy amplifiers ................. 81
   J. Magill, C.O. Carrol, P. Gerontopoulos, K. Richter, J. van Geel
Mixed plutonium-thorium fuel use in fast breeding reactors - Ecological way
of accumulated plutonium burning ...................................... 97
   E.Ya. Smetanin, V.B. Pavlovich, G.N. Kazantsev, I.Ya. Ovchinnikov,
   B.Ya. Zilberman, L.V. Symnik

CANDU AND LWR (Session 3)

Annihilation of plutonium in CANDU reactors ............................. 103
   D.A. Menelay, A.R. Dastur, R.A. Verrall
Plutonium burning via thermal fission in unconventional matrices ............... 115
   C. Lombardi, A. Mazzola
Reactor physics characteristics of possible fuel materials for plutonium-incinerating LWRs .. 135
   R. Chawla, U. Kasemeyer, J.M. Paratte
Physical and technological aspects of cermet fuel. Application of Pu burning in
WWER reactors ....................................................... 149
   V.M. Dekusar, A.G. Kalashnikov, E.N. Kapranova, A.D. Karpin, I.S. Kurina,
   V.V. Popov
Fuel with carbon matrix for burning plutonium in thermal reactors ................. 155
   E.A. Ivanov, G.N. Kazantsev, I.Ya. Ovchinnikov, F.P. Raskach

MOLTEN SALT (Session 4)

Some ideas about hydrid system concepts .................................. 165
   P.A. Landeyro, A. Buccafurni, A. Santilli
Rational Pu-disposition for 233U production by THORIMS-NES (thorium
molten-salt nuclear energy synergetics) .................................. 169
   K. Furukawa, K. Mitachi, S.E. Chigrinov, Y. Kato, A. Lecocq, L. Berrin Erbay
Neutronic examination on plutonium-transmutation by a small molten-salt
fission power station .................................................. 183
   K. Mitachi, K. Furukawa, Y. Yamana, T. Suzuki, Y. Kato
Study of the possibility of using molten salts for plutonium utilization and actinide transmutation. ............................................. 197
  V.S. Naymov, A.V. Bychkov, O.V. Skiba, P.T. Porodnov

FAST REACTORS (Session 5)

Study on Pu burner fast reactor cores without uranium. ..................... 207
  M. Ishikawa, A. Shono, T. Wakabayashi
Pu burning in fast reactor cores using unconventional fuel without $^{238}\text{U}$ ..................... 219
  I.Y. Krivitski, G.G. Byburin, A.P. Ivanov, V.I. Marveev, E.V. Marveeva
Mononitride mixed fuel for fast reactors. ..................................... 229
  B.D. Ragozkin, N.M. Stepennova, Yu. E. Fedorov, M.G. Shishkov,
  O.N. Dubrovin, L.V. Arseenkov
Plutonium disposal and burning in lead cooled fast reactor .................. 237
  V.V. Orlov
Physics of fast reactor - converter Pu to $^{239}\text{U}$ ......................... 247
  D.N. Ziabletsev, V.G. Ilynin, M.S. Kolesnikova, A.V. Magaev

WEAPONS GRADE PLUTONIUM (Session 6)

Plutonium rock-like fuel integrated R&D (PROFIT) ................................ 253
  T. Muromura, N. Nitani, H. Akie, H. Takano
Combining an accelerator and a gas turbine modular helium reactor for near total destruction of weapon grade plutonium ................................. 263
  A.M. Baxter, D. Alberstein
Physical and technical-economic aspects of weapon grade Pu utilization in HTGRs .......... 275
  Yu.P. Sukharev, A.I. Kiryushin, N.G. Zuzavkov, N.G. Kodochigov,
  A.S. Kudryashov, N.N. Ponomarev-Stepnoi, N.E. Kukharkin,
  E.S. Glushkov, V.N. Grebennik

List of Participants ............................................................................. 285
SUMMARY OF THE TECHNICAL COMMITTEE MEETING

Introduction

In recent years, the IAEA has been working to collect and analyse information on the overall developments and trends in spent fuel processing and plutonium utilization. In its 1993-1994 programme, the Nuclear Fuel Cycle and Waste Management Division, following the current world situation, has been giving more attention to the collection, analysis and exchange of information on both plutonium accumulation and its utilization.

Presently a limited number of countries are involved in plutonium disposition issues, although the subject is gaining greater attention in the light of global developments in the 1990s. This meeting focused on unconventional concepts of plutonium disposition, both from the point of view of the fuel cycle and from the point of view of fuel type, since they have not been reviewed and discussed in depth elsewhere. These unconventional concepts include options which are based on an evolution of technology (e.g. modified fuel designs, metallic fuel, advanced LWRs, reconsidered fast reactors, etc.) and require further development and validation as well as options are based on new concepts still to be further elaborated by appropriate R&D (e.g. PuO₂ dispersion in non-fuel oxides, nitride fuels, combination with a Th cycle, HTRs, etc.). The main emphasis was to collect a technical description of those concepts and to be informed on their status and the schedule for further development and potential implementation.

Plutonium is formed in all existing nuclear power plants and contributes to a large extent to the power generated by any nuclear fuel. The unconsumed part remaining in spent fuel is a generic by-product of nuclear power generation. It is generally designated as "civil" or "reactor-grade" Pu and will be referred to as "RPu" in this document. Disposition of RPu can be envisaged along three scenarios:

1) Leave it in the spent fuel for long-term storage and final disposal. The source term of spent fuel radiotoxicity is dominated during the first 30 to 70 years (depending on fuel type and burnup) by the fission products, thereafter by americium resulting from the decay of Pu, and then by the Pu itself. This scenario is being developed in a number of countries;

2) Strip the spent fuel from most of its fission products and refabricate the resulting mix of U and Pu into fuel. The synergistic fuel cycles involving light water (cooled and moderated) reactors (LWRs) and pressurized heavy water reactors (CANDUs), being studied in Canada and the Republic of Korea, and the recycle based on pyroelectrochemical technology for liquid metal (cooled) fast reactor (LMFRs) mixed oxide (U, Pu)O₂ (MOX) fuel, already demonstrated on a pilot-scale in Russia, are typical examples;

3) Separate uranium, plutonium and fission products by reprocessing and recycle U and Pu. This scenario is being industrially pursued by a number of countries. Further developments aim at minimization and improved management and disposition of the minor actinides (MA) to further reduce the radiotoxicity of the nuclear cycle wastes.

Whatever the scenario, with storage time, the radioactivity of the stored material increases, and its technical value as a fuel decreases. The rate of ageing of RPu depends on its type: it is lower for Pu from low burnup gas cooled reactor (GCR) fuel and highest for Pu from high burnup (or MOX) LWR fuel. Aged RPu can if necessary be cleaned from its Am (which must then be disposed) or diluted by fresher Pu.

In April 1993, an IAEA Advisory Group Meeting on Problems Concerning the Accumulation of Separated Pu evaluated inventories of separated RPu, stating that about 120 t would accumulate early in the next century. A more recent update by the IAEA projected 160 t, and various other evaluations have ranged from 110 to 200 t in the same period. This inventory buildup is due to a
shortage of available MOX fabrication capacity. National strategies, now being pursued, will result in a subsequent reduction of the stockpile of separated RPu worldwide, and technologies capable of safely and securely using and handling Pu are being further developed.

The decision to dismantle nuclear warheads and eliminate military Pu stockpiles will add to the RPu stockpile more than 100 t weapon-grade plutonium ("WPu"). It should be mentioned that WPu has a negligible ageing rate and could therefore be stored almost indefinitely without technical inconvenience. However, nontechnical considerations call for early disposition of WPu, at least sufficiently advanced to make further military usage unattractive. This could for instance be done without delay by blending WPu, as soon as available, to aged RPu, thereby rejuvenating the latter. Different locations and ownership of the two types of Pu make such simple and straightforward solutions inapplicable, except for restricted amounts of RPu and WPu.

**Objectives of plutonium disposition**

All Pu utilization options pursue multiple objectives. New concepts of plutonium disposition place emphasis on one or more of the following objectives:

- elimination of the Pu stockpiles;
- maximum consumption of Pu (sometimes even without production of new Pu);
- reduction of radioactive wastes;
- reduction of personnel exposure throughout the reactor cycle;
- recovery of energy resources.

When several objectives are to be considered, compromise solutions usually have to be adopted. As a result, several alternatives are presently being developed.

No single scenario can be universal. The selection is dictated by national policy, bilateral (or multilateral) agreements, availability and/or accessibility of the technology, specific sensitivities of public opinion, etc. It is therefore recognized that different scenarios will be selected or considered in the various countries.

In assessing options and developing scenarios for Pu disposition, several criteria must be considered, some of technical nature, some based on political arguments. They include:

- existence or potential development of the technology: fuel processing (conversion, fabrication, reprocessing and/or waste vitrification) of the material, fuel performance, reactor and auxiliary facilities, transportation, etc.;
- licensing consideration, including the required database;
- timeliness, taking into account not only the annual capacity of Pu disposition but also the time schedule for development, demonstration and industrial implementation;
- safeguards, proliferation resistance and, if addressed by the scenario, irreversibility of the demilitarization;
- energy resource utilization;
- radiotoxicity of the products and of the high level waste arising from the scenario;
- amount of radioactive waste from the fuel cycle and reactor operation and from dismantling of those nuclear installations;
- environmental impact;
economics, including the development, demonstration and first-of-a-kind costs;
- public acceptance or acceptability.

Technical options

The alternative options to MOX fuel in LWRs, LMFRs and advanced thermal reactors (ATRs) are based on Pu in inert matrixes or on fuel type incorporating Th. The first group usually necessitates the simultaneous use of burnable or removable absorbers, which deteriorate the neutron economy. The utilization of Th results in the production of $^{232}$U and thereby conserves the energy resources to a variable degree, depending on the nuclear characteristics of the option. The danger of creating thereby a new weapon-grade material can be intrinsically avoided by tuning the generation of $^{231}$U towards significant contents of $^{232}$U, whose daughter products are high energy gamma emitters. While this contributes to proliferation-resistance, it also leads to inconveniences in the subsequent utilization of this fissile material: heavy investment in reprocessing and fabrication plant development and construction and increased exposure level of the fuel cycle and reactor personnel. Some options consider the flexibility to utilize either type of fuel and even to return to conventional MOX utilization. Amongst all the fuel options, the ones based on oxides are probably applicable on the shortest time-scale: foreseeing 10 years development and data acquisition (including a representative performance data-base), followed by 10 years for establishing an industrial fabrication capacity is not unreasonable.

While the time-scale for fuel cycle accommodations and the related investments are important to take into consideration, the time-scale and investments necessary to develop new reactor systems are potentially many times larger. But simultaneously, it is the reactor system that plays a predominant role in the annual rate of Pu disposition and in the Pu transmutation level. Depending on where the emphasis is placed, a large variety of options can be considered. Various aspects of the use of LWR, CANDU, LMFR, high temperature gas cooled reactor (HTGR) and molten salt (cooled and fueled) reactor (MSR) NPPs were described at the meeting, as well as concepts based on accelerator-driven subcritical reactors, to be utilized either as a basic installation or a “post-combustion” installation.

Conclusions and recommendations

The several objectives being pursued and the multiple criteria being considered are difficult to reconcile in any single option proposed or considered for Pu disposition.

Conventional options (MOX fuel in LWRs, LMFRs and ATRs) already applied industrially are able to cope with the disposition of separated Pu stockpiles, provided sufficient MOX fabrication capacity is deployed. Some of the other objectives and criteria are not met, or only partially met.

The unconventional options being suggested or developed aim at addressing objectives and/or criteria not met by the present MOX utilization routines. In turn, none of those unconventional options meet all objectives and criteria. Certain systems place emphasis on flexibility to switch the option(s) to meet changing emphasis with time on particular objectives or criteria. Even so, the technical experts evaluating or developing unconventional options feel uneasy about the absence of general consensus on the ranking of the objectives and criteria. It is a concern worthy of being addressed, since some of those options required considerable time and budget for development, demonstration and industrial implementation. For all these reasons, the effectiveness, cost-benefit balance and timeliness of most of the presented options are difficult to evaluate.

A rather long list of proposals and recommendations were discussed at the working groups and plenary session, and the main ones are presented below:
1. It is important to evaluate unconventional alternatives being considered for Pu management. This meeting dealt with many technical aspects of various potential alternatives, enabling each of them to be put in perspective. Regular reviews of this field of development were considered highly valuable at the international level.

2. While all the presentations provided a good insight to technical aspects, most of the papers did not provide a clear picture of the time-scales and costs to be considered for the development stage, the demonstration stage, and the industrial implementation stage (after due evaluation of lessons learned from the demo period). These aspects need to be addressed in the future.

3. Future working groups of this type should additionally discuss a few specific topics, including, for example, the use of Th in conjunction with Pu disposition, selection of the inert diluent and radiotoxicity issues.

4. Quantitative criteria should be developed to assess radiotoxicity and proliferation-resistance requirements.

5. International collaboration is recommended in the near-term, for development of a material technology database for candidate inert-matrices, and to pursue a suitably formulated, numerical benchmark exercise for neutronics calculations relevant to particular reactor systems.

6. Further work should be conducted to prove the acceptability of inert carrier materials for fast reactor service. Experimental investigations are necessary to verify the characteristics of conceptual core designs with different compositions. In addition to R&D on in-core performance characteristics, it is recommended that work be continued to ensure the technical ability, at some time in the future, to reprocess used inert-carrier fuel.

7. Considering the inadequate experimental data base on accelerator-driven components, the performance testing and development of targets should proceed to some confirmation stage.

The meeting featured six sessions, which are summarized below:

Session 1. General Overview, chaired by V. Murogov and H. Bairiot

All presentations pointed towards the accumulation of separated Pu from the reprocessing of thermal reactor fuel, and to a smaller extent, fast reactor fuel. The decision to dismantle the nuclear warheads will add to separated RPu an equivalent quantity of separated weapon grade Pu. The concerns about radiotoxicity of the Pu itself and of its decay products and the desirability to prevent misuse of weapon grade Pu have promoted initiatives for considering several options for Pu disposition. While use as MOX fuel for LWRs and LMFRs is developed on an industrial scale, alternative options are being considered in order to decrease separated Pu accumulation or to reduce Pu production even more.

Exploration of the long times needed for any alternative option strongly suggests that, whatever long-term path is taken, safe, secure and verifiable storage of separated plutonium is essential. The choice of Pu management is not only related to technological, safety and economic considerations but is also influenced by non-proliferation, national policy, and public acceptability implications. As a result, international organizations have reviewed and are currently reviewing several related aspects. The IAEA is maintaining and updating a database and forecast of civilian Pu generation, separation and utilization rates, as well as fuel performance updates. In this regard, the IAEA is in the process of publishing topical Pu-related safety standards. The OECD/NEA has been involved traditionally in reviewing the economics and logistics of the fuel cycle, in general, and has pursued and is pursuing activities in these fields on MOX fuels. Currently, an expert group is reviewing and evaluating the broad technical questions concerning Pu management, and another group is reviewing the physics of Pu recycling.
As an example of the constraints and options typical for each individual country, the Russian Pu management perspectives were presented: the availability of LMFRs and their good performance, on the one side, and, on the other side, the quite large development program to be considered for MOX utilization in WWERs favour, as first priority, the implementation of an industrial capacity of LMFR MOX fabrication and the construction of additional LMFR plants, to utilize the Pu in the most expeditions time-scale.

The utilization of Th in conjunction with burning Pu has been considered by several organizations, since the excess reactivity of Pu and its evolution can be controlled by the $^{233}$U bred from Th without producing further Pu. The use in CANDUs of Pu fuel with either ThO$_2$ or inert carrier matrices is under active investigation. One presentation focused on assessment of (Th-Pu)O$_2$ fuel from the perspective of long term waste disposal by comparison with the existing repository design for used UO$_2$ fuel: the only issues requiring further investigation are segregation during fabrication and build-up of excess oxygen during irradiation. The new THORIMS-NES strategy is a candidate for a Th-Pu cycle maintaining the world energy inventory and effectively consuming Pu.

Session 2. Gas Cooled Reactors and Thorium Aspects, chaired by R. Chawla and E. Kudriavtsev

The four papers in this session (two HTGR-related and one each concerning fast reactors and the energy amplifier concept, respectively) considered achieving high levels of Pu elimination by discontinuing the use of $^{238}$U. There was general consensus that, for both reactor-grade and weapon-grade plutonium, a high burnup capability needs to be aimed at relative to conventional methods. The feeling was expressed that effective plutonium disposition will take several decades to complete and that, as such, certain novel (unconventional) methods would not necessarily extend the overall disposition time-frame.

Three of the four papers considered the control of excess reactivity for the fresh core through the replacement of $^{238}$U by thorium. It was recognized that the latter's usage in the context of plutonium reduction would raise new technical issues with regard to non-proliferation and spent-fuel radiotoxicity. Differences in viewpoints were linked, on the one hand, to the desirability of generating $^{233}$U as a valuable thermal reactor fuel and, on the other hand, to the perceived replacement of one type of fissile inventory by another. Proliferation concerns associated with $^{233}$U were expected to be mitigated by its easier detectability (due to the high-energy radiation from $^{232}$U), as well as by the denaturing possibility with $^{238}$U. Regarding long term radiotoxicity, one of the points made was that the Th/$^{233}$U cycle, considered in once-through operation, was not necessarily advantageous relative to the $^{238}$U/Pu cycle.

Session 3. CANDU and LWR, chaired by R. Page and L. Zabudko

With LWRs likely to continue to dominate the nuclear energy scene, it is important to consider their potential for reducing plutonium inventories worldwide. The current use in LWRs of MOX fuels constituting 30 to 40% of the core, while effectively decreasing the separated Pu inventory, does not, however, represent a net reduction of plutonium inventories as approximately the same amount of new plutonium is generated as that consumed.

Designs for 100% MOX cores are being developed, and effective Pu reduction should be achievable with them. The build-up of new plutonium from $^{238}$U present in MOX, however, remains disadvantageous in the perspective of eliminating further Pu production and consequently designs of plutonium burning fuels are being considered without uranium. The fuel would consist of plutonium dispersed within an inert matrix, e.g. ZrO$_2$, Al$_2$O$_3$, MgO, zirconium alloys, or graphite. The use of such a fuel in water reactors would allow for an overall total plutonium reduction of 60 - 70% with a fissile $^{239}$Pu reduction of the order of 90 - 95% from a single reactor cycle. The absence of uranium (or Th) results in core physics effects which need to be investigated in detail in order to ensure that controllability and safety are not significantly affected. Scoping investigations indicate that the use of
appropriate burnable poisons, possibly combined with advanced fuel concepts developed in other
contexts (such as cermet fuel containing small amounts of fertile material), could provide a suitable
route towards implementation in LWRs of current-day design.

The CANDU reactor system, with its high neutron economy and well-thermalized neutron
spectrum, reduces $^{239}$Pu formation by increasing the fission-to-capture cross-section ratio of $^{239}$Pu and
eliminates the need for reactivity suppression through rapid on-line refueling. Using inert matrix fuels
and in the absence of $^{238}$U, it can burn 90% of the initial fissile inventory in a once-through mode
cycle and achieve further reductions through a multiple shuffling. The absence of $^{238}$U has a limited
effect on the dynamics and thus can be addressed with the use of current reactor technology. Certain
aspects such as the refueling schedule require further clarification.

The development of LWR and CANDU inert-matrix, Pu-incinerating fuels is still at a
relatively early stage, with work on candidate matrix materials having just started. It will be necessary
to implement a major fuel design and testing programme of considerable duration before the fuel will
be available for full-scale application. There was some indication that possibly unrecognized
technological difficulties may exist with some of the candidate materials being considered. Valuable
test data of relevance could already be available in individual laboratories, and it is highly desirable
that this information be pooled before major new efforts are undertaken.

There was some difference of opinion regarding the virtue of once-through schemes. Inert
matrices not amenable to reprocessing (intended to provide a spent-fuel form for direct disposal as
waste) would, on the one hand, offer greater proliferation resistance but, on the other, make further
reduction of radiotoxicity (e.g. via recycle in fast reactors) more difficult.

Session 4. Molten Salt, chaired by D. Alberstein and G. Pshakin

All papers presented in this session addressed approaches to Pu elimination that involved use
of molten salt systems - some in conjunction with an accelerator-driven component, others without.
All systems presented make use of non-aqueous, molten salt processing to maximize plutonium
reduction and minimize waste volume production. In all but one of the papers use of thorium for
production of new fissile material was described. In the remaining paper, the focus was only on
plutonium consumption, without production of new fissile materials.

The design and overall Pu elimination and $^{233}$U production capability of an accelerator-driven
molten salt breeder reactor and a molten salt fission power station were presented to provide more
explanation of the THORIMS-NES concept presented in Session 1. In this system rather than simply
burning out excess plutonium inventories, the plutonium can also be used to produce new fissile $^{233}$U.
The applicability of THORIMS-NES in a scenario for plutonium elimination and initiation of a
thorium era on a large scale was evaluated, and required installed capacities of each reactor were
quantified.

With regard to the accelerator-driven component of these systems, evaluations to date have
addressed feasibility, but they are supported by minimal experimental data. Proposals for target
performance testing have been prepared but not yet funded.

With regard to molten salt processing systems, previous experience with these systems has
been accumulated from the Oak Ridge Molten Salt Reactor developmental activities. Systems under
consideration today differ in some details from the Oak Ridge system. The system evaluated and
presented by the RIAR was derived from thirty years of investigations of fast breeder reactor MOX
fuel reprocessing via chloride molten salts. It indicates a possibility of applying pyroelectrochemical
and pyrochemical techniques. The scientific bases of this approach have been formulated, and the
initial aspects of the closed fuel cycle have been developed. It was indicated that, after it is adapted
to molten fluoride salt processes, this process can be included in the overall process system for the
molten salt reactor. It was also agreed that molten salt processing systems should proceed to the
The molten salt systems with thorium fertile material can effectively reduce excess plutonium inventories but also offer an opportunity to redirect the focus of nuclear energy programs from the uranium/plutonium cycle to a closed thorium/uranium cycle. Efforts are beginning to organize and increase worldwide cooperation in the evaluation and development of molten salt reactor technology. To the extent that fluoride systems are being considered for use in molten salt reactor systems, attention is being focussed on properties of fluoride molten salts such as actinide electrochemical properties and behaviour of rare earth elements in these melts.

Session 5. Fast Reactors, chaired by D. Menelay and V. Kagramanjan

Several countries have developed LMFR technologies, and about 20 liquid metal cooled reactors have accumulated more than 200 reactor-years of operating experience over the past 25 years. The special characteristics of these reactors can be utilized to enhance the capability for plutonium consumption (including Pu of low fissile content) as well as to meet other future objectives.

In addition to plutonium disposition, it has been suggested that long-term disposal of wastes from all types of reactors can be improved by consuming minor actinides in addition to plutonium. This session was devoted to examination of the technologies needed to accomplish these tasks using fast reactors. Most LMFR designs utilize sodium as coolant, and this is taken here as the reference concept for fast reactors. The alternate coolant proposed was lead; from the point of view of plutonium disposition this concept is similar to the reference concept and so will be considered equivalent.

Plutonium can be efficiently and effectively dispositioned in fast reactors, first by removing the "blanket" uranium. This capability has already been demonstrated. Plutonium consumption can be further increased in a MOX core, for example, by decreasing the fuel volume fraction, increasing core leakage, or by adding neutron absorbing materials.

In addition to the above, it may be possible to remove the plutonium-producing isotope $^{238}\text{U}$ from fuel in the core region so that the net plutonium consumption increases significantly. Three papers in this session proposed this approach. Such a core can achieve a very high rate of plutonium consumption with very low coolant void reactivity. Some design issues remain to be resolved; in particular, the large reactivity loss with burnup and the small Doppler effect. Alternative to the use of inert matrix carriers, one author proposed replacement of the core $^{238}\text{U}$ with $^{225}\text{Th}$. A number of challenging problems remain to be solved for this option.

Options seeking more complete annihilation of plutonium and minor actinides in fast reactors require reprocessing of the spent fuel at the end of each operating cycle. The design characteristics of these reactors are changed by core reconfiguration, and careful examination is required to ensure the performance and safety of the system are not compromised. Scoping studies have given sufficient confidence that these goals can be achieved.

The search for suitable "carrier" materials for plutonium fuel was a central theme of the session. A considerable amount of work has been done on inert matrix materials, especially in Russia. Preliminary irradiation test results were reported, mainly for mono-carbide and mono-nitride forms as well as for porous zirconium carbide saturated with fuel oxide. Plans are underway in France for irradiation tests of solid solutions, cermet, and cercers (ceramic-ceramic). Final selection will require several years of work, given the many variables to be investigated and the long times required for irradiation and examination. It appears that opportunities exist for international collaboration in some aspects of this work.
All three papers presented at this session addressed destruction of excess WPu. Two papers focussed on high temperature gas cooled reactors, and one paper on light water reactors. All approaches aimed at final disposal of spent fuel with no further reprocessing. Issues with regard to radiotoxicity of spent fuel were not addressed in any of these papers.

In the LWR presentation, preliminary inert matrix fuel development activities were described, and a schedule for further development was presented. A number of technical issues regarding fuel behaviour under both normal and accident conditions (both design basis and beyond design basis accidents) were identified as needing further evaluation. These include fuel matrix/coolant interactions, fuel matrix/cladding interactions, general irradiation behaviour and others.

Both gas cooled reactor presentations indicated that this reactor is capable of achieving high levels of excess weapons grade plutonium reduction. There are some data from irradiation of plutonium coated fuel particles in HTGRs that confirm the capability of the fuel to achieve high burnup. The presentation on combining the gas cooled reactor with an accelerator acknowledged that this approach would require more development work than the much more well developed approach of using only the gas cooled reactor for plutonium destruction.

Acknowledgements:

The success of this meeting was in part due to the able leadership provided by H. Bairiot of Belgium.

ABBREVIATIONS

AGM  advisory group meeting
ATR  advanced thermal reactor (Japan)
CANDU  Canada deuterium-uranium (reactor)
GCR  gas cooled reactor
HTGR  high temperature gas cooled reactor
IPPE  Institute of Physics and Power Engineering (Obninsk)
LMFR  liquid metal (cooled) fast reactor
LWR  light water (cooled and moderated) reactor
MA  minor actinides (Am, Cm, Np, etc.)
MOX  mixed oxide (U, Pu)O₂
MSR  molten salt (cooled and fueled) reactor
NPP  nuclear power plant
RIAR  Research Institute of Atomic Reactors (Dimitrovgrad)
RPu  reactor-grade Pu
THORIMS-NES  thorium molten salt nuclear energy synergetics (based on molten salt converter, accelerator-driven molten salt breeder and dry processing plants)
WWER  water moderated, water cooled reactor (Russia)
WPu  weapon-grade Pu
SESSION 1: GENERAL OVERVIEW
Abstract

The Russian concept of plutonium management (both civil and weapons) is based on the postulate of the outer fuel cycle closure, necessity to enhance fuel efficiency, and decreasing radioactivity of disposed long-lived wastes. The key problem for all countries considering recycling options is now to optimize for the short-, medium- and long-term. Short-term plutonium management in Russia is based on safe and reliable storage of separated civil plutonium and ex-weapons plutonium until it can be used in reactors. Using of accumulated plutonium in fast reactors is a question of medium-term plutonium disposition options. Long-term options to be defined consider use of plutonium in VVER-type reactors and the burning of plutonium excess and minor actinides in fast reactors with new cores.

1. INTRODUCTION

The increase in the accumulation of plutonium stocks in the process of today's reactor operations, as well as the plutonium released as a result of armament reduction evokes steadily raising concern of Russian and foreign societies about its future. Expected quantities of civil plutonium accumulated in the spent fuel of now functioning Russian NPPs with a life time of 30 years are given in the Table I. Also in this table are the quantities of ex-weapons plutonium expected to be released in Russia.

The problem with plutonium is especially acute due to the public desire to make the disarmament process irreversible. There are two opposing points of view on plutonium. One view is to regard it only as a waste that should be disposed in deep geological formations. The other view insists on the need of plutonium recycling. Plutonium recycling increases national energy resources and decreases the radioactivity of long-lived nuclear wastes. From our standpoint this divergence in views extends from the difference in the status of fuel cycle technology and the comprehension of the role of nuclear power in particular countries.

If a country's view of the future of nuclear power is optimistic and fuel cycle technology related to plutonium recycling is sufficiently developed to:

- follow non-proliferation policy;
provide a safe and ecologically acceptable plutonium based fuel cycle;

be economically competitive,

then plutonium is considered a national asset. Otherwise, it is a permanent global threat. The key problem for all countries considering recycling options is now to optimize for the short-, medium- and long-term.

In Russia we are considering the recycling option along with these additional considerations:

-growing plutonium inventories because of:
  - fast reactor development delay and
  - arms reduction;

-good experience outside of Russia using plutonium in thermal reactors;

-increased public anxiety on long-lived radiotoxic nuclear waste;

-changes in the Russian economic and political environment;

-great uncertainty in the future of nuclear power development;

<table>
<thead>
<tr>
<th>Reactor type</th>
<th>Power, GWt(e)</th>
<th>Pu¹, t</th>
<th>Pufis², t</th>
</tr>
</thead>
<tbody>
<tr>
<td>VVER-440</td>
<td>3</td>
<td>27 (3,7)</td>
<td>20 (16)</td>
</tr>
<tr>
<td>VVER-1000</td>
<td>7</td>
<td>55 (9,1)</td>
<td>40 (33)</td>
</tr>
<tr>
<td>RBMK-1000</td>
<td>11</td>
<td>76 (8,3)</td>
<td>45 (37)</td>
</tr>
<tr>
<td>BN-600</td>
<td>0,6</td>
<td>12 (0)</td>
<td>11 (11)</td>
</tr>
<tr>
<td>Weapons Pu</td>
<td>-</td>
<td>100 (0)</td>
<td>95 (95)</td>
</tr>
<tr>
<td>In total:</td>
<td></td>
<td>270 (21)</td>
<td>211 (193)</td>
</tr>
</tbody>
</table>

¹ in brackets mass of Pu-241+Am-241
² in brackets counting on Pu-241 decay after 30-years storage
-readiness of political, scientific and industrial structures of western countries and Japan to help address plutonium utilization in Russia.

We suppose that in the long-term perspective, optimum plutonium recycling can have many identical features in Russia and other countries. As for the short- and medium-term, we think that Russian plutonium utilization plans could differ to some extent from those of other countries. The aim of this presentation is to clarify this point.

2. EXPERIENCE GAINED WITH PLUTONIUM

Russia began using plutonium as a nuclear fuel in the second half of '50s. In 1957 a core was fabricated of metallic plutonium alloy for the pulsing fast reactor IBR-30. In 1959-65 plutonium dioxide was made for the BR-5 and IBR-2 reactors.

Systematic studies of plutonium-bearing fuel began in 1970 in the BOR-60 reactor. This work were conducted in the framework of the fast breeder reactor development program and was aimed at the most effective use of plutonium to expand nuclear power fuel resources.

Until now, two cores of weapons grade plutonium have been tested in the BR-10 experimental fast reactor. Large batches of MOX-fuel pins made by different technologies with plutonium of various isotopic composition have been tested in the BOR-60 research reactor. For many years this reactor has been operated recycling its own plutonium.

With continued successful MOX fuel tests, we expanded the scope of the tests to the prototype fast reactors BN-350 and BN-600. Both reactors have been fueled from the very beginning with enriched uranium. The PO "Mayak" (Chelyabinsk) semi-industrial MOX fuel fabrication installation "Paket" provides these reactors with MOX fuel. Mayak has an annual capacity of 10 subassemblies. In the BN-350 reactor the tests have been performed with the subsequent investigation and chemical reprocessing of test fuel subassemblies with MOX fuel (350 kg of weapons grade plutonium). More than 2000 such fuel elements have been fabricated and tested in the BN-350 and BN-600 reactors. Post-reactor investigation showed that fuel element endurance was not exhausted with a burn-up of 9-11% h.a.

Serious studies were performed in Russia on vibrocompacted fuel. To this end granulated fuel, obtained by different methods, was used. Among them is a method of electrochemical uranium oxide and plutonium oxide coprecipitation. All the fuel elements loaded into the BOR-60 reactor have been fabricated using this technology. Also, subassemblies have been tested in the BN-350 and BN-600 reactors.

Successful MOX fuel testing for BN-type fast breeder reactors made it possible to begin design and construction of a small series (3-4 units) of fast, plutonium breeder reactors, BN-800 on the South Urals and Beloyarskaya sites. A special plant for MOX fuel fabrication ("Complex-300") is planned to be built to provide fuel for the operation of these reactors. It will produce a maximum
of 900 subassemblies annually. Construction of the first two BN-800 units and of Complex-300 has been suspended due to financial difficulties. The reactors are in the initial stage of construction while construction of Complex-300 is 50% complete.

Works is planned to back fit the "Granat" and "Paket" pilot installations at the Mayak site to meet the latest safety and environmental requirements of the Governmental Regulatory Organization (Gosatomnadzor).

Studies of plutonium utilization in thermal reactors are only now beginning in Russia. For a long time in Russia, much longer than in other countries, plutonium was meant to be used solely in fast breeder reactors as the most effective use of this nuclear material.

Now work is underway to study the fabrication technology of experimental subassemblies with MOX fuel for the VVER-1000. Critical assembly "SUPR" is being constructed at IPPE, Obninsk.

General information on MOX production in Russia is presented in the Table 2.

The nuclear fuel closed cycle realization started with the commissioning of the first Russian reprocessing plant, Mayak, in 1976. The RT-1 plant is a multi-purpose enterprise which provides reprocessing of spent fuel from VVER-440 reactors, fast converter reactors (BN-350 and BN-600), icebreaker and submarine transport units, research reactors, and other power units. The plant capacity is 400 t/year in terms of the main fuel type, that is, that of the VVER-440 reactor, making possible to reprocess the fuel from not only Russian NPPs but also from foreign reactors of the same type. About 3000 tons of spent fuel have been reprocessed in this plant.

**TABLE II. MOX PRODUCTION CAPACITY IN RUSSIA**

<table>
<thead>
<tr>
<th>Plant/Facility</th>
<th>Reactor</th>
<th>Annual capacity</th>
<th>Production in 1992</th>
</tr>
</thead>
<tbody>
<tr>
<td>&quot;Paket&quot; at Mayak, Chelyabinsk</td>
<td>BN-350</td>
<td>10-12 FAs</td>
<td>4 FAs</td>
</tr>
<tr>
<td></td>
<td>BN-600</td>
<td>300 kg MOX</td>
<td>100 kg MOX (about 20% Pu)</td>
</tr>
<tr>
<td>&quot;Paket&quot; (modified) since 1993</td>
<td>BN-600</td>
<td>40 FAs</td>
<td>1 tonne MOX</td>
</tr>
<tr>
<td>Facility at RIAR (Dimitrovgrad)</td>
<td>BOR-60</td>
<td>1 tonne MOX</td>
<td>600 kg MOX</td>
</tr>
<tr>
<td></td>
<td>BN-600</td>
<td>(vibropack)</td>
<td></td>
</tr>
<tr>
<td>Plant at Chelyabinsk complex (50-60% complete)</td>
<td>BN-600</td>
<td>60 tonnes HM</td>
<td></td>
</tr>
<tr>
<td></td>
<td>BN-800 (VVER-1000)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Plant in Krasnoyarsk</td>
<td>VVER-1000</td>
<td>planned for future</td>
<td></td>
</tr>
</tbody>
</table>

Note: FA=fuel assembly; HM=heavy metal
Spent fuel reprocessing produces the main product, enriched uranium and civil plutonium. When the RT-1 plant is under full load, plutonium production is 2.6 tons per year. Last year's plutonium production dropped to 0.6 tons.

The total quantity of extracted civil plutonium stored at the Mayak site is about 30 tons. This plutonium is stored in the form of dioxide to be used in fast reactors. The main problem of this forced, protracted storage of civil plutonium is decay of the shot-lived fissile isotope plutonium-241 into americium-241. This isotope is very troublesome from an ecological point of view. About 10% of the initial unloaded plutonium is plutonium-241.

A new powerful radiochemical plant RT-2 is under construction not far from Krasnoyarsk. Its principle objective will be to reprocess VVER-1000 spent fuel. In its first phase, to be put in operation after 2005, it will reprocess 1500 tons of fuel per year. The plant cooling pond already contains 1100 tons of VVER-1 spent fuel subassemblies, while about 1000 tons of this fuel are currently at other NPPs.

Accumulation of the above mentioned plutonium at the Mayak site, considerable scales of planned plutonium extraction at the RT-2 plant of up to 10 tons of plutonium per year, and the expected release of ex-weapons plutonium of up to 100 tons when intensive deployment of BN type reactors in the nearest future has a low probability, require corrections to be made in the earlier existing strategy of plutonium utilization mainly in fast breeder reactors. There are no doubts that in the near-term management measures are to be taken for reliable plutonium storage. In addition to the storage of civil plutonium, storage for released ex-weapons plutonium should also be arranged. At the same time it is obvious that such storing should not be too long because of economic, ecological, and political considerations.

3. MANAGEMENT OF SEPARATED CIVIL AND EX-WEAPONS PLUTONIUM

3.1. Medium-Term Option

The Ministry of the Russian Federation for Atomic Energy has developed a basic concept on the management of extracted civil and released ex-weapons plutonium based on the following leading principles:

-Russian experience of plutonium management should be maximized;

-plutonium diversion resistance should be considered an important criteria;

-proposed medium-term option of plutonium management should be ecologically and economically acceptable and lead to the development of a long-term fuel cycle option.

Separated plutonium utilization based on a "nuclear center" at the Mayak, including the Shop-300 plant for MOX-fuel manufacture and an NPP with the 3 BN-800 units together with the operating RT-1 plant for chemical processing of
uranium spent fuel, fully meets these criteria. Construction of this center has already been foreseen by the Russian nuclear program. The main purpose of these reactors was to utilize civil plutonium accumulated at Mayak as result of chemical processing of uranium reactor spent fuel. The BN-800 reactor design is described by a breeding ratio (BR) of about 1. That is, it was expected that these reactors will use plutonium from thermal reactors as fuel for their first loading only, changing to their own plutonium in the future. For this purpose it is foreseen to organize BN-800 spent fuel processing.

Calculations made show that the problem of not only the accumulated civil plutonium at the RT-1 but also that of all released ex-weapons plutonium could be solved by creating a nuclear center at Mayak with the addition of one BN-800 reactor to be constructed on the Beloyarskaya site. It would only be necessary to postpone chemical reprocessing of the BN-800 reactor spent fuel until a considerable portion of already released civil and weapons-grade plutonium will have been involved in the fuel cycle.

Fissionable material nonproliferation is addressed first, by limitation of released civil and weapons-grade plutonium travel within the militarized zone on the Mayak site and second, by minimization of the time required to

![Pu balance at the PO "Mayak" as a function of BN-800 reactors number](image)

*Figure 1. Pu balance at the PO "Mayak" as a function of BN-800 reactors number (one through cycle, RT-1 capacity 2.6 t Pu/a)*
transform already released plutonium into spent fuel. Radioactive fission products present in the spent fuel serves as a reliable barrier against diversion, and the complex isotope composition of civil plutonium makes warheads very difficult to manufacture.

In order to decrease plutonium content in the spent fuel and the costs of spent fuel subassembly storage, an improved BN-800 reactor design has no blankets.

Weapons-grade plutonium consumption for one BN-800 reactor produces about 1.6 tons a year. If the BN-800 reactors were put in operation according to the program, all civil plutonium accumulated at Mayak and all released weapons-grade plutonium could be "disarmed" (that is, transformed into spent fuel) during the first two or three decades of the next century. (See Figure 1). Thus, weapons-grade plutonium utilization in the context of the basic concept is inseparably linked with the realization of the program on fast reactors using traditional MOX fuel.

In general, it is possible to start "disarming" ex-weapons plutonium using a light water reactor as suggested by the United States. For Russia, however, this solution is difficult to reach for several reasons. First, Russia has only just begun its experimental research using MOX fuel VVERs. We are considerably behind our foreign partners in this field. There is no thermal reactor operating in Russia designed to use MOX fuels. Safety levels at the VVERs, even operating with uranium, do not meet long-term safety requirements for new generation reactors. Therefore, licensing a changeover from uranium fuel assemblies to MOX fuel assemblies at operational VVERs is highly doubtful.

Second, a changeover fuel to MOX-fuel loaded core VVERs "disarm" up weapons-grade plutonium would require twice as the number of BN-type reactors because annual consumption levels of plutonium for fabricated fuels for VVER and BN reactors are different. In France, for example, they load one-third of the core with MOX assemblies. This means that five-six times more VVERs will be needed than BNs.

Third, the question of radiotoxicity of spent fuel is important. It is well-known that long-lived isotopes in spent fuel such as americium, neptunium, and curium complicate both MOX-fuel recycle technology and finding solutions to the long-term waste disposal. To a great extent, these problems are attributed to the accumulation of plutonium-241 in spent fuel. The specific radiotoxicity of plutonium-241 is 40 times higher than that of the basic isotope plutonium-239. During storage, the plutonium-241 isotope is transformed into the toxic americium-241 with a half-life of 433 years. This isotope is the main contributor to the radiotoxicity of transuranium elements in the spent fuel after the decay of short-lived fission products. During the operation of light water reactors with uranium, about 250 kg/GWt(e)*year of reactor grade plutonium are built up. About 30 kg of this mass is plutonium-241. "Disarming" ore burning-up weapons-grade plutonium in thermal reactors increases three-fold the annual build-up of plutonium-241, compared with burning uranium-based fuel in VVERs. If this spent fuel is stored for the long-term, a considerable amount of the plutonium-241 will be transformed into americium-241, highly complicating further utilization of plutonium and waste disposal.
Fourth, the burning of weapons-grade plutonium at VVERs may produce several times more minor actinides as compared with the VVERs using uranium-based fuel. The burning of the basic isotope plutonium-239 during weapons-grade plutonium utilization at Mayak facilities would result in an accumulation of nuclides with a total radiotoxicity three times higher than that of the VVER/uranium variant.

The situation is quite different however, if we use BN-type reactors to utilize weapons-grade plutonium. The radiotoxicity of the "disarmed" plutonium does not, for all practical purposes, exceed that of the initially loaded plutonium. Figure 2 shows efficiency indices for weapons-grade plutonium utilization in VVERs and BNs. The VVER/uranium radiotoxicity index is given for comparison. We can see that BN gives the best radioecological effect; it is the most effective burner of plutonium-241. Therefore, with all other conditions being equal, it is more logical initially to utilize reactor-grade plutonium and only after, weapons-grade plutonium. The radiotoxicity index factor is highly important for the Mayak plant conditions with its difficult ecological situation.

The question of plutonium utilization in thermal reactors in many countries is connected with the relatively high cost of fast reactors. In Russia, the electricity generated by the first BN-600 is 40 per cent more expensive than that of VVER-1000. But the experience of BN-600 was taken into account in the BN-800 project and the metal consumption factor is only 80 per cent of the

![Figure 2. TRU radiotoxicity evolution after 1st recycle of weapon Pu (WPu) and civil Pu (CPu) in VVER and BN type reactors.](image-url)
BN-600 project. The improved economic characteristics of the BN-800 fuel cycle have been reached through the transition from uranium (which is ineffective for fast reactors) to MOX fuel and the further improvement of the burn-up factor.

The economic characteristics of the fast and thermal reactors have also been compared, based on the increased safety standards required in our country after Chernobyl. Inherent characteristics of fast reactors, as well as new technical solutions, enabled us to upgrade the BN-800 project to that of a world-level nuclear power plant of a new generation, with improved safety characteristics. It is the only project which has passed all, including ecological, necessary certifications. Local authorities have already approved the project and have reached an agreement with Minatom.

But work is not completed on new projects with new, safer uranium reactors (VVER-500, VPBER-600). Preliminary estimates show that average capital investment in these projects is higher than in the VVER-1000, but corresponds approximately to the investment necessary for the construction of three BN-800 units as Yuzhno-Uralskaya. Another factor to be considered is that Russia has no industrial-scale MOX fuel fabrication for VVERs. Such a technological line is to be at the RT-2 facility, to be commissioned after 2005.

Regarding BN-type reactors: as mentioned above, the Complex-300 is 50 per cent completed. With sufficient investment, it can be commissioned by the end of the century, before the commissioning of the first BN-800 unit. After putting into operation three BN-800 units, Complex-300 will reach its maximum capacity in MOX fuel fabrication.

3.2. Long-Term Option

The second stage of plutonium technology mastering in Russia is connected with start up RT-2 plant after the year 2005 (near Krasnoyarsk) meant for VVER-1000 reactor spent fuel reprocessing. The projected quantities of plutonium to be extracted will be much larger than the possible needs of fast reactors. Plutonium to be extracted at the RT-2 plant is to be used in VVER-1000 reactors. A special workshop is being planned at the RT-2 plant for MOX fuel fabrication.

It is understood that plutonium use in thermal reactors is a forced decision and is limited by the number of recycles because of radiotoxic minor actinide accumulation. Fast reactors will complete the fuel cycle for thermal reactors, solving the problems of plutonium isotopic composition and the burning of minor actinides.

The aim of these system studies is to determine the optimal parameters of thermal and fast reactor cores and the optimal composition of the nuclear power grid. Criteria for fuel cycle efficiency should encompass all aspects of economics, ecology and fissile material nonproliferation.

At the present time in Russia, as it was mentioned earlier, the possibility of using civil plutonium in VVER reactors with MOX fuel up to 1/3 of the
reactor core is being investigated. Also, calculational physical research is underway in the area of VVER reactors with a full load of MOX fuel. Cermet fuel is also under development to provide more effective plutonium burning and increase the safety of the reactor core.

As for breeder reactors, the key direction of research is destruction of plutonium excesses and radiotoxic minor actinides stored in thermal reactors. For these purposes new reactor cores with MOX fuel, and increased content of plutonium in fuel without blankets are under consideration. Also, new fuel composition on the basis of an inert matrix instead of source uranium are being developed.

Along with the traditional uranium-plutonium cycle, fuel cycles with thorium and uranium-233 are under consideration. These will allow us to decrease to a minimum the quantity of plutonium in the nuclear industry and minor actinides in wastes.

In the Figure 3 different options of plutonium management are presented as a scheme. Some of them are already being realized in the countries with developed plutonium technology and some are hypothetical ones, being considered, as a rule, in the countries having no experience of plutonium utilization. We think it is reasonable that all such options be compared using the same methodology and mutually agreed upon criteria.

It is obvious, that the aim of such a comparative analysis is not a refusal from Nuclear Power development and plutonium utilization scenario in such
countries as France and Russia. It is an exchange of information, multiparametric scientific analysis ensuring choice of a scenario optimal in the frames of national strategy, multiobjective substantiation of this strategy.

CONCLUSIONS

1. The Russian concept of plutonium management (both civil and weapons) is based on the postulate of the outer fuel cycle closure, necessity to enhance fuel efficiency, and decreasing radioactivity of disposed long-lived wastes.

2. Short-term plutonium management in Russia is based on safe and reliable storage of separated civil plutonium (at PO "Mayak") and ex-weapons plutonium until it can be used in reactors.

3. Medium-term plutonium disposition options are based on development of a Nuclear Power Center at PO "Mayak" (RT-1, Complex-300 and 3 BN-800s) to use accumulated civil and ex-weapons plutonium in fast reactors.

4. Long-term options to be defined consider use of plutonium in VVER-type reactors and the burning of plutonium excess and minor actinides in fast reactors with new cores.

5. Investigations are underway to estimate plutonium utilization options, taking into account non-proliferation, environment, safety, health and costs.

6. International cooperation with the task not only to develop current technical politic but to determine optimum long-term disposition is desired.
AN OVERVIEW OF THE ACTIVITIES OF THE OECD
NUCLEAR ENERGY AGENCY ON PLUTONIUM

N. ZARIBAS
OECD/NEA,
Issy-les-Moulineaux, France

Abstract

The paper presents the framework of NEA plutonium activities and summarizes past and present relevant work.

1. INTRODUCTION

It is recognised that civilian plutonium management and use are not only related to technological, economic and safety considerations but also have wider political, non-proliferation, national and international policy and public acceptability implications.

The increased stocks of separated plutonium and the postponement or even the abandonment of the implementation of fast reactor programmes have resulted in a growing interest, in a number of countries, in recycling plutonium in light water reactors. While uranium requirements, enrichment and fuel fabrication capacities and requirements, reprocessing capacity and spent fuel production can be reliably predicted on the basis of the present operating and scheduled nuclear power capacity until about 2005, future separated plutonium inventory forecasts have large associated uncertainties and are quite sensitive to mixed oxide fuel fabrication and reprocessing assumptions.

Quantities of separated civilian plutonium have been increasing for the past 30 years and are expected to continue to increase in the near future. Under the first and second Strategic Arms Reduction Treaties (START I and II) and unilateral pledges made by American and Russian Presidents, many thousands of U.S. and Russian nuclear weapons will retire within the next decade. As a result, a large inventory of weapon-grade plutonium on each side is expected to become surplus to military needs. Part or all of this surplus plutonium may eventually reach the commercial market. Quantitatively, its impact on the world natural uranium supply markets is not likely to be significant, but existing civilian fuel cycle facilities, services and programmes will be affected and should probably need to be adjusted accordingly to cope with this material.

Policies adopted by governments will be affected by their knowledge of technologies available in the civil fuel cycle and of likely costs. These aspects, together with scientific studies that provide supporting information, are the subject of work at the NEA – other aspects are treated in other international bodies or in bilateral arrangements between governments.

2. THE GENERAL FRAMEWORK OF NEA PLUTONIUM ACTIVITIES

Technical NEA Committees have many years of accumulated experience in dealing with sensitive issues, including among others plutonium economics, safety, related technological and science aspects and recycling logistics. Relevant work, which is usually performed by ad-hoc Expert Groups consisting of nominated high-level national experts, is primarily addressed to OECD Member Governments. The resulting reports present unbiased views which are always approved by representatives of technical NEA Committees before publication. Historically, circulation of those consensus documents has been quite wide and attracted the interest of the industry, the general public and the media.
Past and present NEA plutonium work has been, essentially, technical in nature. Institutional aspects, non-proliferation and physical security are treated in the International Atomic Energy Agency and elsewhere. Staff members of the IAEA and the Commission of the European Union take active part in NEA activities and provide valuable input, ensuring at the same time proper co-ordination and optimal use of resources.

The objectives of the NEA plutonium studies are generally targeted on the need to identify suitable technical solutions, despite the existing large political uncertainties associated with their implementation. In this respect, it should be noted that national programmes and policies are quite diverse. Public opinion concerning plutonium is, however, quite sensitive in almost all countries irrespective of the material’s original nature, be it civilian or military.

3. PAST ACTIVITIES

The NEA has had in the past a number of plutonium related activities. In the mid-80s the NEA’s Nuclear Development Committee (NDC) conducted a study on plutonium and published in 1989 the resulting report: "Plutonium Fuel - An Assessment". This provided the facts and then current views about plutonium and its civil use in the short and medium terms. It addressed questions influencing the choice of fuel options and illustrated how economic and logistic assessments of the alternatives could be undertaken.

Plutonium handling and safety issues, based on available experience in OECD countries, were addressed in the 1993 NEA study on the "Safety of the Nuclear Fuel Cycle". The report underlined the extreme care which has been taken in both the design of plutonium handling facilities and the detailed operational procedures that have been adopted and noted their very good safety records.

Regarding the economics of plutonium use, the recently published NEA study on the "Economics of the Nuclear Fuel Cycle" included updated analyses of the economics of MOX fabrication and of the entire reprocessing cycle.

4. CURRENT ACTIVITIES

4.1 Expert Group on the management of plutonium

There is clearly a need for decisions about the management of plutonium. Although a number of related national activities do exist and international meetings and conferences are at present frequently organised, it was felt that the authoritative consensus report of an international expert committee would be very welcomed by decision-makers and industrialists alike.

During 1993, an informal group of knowledgeable government officials and industry experts developed an initial NEA study proposal on plutonium management, taking into account that the resulting NEA report would be of use primarily to governments in evaluating their policies and presenting them to the public. That proposal has, in turn, been accepted by the NEA’s responsible Committees.

An ad-hoc Expert Group was, therefore, assembled in early 1994, under the auspices of the NEA’s Nuclear Development Committee, with the task of identifying, examining and evaluating the broad technical questions related to plutonium management. Recognising that this was a subject of interest to all countries, whether or not they had in stock separated plutonium or any intention of using plutonium, the following nominated participants to the Expert Group: Australia, Belgium, Canada, France, Germany, Ireland, Italy, Japan, Korea, The Netherlands, Norway, Switzerland, United Kingdom and the United
States. Exceptionally, experts from Russia were invited to participate in this activity because of the accumulated experience in that country with plutonium production, handling and use. The IAEA and the European Commission have also nominated members for the Expert Group.

The Expert Group met in Paris in full session twice and its third meeting is foreseen for Spring 1995. Designated sub-groups already produced valuable contributions along the lines agreed during the meetings. According to current plans, it is expected that the group’s final report will be published in early 1996.

The scope of this NEA study is detailed below:

"Point of departure: Plutonium arises in operating reactors. Most of it is held in spent fuel. A growing amount has been separated but not yet used. There is public interest in the management of this plutonium including any that may become available to the civil market from ex-military uses. The NEA study is concerned with the technical options for management of this plutonium.

The study’s main analyses will primarily address:

A. Technologies that have already been implemented which provide for medium-term storage of plutonium or for recycling the plutonium through reactors. A review will be provided of experience with them and a technical commentary on their potential deployment over the next 20 years or so.

B. Future technologies that may be joined by a further range which are, in some cases, already under development. A technical review of the additional options that may become available will be provided."

As mentioned before, neither institutional matters would be considered by the expert group nor military plutonium as such, but note will be taken of any specific problems or opportunities associated with plutonium of different isotopic composition.

The study is not expected to make any policy recommendations, however, all plutonium technical management options, reflecting both the current experience and future developments will be covered. Accordingly, the Expert Group agreed during its second meeting that the structure of its report should be as follows:

1. Introduction
   - Scope/objectives
   - Total plutonium arisings
   - Separated plutonium
   - Plutonium in spent fuel
   - Reprocessing
   - Report organisation

2. Experience gained and current trends
   - Storage
   - Purification
   - Transportation
   - MOX fabrication
   - Experience with constructing MOX fabrication plants
   - MOX in reactors:
     - thermal
     - fast
   - CANDU
   - MOX reprocessing
3. **Future developments**  
   - Advanced reactors, cores, fuels and fuel cycles  
   - Disposal of plutonium

4. **Country programmes**

### 4.2 **Working Party on physics of plutonium recycling**

The Nuclear Science Committee of the NEA set up in early 1993 a Working Party to deal with the status and trends of physics issues related to plutonium recycling with respect both to the back-end of the fuel cycle and to the optimal utilization of plutonium.

The objectives of this project are to provide Member Countries with up-to-date information on:

- **a)** the core and fuel cycle issues of:
  1. multiple recycling of plutonium in partly-loaded LWRs with MOX fuel,  
  2. full MOX fuel recycling LWRs cores;  
- **b)** the flexibility of fast reactors to produce or to burn plutonium within standard fuel cycles (eg.: using MOX fuel), or advanced fuel cycles (eg.: metal, nitride fuels);  
- **c)** the core and fuel cycle physics issues related to the use of plutonium fuel without uranium (eg.: plutonium on inert matrices) to enhance plutonium burning capability;  
- **d)** the physics issues related to the use of uranium from recycling;  
- **e)** the core and fuel cycle issues of plutonium recycling through advanced converter reactors (eg.: CANDU PHWR, ATR).

The Working Party will also provide advice to the nuclear community on the developments needed to meet the requirements (data and methods, validation experiments, strategic studies) for implementing the different plutonium recycling approaches.

Belgium, Canada, France, Germany, Italy, Japan, the Netherlands, Switzerland, United Kingdom, United States and the IAEA have nominated members to the Working Party which has already met twice. A number of benchmark exercises have been completed and the Group is expected to finalize its work and produce its state-of-the-art report on scientific aspects of plutonium recycling by early 1995.

5. **SUMMARY**

The growing plutonium inventories may well exacerbate the discontent with nuclear energy expressed in some quarters. To the extent that it is seen as desirable that the option of using plutonium in the medium- to long-term should be maintained, it should be demonstrated that civilian stocks of plutonium will continue to be managed well in the short-term. Possible national approaches should give full consideration to the following:

- the rationales for decisions adopted by those countries interested in pursuing plutonium use;  
- the availability of and experience in using technology for all segments of the plutonium recycle route;
the potential for further improvements in technology aiming, for example, at even higher standards of environmental protection, safety, worker health and reduced costs;

- priorities for further research and technical developments.

The past and present NEA programme on plutonium aimed at addressing the above issues and provided assistance to Member Governments, decision-makers and the industry by producing authoritative and comprehensive evaluations of related technologies, scientific aspects and economics. The NEA will continue in the future with similar activities and may also expand its programme, as necessary.
This report is a brief overview of issues relevant to the performance of irradiated (Th,Pu)O₂ as a waste form for geological disposal. Fuels of this type, among others, are being considered for burning plutonium from dismantled nuclear weapons in power reactors, including CANDU systems. The high chemical stability and low aqueous solubility of thoria make this type of fuel attractive as a waste form. In contrast with UO₂ fuel, the inertness of thoria to oxidation dominates most of the chemical issues of fuel disposal. The overall performance of a thoria-based fuel waste form is likely to be determined by the "instant" release of the gap inventories of mobile fission products such as ²³⁹I. This, in turn, will be controlled largely by the in-reactor power history and probably also by details of fuel fabrication. Limited experience with thoria-based fuels (chiefly (Th,U)O₂) indicates that, for given power and burnup levels, gas releases can be substantially lower than with UO₂ fuels. The gap and grain-boundary inventories of fission products are expected to be correspondingly low. A fabrication route involving molecular-level mixing (e.g., sol-gel process) would be preferable to powder blending, because microscopic heterogeneities in the fuel could adversely affect the retention of fission products. Pilot-scale irradiation, post-irradiation examination and leaching studies are required to support this preliminary assessment. Other issues that need to be addressed include impurity specifications (to minimize formation of long-lived activation products), and criticality and safeguards issues that might influence the design of fuel-handling facilities.

1. INTRODUCTION

There is an urgent need to identify appropriate methods to deal with the plutonium recovered from dismantled nuclear weapons and military stockpiles, and make it inaccessible for future weapon use. At present, debate centres on the options of either direct disposal of the plutonium in a nuclear waste glass, or burning it in a nuclear reactor and disposing of the irradiated plutonium-bearing fuel (Holdren 1994). The latter approach has the advantage that useful energy would be obtained from the plutonium, and hence some return could be obtained on the large investment.

A number of different reactor and fuel types are being examined for their capability to burn weapons plutonium. The CANDU[1] reactor system is particularly attractive, because it has the flexibility to operate with a variety of different fuel types, and it is able to accommodate a full core loading of plutonium-bearing fuel (Dastur et al. 1994).

Three generic fuel types could, in principle, be used to burn plutonium in a power reactor: (a) Uranium-based mixed oxides, (U,Pu)O₂; (b) Thorium-based mixed oxides, (Th,Pu)O₂; (c) Thorium-based mixed oxides.

[1]: Canada Deuterium Uranium, registered trade mark of Atomic Energy of Canada Limited
(Th, Pu)O$_2$; (c) Inert-matrix fuels, in which PuO$_2$ is dispersed in an inert diluent such as zirconia (Akie et al. 1994). The (Th, Pu)O$_2$ fuel option is attractive because breeding of $^{233}$U would allow conversion of over 90% of the $^{239}$Pu at high burnup in a once-through cycle (Veeder and Didsbury 1985).

Thorium-based mixed oxides are also appealing from a waste management perspective, because ThO$_2$ is chemically stable and almost insoluble in groundwater. In this report, we explore waste-management aspects of thoria-based plutonium fuels. We compare some of the key chemical and physical properties of UO$_2$ and ThO$_2$, and draw from experience in the assessment of geological disposal of irradiated UO$_2$ fuel (Johnson et al. 1994), to identify issues that would need to be addressed in a thorough assessment of thoria-based fuel disposal. We assume that the fuel would contain about 3 wt.% PuO$_2$ in ThO$_2$. We also assume a similar disposal scenario to that envisaged in the Canadian Nuclear Fuel Waste Management Program, i.e., direct disposal of used fuel bundles in corrosion-resistant containers, surrounded by a clay-based buffer material, within a vault excavated deep in granite (Hancox and Nuttall 1991).

2. CHEMISTRY OF THORIA

2.1 Redox Chemistry

By far the most important chemical difference between ThO$_2$ and UO$_2$ is the fact that thorium is present in its maximum oxidation state, Th(IV), whereas uranium is not. Under oxidizing conditions, UO$_2$ can be converted to the comparatively soluble uranyl cation, UO$_2^{2+}$, and its derivatives. This reaction and the corresponding reduction dominate the geochemistry of uranium (see below), and an understanding of the kinetics of oxidative dissolution of UO$_2$ is central to the performance assessment of irradiated UO$_2$ fuel as a waste form. Oxidative dissolution of the matrix is not an issue with thoria fuel. Redox conditions could affect the leachability of $^{233}$U from irradiated thoria, but this would be limited to surface dissolution and is unlikely to be a major concern.

The inertness of thoria to oxidation is also relevant to interim dry storage of irradiated fuel before geological disposal. The maximum acceptable temperature for dry storage of CANDU UO$_2$ fuel in air is typically 150-175°C, because at higher temperatures oxidation of UO$_2$ to U$_2$O$_8$ in defected elements can cause powdering of the fuel matrix and splitting of the fuel cladding (Boase and Vandergraaf 1977, Hastings et al. 1986, Novak et al. 1983, Wasywich et al. 1993). Somewhat higher temperatures may be acceptable for higher-burnup UO$_2$, such as LWR fuel, based on data reported by Thomas et al. (1993). Matrix oxidation is not an issue with thoria-based fuels. Furthermore, the thoria structure can easily accommodate oxidation of minor solid-solution components such as U and Pu (see Section 2.4). For example, detailed investigation of the U-Th-O system has shown that thoria-rich solid solutions, $(\text{Th}_{1-x}\text{U}_x\text{O}_{2+y}$), are stable in air if $x \leq 0.5$, and $\text{U}_5\text{O}_8$ is only formed from more uranium-rich compositions (Cohen and Berman 1966). Thus, fuel oxidation is unlikely to be a concern during dry storage of (Th, Pu)O$_2$, and hence the maximum storage temperature would be limited by some other factor, probably cladding degradation (EPRI 1989).

2.2 Aqueous Chemistry

Most metal(IV) dioxides have very low solubilities in aqueous solutions except at extreme pH conditions, or under redox conditions favouring either reductive or oxidative dissolution. Because ThO$_2$ is inert to oxidation and extremely difficult to reduce, it is virtually insoluble over a wide range of solution conditions.
The solubility of crystalline thoria at 25°C and pH>5, in the absence of complexing agents, has been estimated at $10^{-14}$ mol/kg, or 2 parts per quadrillion (Langmuir and Herman 1980). Even lower solubility estimates for crystalline thoria have been suggested by Ryan and Rai (1987), based on solubility measurements on hydrous thorium(IV) oxide and estimates of the difference in stability between the hydrous and crystalline forms. The solubility increases rapidly as the pH falls below 5. In addition, both inorganic and organic ligands can enhance the solubility of ThO$_2$ in neutral to acidic groundwaters (Langmuir and Herman 1980, Felmy et al. 1991).

Lemire and Garisto (1989) calculated solubility limits of thorium in groundwaters contacting UO$_2$ fuel in a granitic waste vault of the type envisaged in the Canadian Nuclear Fuel Waste Management Program. The calculated values represent the solubility of hydrous thorium oxide, as opposed to crystalline thoria, and are therefore very conservative (i.e., overestimates) for the purpose of the present report. The calculations were performed for a distribution of water chemistry parameters representing expected conditions in a granitic waste vault, and a distribution of thermodynamic values representing experimental uncertainty limits. The most frequent values for the calculated thorium solubility (for 40,000 randomly selected cases) are about $10^{-9.5}$ mol(Th)/kg(H$_2$O) at 25°C and $10^{-11}$ mol/kg at 100°C. These values are slightly higher and slightly lower, respectively, than corresponding values for uranium under reducing conditions. Small but finite tails in the distributions extended to much higher solubilities; in the case of thorium, these are mainly an artifact of high uncertainty limits on some complexation constants, and are not thought to be realistic (R.J. Lemire, personal communication, September 1994).

We conclude that ThO$_2$ solubility is extremely low (comparable to UO$_2$ under reducing conditions) over a wide range of aqueous solution conditions. Nonetheless, it would be desirable to perform experiments and calculations to define more accurately the conditions (e.g., pH and groundwater compositions) under which solubility may be elevated.

The release of actinides and those fission products that are retained by the thoria matrix is expected to be limited by the solubility of ThO$_2$. Such release would be exceedingly slow in an engineered disposal vault of the type envisaged for CANDU UO$_2$ fuel. In nature, thorium is often closely associated with clays or oxide minerals such as bauxite. Thus, one might expect some acceleration of thoria dissolution if it is coupled via diffusion to sorption on clay buffer material or other mineral surfaces. The enhancement of the dissolution rate of UO$_2$ as a result of mass-transport-precipitation coupling has been studied by Garisto and Garisto (1986, 1988). This phenomenon becomes important if a redox front exists near the dissolving UO$_2$. The results imply that the dissolution rate of thorium could be similarly enhanced by coupling to strong adsorption in the buffer, although the impact is likely to be smaller because of the extremely low solubility of thorium under the expected groundwater conditions.

Similar arguments would apply to possible alteration processes, such as the conversion of ThO$_2$ to ThSiO$_4$ by reaction with dissolved silica. In other words, this conversion will be exceedingly slow in typical groundwaters, even though it is thermodynamically favoured (Kamineni and Lemire 1991).

### 2.3 Radiation Effects

Radiation damage to nuclear fuels has been studied extensively (Matzke 1982). In general, radiation damage to refractory oxides increases their susceptibility to dissolution
and leaching. However, dissolution experiments, along with isotopic studies of minerals suggest that radiation damage to thorium is fairly readily annealed (Eyal and Fleischer 1985a, 1985b, 1985c, Vance and Gascoyne 1987). Neither ThO$_2$ nor UO$_2$ is susceptible to metamictization (radiation-induced transformation to an amorphous state). This can be understood on the basis of a low crystallization temperature relative to the melting point; recrystallization is highly favoured in the thermal spike associated with an ion track. The low degree of radiation-induced damage, along with a tendency for defects to anneal, suggests that radiation-enhanced dissolution of ThO$_2$ fuel will be slight.

Radiation-induced enhancement of U and Pu diffusion in UO$_2$ and (U,Pu)O$_2$ at temperatures below 1100°C is well known (Matzke 1992); this has an impact on waste management issues because in-reactor grain growth is limited by metal ion diffusion. There are no significant differences in the diffusion rates of tetravalent metal ions (Th$^{4+}$, U$^{4+}$, Pu$^{4+}$) in ThO$_2$ and the rate of metal ion diffusion is somewhat slower than in stoichiometric UO$_2$. We thus expect in-reactor grain growth and related fission-product segregation to occur to the same extent, or less, as in UO$_2$.

The effect of radiation on the rate of fission-gas release is qualitatively the same for both UO$_2$ and thorium-based fuels (Naik et al. 1981, Kaimal et al. 1990). In each case the diffusion coefficient at a given temperature is constant at low doses, then drops dramatically as the dose is increased to $10^{24}$ fissions/m$^3$ because fission gas atoms are trapped by radiation-induced defects (Matzke 1992, Naik 1992). Radiation-induced damage is thus not expected to have adverse effects on the rate of fission-product release from a thorium-based fuel.

A separate radiation issue for used UO$_2$ fuel is the effect of radiolysis of water on the oxidative dissolution of UO$_2$ (Sunder et al. 1990, Shoesmith and Sunder 1992). This is another process that involves the oxidation of Th(IV) to Th(VI), and is not a concern with thorium. It is therefore unlikely that radiolysis will have any significant influence on thorium dissolution in a disposal vault.

2.4 Natural Analogues

Useful information about the behaviour of UO$_2$ fuel in a disposal vault has been inferred from the mineralogy and groundwater chemistry of uraninite mineral deposits. An example that has several close parallels to a fuel disposal vault is the Cigar Lake ore body in Saskatchewan, Canada (Cramer and Smellie 1994). Natural analogues for thorium fuel disposal are more tenuous because thorium is not highly concentrated by any geochemical processes; this is another consequence of its inertness to redox processes (Langmuir and Herman 1980, Bayer et al. 1974). In addition, minerals in which thorium is the major component, such as thorianite (ThO$_2$) and thorite (ThSiO$_4$), are rare. Large thorianite crystals do occur rarely in some pegmatite rocks (Bayer et al. 1974). Even though there is no natural formation rich in thorianite, the general immobility of thorium in nature may be instructive. For example, concentrations of dissolved thorium in surface waters are exceedingly low (Langmuir and Herman 1980). Natural deposits of thorium usually occur either as resistate minerals (i.e., insoluble residues of chemically resistant accessory minerals, such as monazite, a substituted rare-earth phosphate, that remain after rock weathering) or through adsorption and retention on clay or oxide alteration minerals (if the thorium originates as a minor component of the major rock-forming minerals in a weathered formation).
2.5 Compatibility of Actinides with Thoria

Thoria crystallizes with the fluorite structure, as do all other actinide dioxides (Wells 1975). Extensive solid-solution formation occurs between these oxides, and the fluorite structure can also accommodate substantial levels of actinides in other oxidation states, such as Am(III) and U(VI), as well as many fission products (Brett and Fox 1966, Kleykamp 1985, Mayer et al. 1993). Thus, no phase segregation of actinides is expected to occur within the fuel, either during operation or after disposal, and it is reasonable to assume that release of actinides will be controlled by the slow dissolution rate of the thoria matrix, provided that the fuel is initially homogeneous. Partial segregation of uranium and plutonium has been shown to occur by incongruent vaporization under the steep temperature gradients experienced under severe reactor operation conditions (Green et al. 1983, Baptiste 1984). This is unlikely to be sufficiently extensive to affect the waste-form performance of mixed actinide fuels, but it should be addressed in post-irradiation examination of (Th,Pu)O$_2$ fuels.

3 FISSION-PRODUCT SEGREGATION

Calculated environmental releases and subsequent radiation doses arising from a CANDU UO$_2$ fuel disposal vault are dominated by the "instant" release of soluble and mobile fission products (in particular, $^{129}$I) from the fuel/sheath gap of the fuel (Stroes-Gascoyne et al. 1987, Johnson et al. 1994). Grain-boundary inventories may also be released rapidly, as compared with matrix dissolution. It is likely that similar findings would emerge from a detailed assessment of thoria/plutonia fuel disposal, especially given our expectation of extremely slow matrix dissolution. Therefore, it is important to consider the irradiation history and microstructural behaviour of the fuel, and to have reliable information on the segregation of mobile fission products to the gap and grain boundaries in thoria-based fuels. Such analysis is also important because several fission products have been implicated in fuel failures caused by stress-corrosion cracking of the cladding (Cox 1990); in addition, high levels of fission-gas release can lead to element swelling.

At present, international experience with irradiation of thoria fuels in general, and information on detailed post-irradiation examination in particular, is limited. Promising results have been reported on gas releases from thoria-based fuels (see Section 3.1). In a review of post-irradiation examination of various plutonium-bearing fuels, Cadelli and Lippens (1984) refer only to isotopic analysis of (Th,Pu)O$_2$ fuels. Pinheiro et al. (1988) concluded that (Th,Pu)O$_2$ fuels show promise for extended-burnup, once-through use of recycle plutonium, but most of their materials characterization work was based on the use of cerium as a surrogate element for plutonium in fuel formulations. Katz (1986) described the metallography of thoria-based fuel materials, but with emphasis on method development and the importance of impurities in fuel fabrication. He reported nothing unusual in the porosity and fission-gas bubbles observed in irradiated (Th,U)O$_2$ fuel from the Shippingport reactor.

3.1 Grain Growth and Fission-Product Segregation

Grain growth in the central region of fuel pellets is a major cause of fission-gas release to the gap, because the gases and other incompatible elements are swept from their original resting places in the fuel matrix and become concentrated at the grain boundaries. There, they form features such as fission-gas bubbles and noble-metal particles (Matzke 1980). Interlinkage of fission-gas bubbles on grain-boundary
intersections eventually creates tunnels that permit venting of other fission products to the fuel-cladding gap. Thorium oxide is a somewhat better thermal conductor than UO$_2$; it also has a higher melting point and slower cation diffusion (Section 3.3). Therefore, for a given power rating and fuel geometry, it would be expected to run cooler and undergo less grain growth (Meieran and Westermann 1977). Both the grain-growth kinetics and the thermal conductivity, however, are sensitive to impurities such as fission products. Therefore, direct measurements of grain growth and gas release under power-reactor conditions are essential to identify appropriate operating conditions and to assess disposal performance of the fuel.

Fission-gas release data for (Th,Pu)O$_2$ are not available, but indirect evidence suggests that release rates will be somewhat smaller for thoria-based fuels than for UO$_2$. This conclusion is based on the lower diffusion rate for xenon in ThO$_2$ than UO$_2$ (Matzke 1992, Shiba 1992) and the smaller burst release in ThO$_2$ (Kaimal et al. 1990). The expected low gas release rates from thoria-based fuels are supported by in-pile experiments on ThO$_2$ and (Th,U)O$_2$ fuel assemblies. Goldberg et al. (1977, 1978) measured fission-gas release in a set of 51 thoria-based fuel rods over a range of linear powers, burnups and compositions. They gave an expression for the rate of fission-gas release, which suggests that rates are considerably lower than for UO$_2$ under comparable operating conditions. Accelerated gas release has been reported at high burnup for (Th,U)O$_2$ fuels, but the onset of this phenomenon appears to be delayed, as compared with UO$_2$ fuel (Giovengo et al. 1981). Also, higher-than-expected gas releases have been observed in some Canadian tests with (Th,U)O$_2$ fuels. This has been attributed to a granular microstructure, which both impeded heat transfer and provided networks of porosity that facilitated gas release (Smith et al. 1985).

In many cases, the segregation and hence the leachability of volatile, non-gaseous fission products, such as cesium and iodine, is correlated with fission-gas release (Stroes-Gascoyne et al. 1987), and thus we expect the release of these fission products to be lower for a thoria-based fuel than UO$_2$. Jones et al. (1977) reported low gas releases for (Th,U)O$_2$ fuels, and also noted that fission-product release from defected thoria elements was one to two orders of magnitude lower than for UO$_2$. Experimental data obtained by Matzke (1967) supported this notion; he found that the release of Br, Cs and Rb from thoria was generally slower than from UO$_2$.

For the purposes of this report, we assume that fuel failures would be rare, and that the primary waste-form would be intact (Th,Pu)O$_2$ fuel bundles. The above discussion suggests that this is a reasonable expectation. Obviously, any large-scale use of (Th,Pu)O$_2$ fuels would be contingent on the demonstration of acceptable failure rates. Failure mechanisms for experimental (Th,U)O$_2$ fuel assemblies have been discussed briefly by Bain et al. (1977).

### 3.2 Oxygen Potential and Fission-Product Chemistry

Actinide fission changes the O/M ratio and hence the oxygen potential of oxide nuclear fuels. Fission of $^{235}$U causes only slight changes in oxygen potential, whereas $^{239}$Pu fission releases a significant quantity of surplus oxygen, due mainly to the higher yield of noble metals (Kleykamp 1985). To assess the effect of fission on oxygen potential in (Th,Pu)O$_2$ fuel, some knowledge of the influence of $^{233}$U as well as $^{239}$Pu fission is required. The fission-product spectrum for $^{233}$U is depleted in noble metals, as compared with $^{239}$Pu, therefore it may mitigate the oxygen surplus generated by $^{239}$Pu at extended burnups.
Changes in oxygen potential of the fuel can markedly affect the chemistry of some fission products, for example, molybdenum may be oxidized from the metallic state to MoO$_3$ (Kleykamp 1988, 1990). A high oxygen potential may also increase the mobility of $^{99m}$Tc, one of the radionuclides of particular concern in nuclear fuel waste management (Johnson et al. 1994). Uranium dioxide has a high capacity to accommodate surplus oxygen as interstitial defects or defect clusters in the UO$_2$ lattice (Willis 1987); the surplus oxygen is compensated by oxidation of uranium to form U(V) or U(VI) species. Pure thorium does not have this capacity, hence the effect of surplus oxygen on fission-product chemistry (or possibly on the cladding) may be more pronounced than is the case with UO$_2$. Clearly, the evolution of oxygen potential during operation of (Th,Pu)O$_2$ fuels will be quite different from that in UO$_2$ fuel, and this process and its possible consequences need to be addressed.

3.3 Diffusion Properties of Thoria

In general, diffusion kinetics in the oxide matrix control fission-product segregation, except in the core of fuels operated at high power, where grain-growth occurs. Three distinct diffusion processes need to be addressed. Oxygen self-diffusion is much faster than cation diffusion in both ThO$_2$ and UO$_2$, but is involved only indirectly, if at all, in fission-product release mechanisms. Diffusion of the principal cation (i.e., Th or U) governs grain growth, which in turn affects fission-product release at high power ratings. Finally, diffusion of the fission products themselves governs their release under most operating conditions.

Thorium ion self-diffusion in stoichiometric ThO$_2$ is slower than oxygen diffusion by five or six orders of magnitude. The slow rate of this process makes experimental measurements difficult, and early data show generally poor reproducibility (Shiba 1992). More recent data indicate that the cation self-diffusion rate in ThO$_2$ approaches that in UO$_2$ above 2000°C, but drops off somewhat more quickly at lower temperatures, because of a larger activation energy, 627 vs. 540 kJ/mol (Matzke 1987, 1992). Furthermore, a small degree of hyperstoichiometry increases thermal lattice diffusion of cations in UO$_2$ by several orders of magnitude because of its impact on the uranium vacancy population, whereas ThO$_2$ practically always remains stoichiometric (Matzke 1987). Grain growth should thus be slower for ThO$_2$ than UO$_2$ fuels, and may be negligible, under the operating conditions normally encountered in water-cooled reactors.

Diffusion of fission products in UO$_2$ and ThO$_2$ remains poorly understood, but generally appears to involve U or Th ion vacancies. High-temperature, out-of-pile annealing experiments on lightly irradiated or ion-implanted samples appear to be consistent with modestly lower fission-product diffusion rates in ThO$_2$ than UO$_2$ - roughly paralleling the difference in cation lattice diffusion (Matzke 1967, 1980, Akabori and Fukuda 1991, Prussin et al. 1988, Naik 1992). Fission-product migration in-reactor involves further complexity; indeed, Matzke (1980) has suggested that five different diffusion coefficients are required to model fission-gas transport! Nonetheless, the overall trend is evidently maintained: under equivalent operating conditions, fission-product segregation and release tend to be lower for ThO$_2$ than UO$_2$ fuels (Section 3.1).

4. FUEL FABRICATION

Much of the preceding discussion hinges on initial homogeneity of the fuel, which raises the issue of fuel fabrication. If the fuel is initially homogeneous on a molecular scale, then its chemical properties will be governed by the chemically stable thorium. If the fuel
is fabricated by blending thoria and plutonia powders, however, it is likely to contain microscopic plutonia-rich domains (grains or subgrains) within a nearly pure thoria matrix. The behaviour of the plutonium and its fission and activation products may then no longer be controlled by the thoria, since they would be concentrated in microscopic domains of very high local burnup.

Microscopic inhomogeneity in (U,Pu)O$_2$ and (Th,U)O$_2$ fuels prepared using powder-mixing technology has been shown to cause significantly enhanced fission-gas release (Cadelli and Lippens 1984, Ishida and Korei 1994, Smith et al. 1985), and similar results could be expected with (Th,Pu)O$_2$. Preferential leaching of other fission products from the high-burnup, Pu-rich grains in such fuels would also be expected. Preferential dissolution of plutonium as Pu$^{4+}$ is also a possibility under reducing conditions. Therefore, it is likely that inappropriate fabrication processes would negate the benefits that arise from the desirable chemical and physical properties of a thoria fuel matrix. Small-scale tests with (Th,U)O$_2$ fuel indicate that sol-gel fabrication procedures yield a more homogeneous product than powder-mixing techniques, and its irradiation performance is promising (Hastings et al. 1982, Onofrei 1986). Further work is clearly needed to verify that suitably homogeneous fuel can be fabricated on a full industrial scale.

Reactor operation also affects fission-product release. At the linear power ratings typical of CANDU reactors grain growth in natural UO$_2$ fuel is slight. At similar power, (Th,Pu)O$_2$ fuels should exhibit little or no grain growth; however, these fuels may operate at higher power densities than natural UO$_2$. CANDU reactors have flexibility in fuel management and fuel design that can ensure that (Th,Pu)O$_2$ fuels would operate at similar or lower linear power ratings, compared to current UO$_2$ fuel. For example, the 43-element CANFLEX bundle reduces peak ratings by about 20% compared to the 37-element bundle (Lane et al. 1993). Furthermore, changes can be made to the internal element design to lower fuel-operating temperatures directly. Examples of such changes are graphite disks between pellets, or annular fuel, or both. Hence there is a real possibility that (Th,Pu)O$_2$ fuel could be operated in CANDU reactors with minimal gas release.

5. OTHER ISSUES

A number of additional issues that would need attention in developing a plutonium-burning and fuel-disposal scheme are addressed briefly here.

In addition to fission products, some activation products are significant in the assessment of UO$_2$ fuel disposal. Specific examples are $^{14}$C (formed from $^{14}$N impurities and $^{17}$O in the fuel) and $^{36}$Cl (from $^{35}$Cl impurities). Surprisingly, it has been shown that $^{36}$Cl can make a significant contribution to the overall dose arising from UO$_2$ fuel disposal (Johnson et al., in preparation). Activation products will also need to be considered carefully when assessing thoria-based fuels. For example, there may be some incentive to set stringent specifications on chlorine and nitrogen impurity levels in (Th,Pu)O$_2$ fuel.

Criticality considerations will influence the design of facilities for storing and handling irradiated (Th,Pu)O$_2$ fuels. Clearly, this is also a major consideration for fuel fabrication. The radiation characteristics of (Th,Pu)O$_2$ fuels may differ significantly from typical CANDU fuels, and thus affect the design of handling facilities. Finally, although the destruction of plutonium in (Th,Pu)O$_2$ fuel is a net benefit from a proliferation
safeguards viewpoint, the fabrication and storage of this fuel will introduce distinct safeguards issues, as compared with conventional CANDU fuel.

6. CONCLUSIONS

The high degree of chemical stability and the low solubility of thoria make irradiated thoria-based fuels such as (Th,Pu)O$_2$ attractive as waste forms for direct geological disposal. Furthermore, there is good reason to expect lower fission-gas releases (and correspondingly lower gap and grain-boundary inventories of other fission products) in thoria fuels than in UO$_2$ with comparable power history. In order to realize these beneficial qualities of thoria-based fuels, an appropriate fuel-fabrication process must be utilized to achieve an acceptable degree of microscopic homogeneity. Detailed post-irradiation examination and leaching studies of thoria-based fuels, coupled with a thorough understanding of their physical and chemical properties, are needed to support these preliminary conclusions.

We thank P.G. Boczar, T.J. Carter, R.J. Lemire, Hj. Matzke, and J. Veeder for helpful comments on a draft of this report.

BIBLIOGRAPHY


Kleykamp, H. 1988. The chemical state of fission products in oxide fuels at different stages of the nuclear fuel cycle. Nuclear Technology 80, 412-422.


Matzke, Hj. 1967. The release of some non-gaseous fission products from CaF$_2$, UO$_2$ and ThO$_2$. Journal of Nuclear Materials 23, 209-221.


RATIONAL Pu-TRANSMUTATION FOR $^{233}$U

A. LECOCQ
EURIWA, Orsay, France

K. FURUKAWA
Institute of Research and Development, Tokai University,
Kanagawa, Japan

Abstract

To achieve $^{233}$U production from Thorium, with simultaneous destruction of Plutonium, nuclear specialists have proposed numerous ways of which the consequences may be very different for the other problems of the peaceful nuclear utilization: Safety, Waste management, Industrial Deployment....

Rational choices have to be made for attempting to solve these difficulties, so as nuclear energy be acceptable.

The Molten-Salt technology, applied for Fission reactors -supplying energy-, coupled with Accelerators -waste destroyers & producers of fissile material ($^{233}$U from Thorium-, allows to define a worldwide strategy, called THORIMS-NES, for the global supply of a clean and abundant energy.

The non-aqueous reprocessing processes, used as a junction between the previous applications of the Fission and these new devices, could resorb the present stocks of unreprocessed spent fuels, by recycling them in THORIMS-NES.

1. INTRODUCTION

The world is changing: the end of the cold war, the economical crisis which concerns as well the East-West relations as the North-South ones, the exponential increase of population which breeds economical and cultural poverty, the renew of conflicts for national and or religious incentives, are sufficient to convinc us that our human society suffers a deep mutation.

The scientific and technical community cannot stay out of this crucial debate for the future of our civilization. Time is come for us -specialists of nuclear energy- to draw the lessons of the past, making benefit from 50 years of nuclear energy researches and industrial deployment.

The positive aspects of present nuclear Fission Energy cannot be denied, as we shall see later, but the need of broad changes urges us to make rational choices to solve its own problems. The GLOBAL SUPPLY OF ENERGY and PLUTONIUM DISPOSITION (or NUCLEAR PROLIFERATION REDUCTION) are the first priorities, as it was understood by the organizers of this meeting under the aegis of IAEA. Besides these first challenges, the nuclear community has also to find rational solutions for the SAFETY of nuclear applications, for the WASTE MANAGEMENT, for the INDUSTRIAL DEVELOPMENT at a world scale, without omitting either the ECONOMIC ASPECT, or the prospects for the FUTURE DEPLOYMENT of the various technologies.

2. THE TWO FIRST PRIORITIES

2.1. Global Energy Supply

After 30 years of industrialization we may observe:
-very few small civilian power stations are deployed due to their high cost and by fear of proliferation.
-only about 2TWe.year were produced since the start.
-only small amounts of spent fuel are reprocessed, inducing a deep thread of pollution for the next centuries.
-Thorium resources are neglected.
Thorium utilization to produce energy was considered in the initial stage of nuclear energy studies, because of the good neutronic properties of $^{233}$U under thermal neutrons flux. But the Thorium element has no fissile isotope, and fissile seeds are necessary to start energy production.

Besides it was soon discovered that the transmutation of Thorium into $^{233}$U, produces simultaneously $^{232}$U. This isotope, parent of strong gamma emitters ($^{212}$Bi and $^{208}$Tl), makes indispensable the remote-handling to recycle into a new Thorium matrix, the fissile $^{233}$U obtained by reprocessing of the spent Thorium fuel.

Several projects using Thorium cycle (MSBR, HWSR, CANDU-Th, LWBR), were compared for their efficiency to solve the energy supply problem [1], but no industrial application occurs, partly on account of the difficulties for $^{233}$U recycling.

But now new factors must be considered. The Greenhouse effect occurrence, as explained in an other paper of this meeting [2], and various problems of the U-Pu fuel cycle (sect. 5.1), raise anew the advantages of Thorium which is 4 times as abundant as uranium, and can supply 100 times more energy. Besides the amount of Thorium waste (TRU) is several orders of magnitude smaller than that of Uranium [3].

Since 10 years, several seminars and publications testify to the new interest for Thorium use,[4-6]. The pollution of recycled $^{233}$U by $^{232}$U, which was an handicap for Thorium use, is considered now as useful to prevent the diversion to terrorism and nuclear weapons production. [7]

2.2. Plutonium Disposition (or Proliferation Reduction)

Two grades of Plutonium are generally defined according their isotopic composition: Weapon Grade Plutonium (WGPu) or military Plutonium made of $^{239}$Pu (>93% purity) and Reactor Plutonium (RPu) which is a mixture of the various isotopes produced in the usual power reactors. The distinction between both is rather spurious since it is possible to make a dirty weapon with RPu, and it will be easy, in the future, to produce WGPu from RPu by Laser enrichment....

Also the big amounts of WGPu (200 to 270 t) and of RPu (1000 t by the year 2000) have to be destroy. Moreover, this long half lived element has a high toxicity.

Many interesting projects have been published for fissioning, in power reactors, either WGPu [8-9], or RPu as it will be seen in this IAEA meeting.

At first sight, it seems rational to use classical power reactors with thermalized neutrons (or fast neutrons) to destroy Plutonium; but these old technologies perfectly mature for electricity production, might be adapted to a new fertile material: Thorium. Safety in operation, reprocessing of the spent solid fuel, recycling of $^{233}$U produced in the Thorium matrix, management of the waste, have also to be considered from the start. These new difficulties have to be overcome by a nuclear industry tightly connected, till now, to the U-Pu fuel cycle.

On the opposite, since more than 20 years ago, Molten-Salt reactors specialists have proposed to start the Thorium fuel cycle, by using Plutonium (WGPu or RPu) as a fissile.[10-11], and with a quite new technology grounded on a Fluoride liquid fuel, experimented successfully at ORNL[12].

Since the last 10 years, many improvements have been brought to the original Oak Ridge project (Molten-Salt Breeder Reactor :MSBR) which was focused on the breeding by the $^{232}$Th-$^{233}$U cycle. In an other paper presented [13], it is possible to appreciate the progresses done, since the first fission of Plutonium in the Thorium salt of the Experimental Reactor, MSRE, more than 25 years ago.

Besides the fission of Pu in Burners Reactors, as mentioned above, the development of the Molten-Salt technology opens a new way for the Plutonium destruction with $^{233}$U
### Table 1: Global energy problems and achievable solutions by NEW FISSION TECHNOLOGY [THORIMS-NES]

<table>
<thead>
<tr>
<th>TECHNOLOGICAL PROBLEMS</th>
<th>ACHIEVABLE SOLUTIONS</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>RESOURCE</strong></td>
<td>Th: non-local, popular</td>
</tr>
<tr>
<td><strong>SAFETY</strong></td>
<td>high thermal efficiency, no NO\textsubscript{x}, SO\textsubscript{x}, no CO\textsubscript{2}, no production, complete isolation, minimize few maintenance, few transportation, 233\textsubscript{U} [high gamma], chemical inert, low pressure &amp; flow, negligible small fuel self-sustain.</td>
</tr>
<tr>
<td>fossil</td>
<td>232\textsuperscript{Th} + n \rightarrow 233\textsubscript{U} (fissile)</td>
</tr>
<tr>
<td>ecosystem</td>
<td>simple m. salt cycle, short: 5~10 years</td>
</tr>
<tr>
<td>nuclear</td>
<td>establishment of breeding cycle</td>
</tr>
<tr>
<td>acid rain</td>
<td>multi-functional: nucl. reaction heat transfer chem. process.</td>
</tr>
<tr>
<td>&quot;Greenhouse&quot; effects</td>
<td></td>
</tr>
<tr>
<td>low level waste</td>
<td></td>
</tr>
<tr>
<td>nuclear-proliferation and -terrorism</td>
<td></td>
</tr>
<tr>
<td>Pu [no gamma]</td>
<td></td>
</tr>
<tr>
<td>basic tech: chemical reactivity</td>
<td></td>
</tr>
<tr>
<td>mechanical failure</td>
<td></td>
</tr>
<tr>
<td>nuclear excess reactivity</td>
<td></td>
</tr>
<tr>
<td>configuration</td>
<td></td>
</tr>
<tr>
<td>operation</td>
<td></td>
</tr>
<tr>
<td>[solid fuel], maintenance</td>
<td></td>
</tr>
<tr>
<td>fuel processing</td>
<td></td>
</tr>
<tr>
<td>core meltdown, recriticality</td>
<td></td>
</tr>
<tr>
<td>BREEDING FUEL-CYCLE</td>
<td></td>
</tr>
<tr>
<td>cycle completion: difficult</td>
<td></td>
</tr>
<tr>
<td>doubling time: long: &gt;10 y</td>
<td></td>
</tr>
<tr>
<td>POWER STATION</td>
<td></td>
</tr>
<tr>
<td>siting</td>
<td>near to utility</td>
</tr>
<tr>
<td>power size</td>
<td>small ~ large</td>
</tr>
<tr>
<td>process-heat supply</td>
<td>industrious district</td>
</tr>
<tr>
<td>From the U-Pu fuel cycle:</td>
<td></td>
</tr>
<tr>
<td>- by historical and technical necessity, the U-Pu cycle was the starting point of nuclear energy, but also of Proliferation.</td>
<td></td>
</tr>
<tr>
<td>- less than 1% of natural Uranium is fissioned and produces energy, due to enrichment necessity and loss of fissile in the spent fuels.</td>
<td></td>
</tr>
<tr>
<td>- it breeds, as main waste, Fission Products (FP), and heavy elements (TRU) with long half-lives and high toxicity. Secondary technological waste appear also in the reprocessing Purex plants. (sect. 5.1.)</td>
<td></td>
</tr>
<tr>
<td>From the applied technology in the Fission Industry:</td>
<td></td>
</tr>
<tr>
<td>- low thermal efficiency, uncertain safety.</td>
<td></td>
</tr>
<tr>
<td>- needs of enriched Uranium for most of the nuclear reactor types;</td>
<td></td>
</tr>
<tr>
<td>- weak flexibility in load operation, very limited in size so that the always-large NPP are unexportable in small and poor nations.</td>
<td></td>
</tr>
<tr>
<td>- reprocessing of the spent fuels (if any), inefficient for waste management, as well as for the recovery of unused fissile materials. (sect. 5.2.)</td>
<td></td>
</tr>
</tbody>
</table>

production by Spallation. Thank to an application of this process invented 14 years ago [14], the Molten-Salt target releases a little energy, but overall a lot of neutrons which transmute Thorium into \(^{233}\text{U}\).

The technical connection between the burners reactors releasing thermal energy by Fission (F-plan),[13] and the production of \(^{233}\text{U}\) by Accelerators (A-plan)[2], opens seducing prospects for the future uses of nuclear energy.

3. PRESENT INDUSTRIAL WORLD DEPLOYMENT

The undeniable penalties supported by the Nuclear Industry grounded on the Uranium Fission, result from the used fuel cycle and the applied technology as shown on Table 1:

From the U-Pu fuel cycle:

- by historical and technical necessity, the U-Pu cycle was the starting point of nuclear energy, but also of Proliferation.
- less than 1% of natural Uranium is fissioned and produces energy, due to enrichment necessity and loss of fissile in the spent fuels.
- it breeds, as main waste, Fission Products (FP), and heavy elements (TRU) with long half-lives and high toxicity. Secondary technological waste appear also in the reprocessing Purex plants. (sect. 5.1.)

From the applied technology in the Fission Industry:

- low thermal efficiency, uncertain safety.
- needs of enriched Uranium for most of the nuclear reactor types;
- weak flexibility in load operation, very limited in size so that the always-large NPP are unexportable in small and poor nations.
- reprocessing of the spent fuels (if any), inefficient for waste management, as well as for the recovery of unused fissile materials. (sect. 5.2.)
4. PROPOSALS OF RATIONAL IMPROVEMENTS

4.1. Safety of Nuclear Reactors

Although there is a general consensus to say that the safety of existing NPP is satisfactory, we know that it is based on stochastic risk analysis, and is relying on engineering safety facilities (ECCS, RHR, ACCS, etc.). These devices aim at correcting the weak points of the fission technology which are the pressure of the coolant, the excess of reactivity of the fuel, the Zirconium cladding chemically reactive with steam, the internal storage of FP...

In our search of rationality, we have to compare these solid fuels reactors which are improved day by day, -with the future hope to become "Inherently Safe"-, to some other proposals with liquid fuels which rely on natural scientific laws.

Table 2
Natural elements and practical separable isotope having tiny thermal cross-sections based on the new data (Igarashi & Furukawa, 1990)

<table>
<thead>
<tr>
<th>Natural element</th>
<th>Cross-section (natural element)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 8O</td>
<td>0.19</td>
</tr>
<tr>
<td>2 2H (100%)</td>
<td>0.59</td>
</tr>
<tr>
<td>3 16O</td>
<td>3.55</td>
</tr>
<tr>
<td>4 116B (80%)</td>
<td>5.6</td>
</tr>
<tr>
<td>5 22ne</td>
<td>6.9</td>
</tr>
<tr>
<td>6 2Be</td>
<td>7.5</td>
</tr>
<tr>
<td>7 35F</td>
<td>9.6</td>
</tr>
<tr>
<td>8 85Kr</td>
<td>33.8</td>
</tr>
<tr>
<td>9 19Ne</td>
<td>39</td>
</tr>
<tr>
<td>10 7Li (92.5%)</td>
<td>45.4</td>
</tr>
<tr>
<td>11 13Mg</td>
<td>53</td>
</tr>
<tr>
<td>12 25K</td>
<td>19</td>
</tr>
<tr>
<td>13 83Kr</td>
<td>171</td>
</tr>
<tr>
<td>14 115P</td>
<td>172</td>
</tr>
<tr>
<td>15 40Zr</td>
<td>185</td>
</tr>
<tr>
<td>16 27Al</td>
<td>231</td>
</tr>
<tr>
<td>17 1H (332 6)</td>
<td>57</td>
</tr>
</tbody>
</table>

Molten-Salt reactors described in this meeting [13], used liquid fluorides mixtures of high thermal capacity, chemically inert to air and water, without pressure. The operating conditions allow to establish a walk-away safety due to a weak excess of reactivity, a negative temperature coefficient and a small residual heat. The engineered safety facilities consist only on static equipment (drain tank). Furthermore, such reactors are so designed that there is no release of FP into the environment in the event of an accident. (no storage of Xenon, Krypton and Tritium in the fuel, no escaping ability for Iodine and Cesium ions which are trapped in the ionic medium)[15-18].

Molten-Salt Accelerators [2] using similar fluorides mixtures with the same macroscopic features, have the same chemical and physical behaviour. Besides they are safer by their subcritical operation.

4.2. General Waste Management.

Nuclear waste are produced from the ore treatment till the eventual reprocessing of the spent fuels, passing by enrichment, assemblies fabrication and electricity production. They are more or less dangerous according their concentration, radioactivity, half-life period, the kind of emitted rays and their ability to enter the living matter.

Till now, the fate of the spent fuels, which are the most dangerous, was either to be reprocessed to recover Plutonium and isolate the high-level radiowaste for storage after vitrification, or to be cooled enough before burying them underground.
Fig 1. Molten-salt breeding fuel-cycle in THORIMS-NES composed of fissile-producers (MSB- and power stations (MSR)
But public who disagree partly with both solutions, welcome the new proposals to transmute FP and to destroy TRU into less dangerous materials, by high neutrons flux irradiation or by spallation.

MSR proponents have preceded this historical change since ORNL people had studied and tested various methods to process on line the Fluoride fuel salt, so as to reach a breeding ratio of 1.06.

4.3. Reprocessing in THORIMS-NES

At the time of ORNL works, the target was to reach a breeding ratio as high as possible, by using materials with very small cross-sections (Table 2), and pulling out of the neutrons thermal flux, by physical or chemical processes, the high cross-section products ($^{149}$Sm, $^{135}$Xe, $^{143}$Nd, $^{147}$Pm, $^{151}$Sm;..), and also $^{233}$Pa, the parent of $^{233}$U, to avoid the formation of $^{234}$Pa.

The whole process, described 25 years ago [19], was achieved in the Fluoride medium.

Nowadays it is proved [20] that the feasible doubling time is 20 years for MSBR and at least 30 years for FBR. These values are too high for bringing a right answer to the fissile needs of the next century, in which the doubling time should be less than 10 years. Also it is more rational and economical to have good converters, easy to operate, without any chemical plant [13], and to have also Accelerators able to produce big amounts of fissile $^{233}$U, with a satisfying doubling time. [2]

But reprocessing has to be done from time to time applying mainly the non-aqueous (or Dry) chemical process experimented by ORNL. At the exception of gaseous FP (Xe, Kr, T), the treatment is done by batches of Fluorides salt, either leaving the burners for extraction of various FP and addition of new fissile ($^{233}$U or Plutonium), or coming from Spallators to take out the Spallation Products (SP) and the production of $^{233}$U, with simultaneous addition of Plutonium.

That is the 1st part of our "D-plan" which is the link, through the chemical reprocessing plants, between Burners and Accelerators.(Fig.1)

4.4. Significant features of waste with THORIMS-NES

Amount of Radio-waste, for a supply of 900 TWe.year: [21]

- Ores treatment: the global demand of Thorium, is surprisingly small and ~2 Mt only for the world in which ~35 % will be transmuted to fission and spallation products.

- Spent fuel reprocessing: the total of FP could be decreased about 30% from that of LWR by the improvement of the thermal efficiency, the produced transuranium elements (TRU) will be two or three order of magnitude less than the amounts in U-Pu fuel cycle, and essentially they are staying in fuel-salt as a fissile or fertile species.

- Today low-level radio-waste are produced at the various stages of the fuel processes: chemical and mechanical fabrication of the fuel, and after irradiation during decladding, chemical reprocessing, and refabrication of the new fuel assemblies. The huge amount of these wastes should be minimized in the order of 1/100 with the dry processes applied to THORIMS-NES.

- Structure material made of Hastelloy N will have an induced radioactivity of 0.7 mrem/hr, after 1 year coming mostly from $^{94}$Nb; it might be fully reused after vacuum remelting.

- Graphite moderator kept in place for the whole reactor life could be partly reused after peeling about 0.5 mm in depth to take away the radio deposits.

Radio-waste Elimination:

- Fissions-Products of half-lives longer than 30 years, which are coming from own MS fission reactors and from other reactors, might be incinerated in THORIMS-NES. [22]
  
  - either in form of molten Fluorides: $^{129}$I, $^{135}$Cs and $^{151}$Sm by MSR or AMSB, and $^{90}$Sr, $^{93}$Zr, $^{126}$Sn and $^{137}$Cs by AMSB;
  
  - or in metallic form casing in graphite, for example: $^{59}$Ni, $^{63}$Ni, $^{94}$Nb and $^{99}$Tc by MSR or AMSB and $^{107}$Pd by AMSB. (these species might be recycled)
TRU are valuable fissile or fertile isotopes, and should be effectively utilized as fission reactor fuels not reproducing them. For this purpose MSR will be the best facility. In practice, we are examining the FUJI IV, modifying the composition of FUJI II, in which about 1/3 of $^{233}\text{U}$ is replaced by TRU of 100~200 Kg; and fertile ThF4 is decreased from 12 mole% to 8 mole% resulting in the fissile, which is supported by the further addition of TRU in burning rate of about 50 kg/year.

5. FROM THE PAST TO THE FUTURE

Another rational choice to do, is to find the best means for passing from the present nuclear industry, which should burn solid fuels with more or less Thorium matrix, towards MS applications in THORIMS-NES [2; 13].

The junction between both fields, that is to say between "PAST" and "FUTURE", is the reprocessing of the spent solid fuels.

5.1. Purex process

The present debate between OPENED FUEL CYCLE (OFC) and CLOSED FUEL CYCLE (CFC) result from the incapability of the Purex process to treat the whole spent fuel production. Practically, this technic is convenient only for ores treatment (U or Th), and for military Pu extraction from slightly irradiated fuels.

The weak points of Purex process are as follows:

- Light water, used as solvent, induces criticality hazards which prevent to have vessels bigger and bigger as it is usual (for economy) in chemical industry.
- Nitric acid and TBP molecules, sensitive to the various rays of the spent fuels, are partly radiolysed, and give by-products which disturb the basic process and need to be destroyed or eliminated as radio-waste.
- With this process, all radio-waste are diluted in enormous liquid volumes flowing in numberless pipes and sub-critical apparatus.... for finally being re-concentrated in volumes as small as possible. It results a large production of contaminated materials and many radioactive solutions to manage.
- Whatever may be the fuel burn-up, the same process is applied, in opposition to the general chemical rule which is to choose a suitable process for each composition range.

5.2. DRY processes

This proposal to use a non-aqueous process, and chemical mineral products stable under atomic rays, avoid all the former difficulties: criticality hazards, radiolyse, dilution...etc

These DRY processes are lying on well known laws of thermo & electro-chemistry, and use the numerous data about oxides, chlorides, fluorides compounds for all the elements of the Mendeleieff Table. (ANL-5750, JUL-565-CT, IAEA-STI/PUB/424, ..etc)

Their main features are as follows:

- Mediums in which these processes are performed -solid, salty or gaseous phases- have no moderators. This allows operation with fissile and radioactive materials of high concentration.
- The stability of reagents makes these methods highly productive, compact, and also forms small quantities of waste.
- Dry fuel reprocessing methods (Fluorides volatilization, pyro-electrochemical or irradiated metal refining) allow to treat fuels of any burn-up and cooling time, with a small amount of technologic stages.

The Fluorides volatilization process studied and experimented for UO2-PuO2 fuels by french-russian teams [23] allows the recovering of Uranium with a decontamination factor of 10E6. An experiment showed that this method would be sufficient for Uranium extraction from U-Th fuel. [24]

The Fluorides salt of MSR and AMSBs are tightly connected with the volatilization process; 15 years ago, neutronic calculations have shown that the LWR spent fuel can be re-
used (after a coarse treatment by Fluorine) as fuel for MSRs; in that application, MSRs can
be the bins of LWRs.[25]

Thus, in our D-plan of THORIMS-NES, the Fluorides volatilization process is the key
to obtain the higher efficiency from the present nuclear fission industry. It allows:
-the elimination of the low toxicity waste
-the recycling of the other PF, Actinides, TRU, to the Burners (F-plan) or to the
Accelerators (A-plan), according their neutronics properties, and the Global energy
demand.

6. RATIONAL PROSPECTS

Let us consider, lastly, two particular prospects for Nuclear Energy:

Nuclear Fission Industry should be used everywhere in the world, and chiefly in
small and poor nations where there is a deep lack of clean and cheap energy. To reach that
goal there is a huge need of small and economical power plants.

Till now, almost the nuclear fission energy was produced for electrical purposes. But
electricity cannot be stored, which is a high penalty for solid fuels reactors of weak
flexibility in power production. Chemical applications of nuclear heat should fill that lack
by increasing the enthalpic content of the products involved in the chemical reactions.

6.1. Small economical Reactors

Nuclear island is only a part of electricity plant, so that the price of energy supplied,
for all facilities using solid fuels, is rather independent of the nuclear type. But the cost of
small power stations is bigger because it is necessary, for each size, to make specific applied
technologic researches about:
-definition and special fabrication for each part of the nuclear cell: reactor vessel,
exchangers, pumps...
-calculations, fabrication and tests for fuels, cladding, assemblies....
-studies and fabrication of internal core structur, control rods, loading machine, safety
devices...
-rules for normal operation, loading and unloading of fuel, emergency conditions...
-spent fuel storage management....

Thus, small nuclear reactors, issue from the old technologies, are as complex as the
big ones, and they are not more flexible; they are freezeed for fuel composition, nature of
production (which is generally the unstorable electricity) and the range of power supplied..

Small MSR have the same economical advantages already described here for the bigger
ones, such as operation, maintenance work, fuel management without fuel handing machine,
etc.[2;13]. But with MSR, the nuclear island should be rather independent of the size of the
plant:
-no specific fabrication and tests of fuel according the size
-no specific and complex safety devices

In addition to these features, comes the specific quality of large MSR which is the
flexibility in power from 10 to 100 %, according the hours, seasons, etc.. So there is no
need to conceive several nuclear reactors of various sizes to answer the various demands.
With a convenient overlapping, two sizes should be theoretically sufficient. For example:
-The Small size, like FUJI I (400 MWth nominal) running from 40 to 400 MWth.
-The Big size, like FUJI II (2200 MWth nominal) operating from 220 to 2200 MWth.

So, it appears that Small MSRs [26-28] could be the energetic tools, escorting the
spontaneous progress of the Developing Nations.

6.2. Chemical heat Applications

Nuclear heat is very little utilized for chemical applications; the main reason is that
the highest temperature in all classical LWR is not sufficient to perform chemical reactions.

With MSRs which operate at higher temperature (530 to 700°C) the thermal
conditions are much better. By the high temperature of the fuel salt (700°C and more)
easily reached with the alloy Hastelloy N, MSRs can supply district and industrial heat (oil deep refining plants, synthetic fuel production...), in better conditions than HTGR, because of the high thermal capacity of the Fluoride salt, which avoids a too large drop of temperature during the heat exchange process.

More interesting for the environment, is the possibility to produce hydrogen by reaction between natural gas (CH4) and steam. [29] Two reactions, called "the methane reforming", and the "Shift reaction" used straight the heat of the salt without passing by electricity production and water electrolysis.

For higher temperatures, easily accepted by the salt, it is proposed, in the future, to use new types of graphite (like glass-fiber carbon), and to realize heat transfert in form of radiant heat exchange. To overcome the high temperature pump problem, it is suggested to use a natural convection regime, and/or gas-lifting.[30]

7 CONCLUSIONS

After 30 years of theoretical and applied research in connection with a strong effort for the industrial development, which have shown the advantages and the limits of the nuclear fission energy, the time is come to give a new impulse by rational choices, to increase both the use of its potenialities, and its acceptance by the public.

Some progresses should be obtained in that purpose by the shifting from the U-Pu cycle to the Th cycle, with simultaneous burning of the big Plutonium stocks. However it is possible to go further with deep improvements in the other fields concerned by the acceptability of Fission Energy: Safety, Waste management, Proliferation, Chemical applications..., by rational issues about the technologies to use for its future industrial deployment.

The applications of Molten-Salt technology, well known by the international nuclear community, can be this essential mutation, by offering a Global strategy for the supply of a clean Energy, in big amounts, everywhere in the World, and for a long time.

The main rational choice for International deciders is to find the right balance between the weight of the Past and the necessity to prepare the Future through an International cooperation.

REFERENCES


SESSION 2: GAS COOLED REACTORS AND THORIUM ASPECTS
PLUTONIUM DESTRUCTION WITH PEBBLE BED TYPE HTGRs USING Pu BURNER BALLS AND BREEDER BALLS

K. YAMASHITA, K. TOKUHARA, R. SHINDO
Department of HTTR Project,
Japan Atomic Energy Research Institute,
Japan

Abstract

It was made clear that pebble bed type HTGRs using Pu burner balls (Pu balls) and breeder balls (Th balls) possesses a potential to burn weapons-grade Pu to 740 Gwd/T\textsubscript{Pu}. The total amounts of Pu and \textsuperscript{239}Pu of can reduced to about 20 and 1 %, respectively.

1. Introduction

There are some problems to be solved in the nuclear design when plutonium is burned in reactors. Power coefficients may become positive due to giant fissile resonance peak of \textsuperscript{239}Pu in thermal energy range. Abrupt power excursions may occur due to small delayed neutron rate $\beta$ of Pu when a positive reactivity is added to reactors e.g. by an unanticipated control rod withdrawal. The pebble bed type HTGR is one of the most promising reactors that can solve the problems. The first problem will be avoided by mixing Pu with fertile material like Th or U. Secondary, the reactivity addition by unanticipated control rod withdrawal will hardly occurs in the operation condition because the excess reactivity for burnup is not necessary due to the continuous fuel loading, and the reactor is operated in the condition where most of the control rods are fully withdrawn[1]. Furthermore, the fuel temperature does not rise abruptly due to the large heat capacity of graphite in core even if a positive reactivity is added by some accidents.

However, a problem remains that fissile materials transmuted from fertile materials exist in the spent fuel even if most of initial Pu is burned up. The residual fissile material made difficult to dispose the spent fuel directly. Otherwise, the reprocessing of the spent fuel is necessary for the effective use of the fissile materials.

Thus, we have found a reactor system in which Pu can be burned out so that the reprocessing of spent fuels is no more necessary. This system is a pebble bed type HTGR using Pu burner balls (Pu balls) and breeder balls (Th balls). The Pu ball contains plutonium not mixed with fertile material of Th. The Th ball contains only fertile material of Th. The core is loaded with these two types of fuel balls randomly. They are removed continuously from the core and reloaded into the core so far the burnups is achieved to the expected values. This system can be applied for burning of Pu obtained from nuclear weapons destructed or spent fuels of LWR etc.

This paper presents the principle of the reactor system and the burnup characteristics when weapons-grade Pu (94 \% 239Pu and 6 \% 240Pu) is burned in the reactor system.

63
2. Movements of balls in core

The movements of the Pu balls and Th balls are shown in Fig. 1. The multi-pass fuel loading is applied for the reactor system so that most of Pu in the Pu ball will be burned up ultimately. The burnups of the Pu balls are evaluated by measuring the strength of γ-lays after they are exhausted from the core. The Pu balls will be reloaded again if the burnup is less than the expected burnup. The Pu balls will be discharged from the reactor system as spent fuels if the burnup reaches to the expected value. The excess reactivity necessary for the reactor operation is maintained by loading fresh Pu balls from top of the core. The Th balls are reloaded repeatedly so far as the fuel integrity is maintained. The technic for the multi-pass fuel loading has been accumulated through the operation of the pebble bed type experimental HTGR (AVR) in Germany for about 20 years. This technic can be applied for the reactor system[2].

2. Destruction of Pu

The achievable maximum burnup of the Pu ball was evaluated with the following relation[3]:
Table I Specifications of core and fuel

| Specifications referred to Russian HTR-Module (5) in development

<table>
<thead>
<tr>
<th></th>
<th>200 MW</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermal power*</td>
<td>200 MW</td>
</tr>
<tr>
<td>Core height*</td>
<td>9.4 m</td>
</tr>
<tr>
<td>Core diameter*</td>
<td>3.0 m</td>
</tr>
<tr>
<td>Moderator</td>
<td>Graphite</td>
</tr>
<tr>
<td>Coolant</td>
<td>Helium</td>
</tr>
<tr>
<td>Pu ball Pu amount</td>
<td>0.5 g/ball</td>
</tr>
<tr>
<td>Pu amount Number in core</td>
<td>about 180000</td>
</tr>
<tr>
<td>Th ball Th amount</td>
<td>14 g/ball</td>
</tr>
<tr>
<td>Th amount Number in core</td>
<td>about 180000</td>
</tr>
</tbody>
</table>

\[ B_n = \frac{2n \cdot B_1}{n+1} \]

where

B_n: burnup of spent fuel of reactors in which the whole fuels are replaced with n-times of reloading (GWD/T)
B_1: burnup of spent fuel of reactors in which the whole fuels are replaced for once (GWD/T).

The burnup of the pebble bed type HTGR (B_n) is twice than B_1 because the n can be regarded as the infinite due to the continuous loading. B_1 was calculated with HTGR fuel lattice burnup calculation code DELIGHT[5]. The specifications in Table I were used for the evaluation of burnup. A simplified model is used for the evaluation of the burnup. Pu, Th and \(^{235}\)U are distributed in the fuel kernel of the coated fuel particles (CFPs) in the ball fuel in the evaluation model. The effective cross section of Th was evaluated by the resonance calculation. The change in the effective multiplication factor k_{eff} is given in Fig. 2. It is shown in Fig. 2 that B_1 becomes 370 GWD/t_Pu. The

![Fig. 2 Change in effective multiplication factor with burnup of Pu ball](image-url)
achievable maximum burnup is 740 GWd/t\textsubscript{Pu}. This is obtained by substituting 370 Gwd/t\textsubscript{Pu} into \( B_1 \) in the equation (1). The change in Pu isotopes with burnup is shown in Fig. 3. The amount of total Pu for 740 GWd/t\textsubscript{Pu} is 20\% of the initial value. The amount of \( ^{239}\text{Pu} \) becomes less than 1\% of the initial values.

The Department of Energy in USA (USDOE) has reported study on Pu disposition for the advanced LMR (ALMR) and advanced BWR (ABWR)[6]. Table II shows residual amounts of Pu isotopes when the weapons-grade Pu of 1000 kg is burned with the ALMR, ABWR and the reactor system conceived here. Pu fuel without fertile materials was burned in case of the ALMR. Pu was mixed with natural uranium in the case of the ABWR. The residual amount of

Table II Residual plutonium and generated energy by loading of 1000 kg plutonium in different reactor types

<table>
<thead>
<tr>
<th>Before burnup</th>
<th>After burnup</th>
<th>This HTGR</th>
<th>ALMR*</th>
<th>ABWR*</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total Pu(kg)</td>
<td>1000</td>
<td>199</td>
<td>618</td>
<td>648</td>
</tr>
<tr>
<td>( ^{239}\text{Pu} )(kg)</td>
<td>940</td>
<td>5</td>
<td>476</td>
<td>279</td>
</tr>
<tr>
<td>( ^{240}\text{Pu} )(kg)</td>
<td>60</td>
<td>72</td>
<td>130</td>
<td>233</td>
</tr>
<tr>
<td>( ^{241}\text{Pu} )(kg)</td>
<td>0</td>
<td>55</td>
<td>12</td>
<td>97</td>
</tr>
<tr>
<td>( ^{242}\text{Pu} )(kg)</td>
<td>0</td>
<td>66</td>
<td>0</td>
<td>39</td>
</tr>
<tr>
<td>Generated energy (J)**</td>
<td>------------</td>
<td>1.1\times10^{14}</td>
<td>4\times10^{13}</td>
<td>9.2\times10^{13} ***</td>
</tr>
</tbody>
</table>

* : They were referred to "plutonium disposition study" in USDOE[7].
** : Energy generated by burnup of fissile materials converted from Th and 238U was included.
*** : Enrichment was regarded as 3.5\%o. This was evaluated from burnup of 37.1GWD/t given in reference[7].
total Pu for the reactor system is about one-third of the ALMR and ABWR. The residual amount of $^{239}$Pu is much less than ALMR and ABWR. The generated energy by loading of 1000 kg for the reactor system is larger than those of the ALMR and ABWR. It results from burnup of $^{233}$U converted from Th. From the results, it can be concluded that the quality of the weapons-grade Pu can be strongly degraded by the reactor system and the Pu can be converted to energy effectively.

The amount of $^{233}$U in Th ball will be saturated to 0.2 g after a certain duration in the core. The burnup of the Th ball is about 7 GWD/t$_{Th}$ for a year. The Th ball can be used for 16 years if it can be supposed that the integrity is maintained to the average burnup of Thorium, Hochtemperatur-Reaktor(THTR-300)(109 GWD/t)[7].

3. Maximum amount of Pu in Pu ball

Power is large in the fresh Pu ball. The power of the Pu ball decrease with progress in burnup. The maximum amount of Pu in Pu ball is limited by the maximum permissible power in a ball. The power of 4.5 kW was used in the design of HTR module as the maximum permissible. This was determined by the maximum permissible thermal stress in the ball. Figure 4 shows the change in the power of Pu ball with burnup days when the Pu ball and Th ball are loaded with 0.5 g Pu and 14 g Th, respectively. Axial power peaking is not considered in power change in Fig. 4. The maximum power appears at beginning of burnup. The real maximum power in the core is about 4 kW. This was obtained by using the axial power peaking of the HTR module (1.7)[8]. The real maximum power is lower than the maximum permissible power of 4.5 kW. It can be concluded from the results that the optimum amount of Pu in the Pu ball is 0.5 g when the core is loaded with the same number of Pu balls and Th balls. The power of the Th ball is almost constant after the saturation of $^{233}$U concentration. About 66000 Pu balls, namely 33 kg of Pu, can be burned by the reactor system specified in Table I for a year.

![Fig. 4 Change in power of a Pu ball with burnup](image-url)
4. Conclusion

It is made clear that the burnup of 740 GWD/t\textsubscript{\textit{Pu}} can be achieved by using the pebble bed type HTGR using the Pu balls and Th balls.

The amount of Pu can be reduced to about 20% of the initial value. The final total amount of fissile materials in the spent Pu ball will be less than 1% because \textsuperscript{241}Pu will decay to Am. The substantive amount of fissile material is a small amount of \textsuperscript{239}Pu in the Pu ball. It could be concluded from very low quality of Pu that it is no more worthy of reusing it for the weapon and reprocessing for new fuel production. Therefore, the spent Pu balls can be disposed in the rock salt mining directly[9].

Energy generated by the reactor system is more for the same Pu amount than those of the ALMR and ABWR.

The irradiation test of coated fuel particles of Pu (Pu-CFP) had been done by in Belgonucleaire in 1974. It was shown the integrity of the Pu-CFP was still maintained for burnup of 600 GWD/t\textsubscript{\textit{Pu}}[10]. The density of the fuel kernel was lowered in the CFP to avoid the pressure rise by fission products. Further reduction in the fuel kernel density may be necessary to realize the burnup of 740 GWD/t\textsubscript{\textit{Pu}}.

The amount of Pu destructed in the reactor system is only 33 kg for a year. It is small against total amount of Pu produced by the destruction of nuclear weapons. The reason for the small amount of Pu destructible is that Pu is burned in the reactor system effectively. However, the problem can be solved by the spiking of Pu balls. The Pu balls are made radioactive by loading in the reactor core so that the diversion of Pu will be made difficult. Large number of Pu balls can be made radioactive in the reactor system because Pu balls is continuously irradiated in operation in the pebble bed reactor. The irradiated Pu ball will be stored temporary and be burned again in the reactor system.

ACKNOWLEDGMENTS

The authors express their sincere thanks to Messrs. I.Murata, A.Saikusa, S.Shiozawa, T.Tanaka and T.Tobioka for valuable comments and support.

REFERENCES

THE PLUTONIUM CONSUMPTION MODULAR HELIUM REACTOR (PC-MHR)

D. ALBERSTEIN, A.M. BAXTER, W.A. SIMON
General Atomics,
San Diego, California, USA

Abstract

The Plutonium Consumption Modular Helium Reactor (PC-MHR) can effectively destroy excess weapons grade plutonium, consuming more than 90% of the initially charged plutonium-239 and 65% of the initial total plutonium in a once through cycle - high plutonium destruction without recycle. Because the PC-MHR uses no fertile fuel, no new plutonium is created. The spent fuel discharged from the PC-MHR has characteristics that give it unusually high resistance to diversion and proliferation. The amount of plutonium per spent fuel element is very low; the spent fuel element mass and volume are high; there is no developed process for separating the residual plutonium from the spent fuel; and the discharged plutonium isotopic mixture is severely degraded - well beyond typical "reactor grade" plutonium. The discharged fuel elements are suitable, with no further processing, for direct disposal in a permanent geologic repository. The PC-MHR has the unique safety characteristics of other MHR designs. The MHR safety design objectives are met through inherent safety features and design selections that take maximum advantage of these features. Some of these features include: (1) helium coolant, which is single phase and inert with no reactivity effects; (2) graphite core, which provides high heat capacity and structural stability at very high temperatures; (3) refractory coated plutonium oxide particle fuel, which allows extremely high burnup and retains fission products at temperatures much higher than normal operation; (4) negative temperature coefficient of reactivity, which inherently shuts down the core above normal operating temperatures; and (5) an annular, low power density core in an uninsulated steel reactor vessel surrounded by a reactor cavity cooling system, which enables passive heat transfer while maintaining fuel temperatures below damage limits. The PC-MHR can be deployed in the near term because it is based on technology demonstrated by more than 50 gas-cooled reactors built and operated worldwide since 1956.

1. INTRODUCTION

As a result of recent arms reduction agreements between the United States and Russia, large quantities of weapons grade plutonium and uranium will be declared excess by the governments of the two countries. While the excess highly enriched uranium can be relatively easily dispositioned by diluting (or denaturing) it with natural uranium and using it as fuel in commercial nuclear reactor systems, disposition of excess weapons grade plutonium presents a much more difficult challenge. A number of options for disposition of excess weapons grade plutonium are being evaluated in the United States and in Russia. Although discussions often focus on use of light water reactors (LWRs) fueled with mixed oxide (MOX) fuel assemblies containing weapons grade plutonium or on use of liquid metal reactors fueled with large quantities of plutonium, other innovative options are available for consuming excess weapons grade plutonium inventories and producing a discharged product that is unattractive for use in weapons applications and, as a result, presents no incentive or need for reprocessing and recycle.
Among the options being evaluated in the United States for disposition of excess weapons grade plutonium is the modular helium reactor (MHR), a graphite moderated, helium cooled reactor system. The Plutonium Consumption MHR (PC-MHR) offers high levels of plutonium destruction (more than 90% of the initially charged plutonium-239 and more than 65% of the initial total plutonium) in a single pass through the reactor - high plutonium destruction without recycle. This high level of plutonium destruction is achieved because the PC-MHR uses no fertile fuel (no new plutonium is created) and because its refractory coated particle fuel is capable of reaching burnup levels in excess of 750,000 MWd/t heavy metal while fully retaining its structural integrity.

The MHR offers a unique combination of advantages for the disposition of weapons grade plutonium. In addition to its ability to achieve high levels of plutonium destruction without recycle, the PC-MHR fuel cycle has unusually high resistance to diversion and proliferation. The plant features all of the unique safety characteristics of the uranium-fueled versions of the MHR. The safety design objectives of the MHR are met through a combination of inherent safety features and design selections that take maximum advantage of these features. The plant relies on passive features to control heat generation and removal under accident conditions without dependence on operator intervention or active, A-C powered safety systems.

2. PC-MHR PLANT DESIGN

The MHR has been under development by the U.S. Department of Energy (DOE) since 1983 as part of DOE’s advanced reactor program. The design draws upon the considerable gas-cooled reactor technology base. More than 50 gas-cooled reactors have been built and operated worldwide since 1956. As a result of this experience, the PC-MHR can be deployed in the relatively near term without need for extensive fundamental research and development. Primary engineering development needs relate to fabrication processes for plutonium fuel in a large scale application and to integration of power conversion system components for the reference design recently adopted for the commercial MHR program.

Two versions of the MHR have been under development in the U.S. in the commercial MHR program. The reference plant design up to 1993 consisted of four 450 MWt reactor modules, each of which is coupled to a steam generator that produces high temperature and pressure steam for a conventional Rankine cycle steam-electric power generating system. A four module facility operates at a net plant efficiency of 38%, resulting in net electrical generating capacity of 692 MWe.

In 1993 extensive studies were conducted on the commercial MHR program to determine the suitability of replacing the steam cycle power conversion system with a closed Brayton cycle gas turbine power conversion system. Use of the Brayton cycle is desirable because it increases the plant efficiency from 38% to about 48%. This dramatic improvement in thermal efficiency results in substantial improvement in plant life cycle costs, less thermal discharge to the environment, less radioactive waste produced per unit of electrical energy generated, and maximum utilization of the energy content of the fuel. In addition, the evaluations examined the feasibility of increasing the thermal power rating of the reactor to the maximum that can be accommodated within the same reactor vessel size.
Based upon these studies it was determined that a 600 MWt reactor core coupled to a helium gas turbine direct cycle power conversion system could be adopted as the commercial MHR reference design. A four module reference MHR plant of this design can produce 1144 MWe from 2400 MWt of reactor power.

As part of the DOE sponsored Plutonium Disposition Study, both the 450 MWt steam cycle MHR and the 600 MWt gas turbine MHR have been assessed. The plutonium consumption, diversion/proliferation resistance, and safety characteristics of the two plants are essentially the same. The plants differ primarily in the number of reactor modules that need to be deployed to disposition a given inventory of weapons grade plutonium within a specified time period (as a result of their different thermal power ratings), in the plant life cycle costs (as a result of their different plant efficiencies, their different power ratings, and their different capital costs), and in the amount and complexity of the power conversion equipment.

DOE has not, as of the time of preparation of this paper, yet published the result of its evaluations of the gas turbine MHR for plutonium disposition. Hence, the PC-MHR design described in this paper is the steam cycle version of the design that uses a 450 MWt reactor module.

The steam cycle PC-MHR reference plant consists of four 450 MWt reactor modules capable of providing a net electrical generating capacity of 692 MWe. The reactor core is contained in an uninsulated steel reactor pressure vessel that is connected by a cross vessel to a steam generator vessel. All three vessels are designed to ASME Section III Boiler and Pressure Vessel Code requirements. They are located below grade in a high pressure, low leakage containment with characteristics typical of containments of U.S. commercial light water reactors. The reactor cavity within the containment is cooled by a passive, natural convection, water cooled reactor cavity cooling system. This system operates at all times. Under accident conditions it provides for passive removal of reactor decay heat via conduction and radiation from the uninsulated reactor vessel to the cooling system panels. Heat is transported by the reactor cavity cooling system via natural convection to a heat exchanger, where it is rejected to the atmosphere, which is the ultimate heat sink. The reactor, steam generator, cross vessel, and primary heat transport system arrangement are shown in Figure 1.

2.1 PC-MHR Fuel and Core Design

Standard refractory coated particle fuel, shown in Figure 2, is used in the PC-MHR. Coated particle fuel has been used in high temperature helium cooled reactors for more than 25 years. The fuel is in the form of tiny plutonium oxide fuel kernels coated first with a porous graphite buffer layer followed by layers of silicon carbide and pyrolytic carbon. This system of particle coatings is referred to as a TRISO coating. The diameter of the coated fuel particle is approximately 615 μm. The particles are mixed with graphitic material and formed into cylindrical fuel rod compacts approximately 12.45 mm in diameter and 49.3 mm in length. The fuel rod compacts are inserted into hexagonal prismatic graphite fuel element blocks, as shown in Figure 2. The completed fuel element is approximately 0.79 m in height and 0.35 m wide across the flats, and it weighs about 115 kg. A standard fuel element contains 202 fuel holes, 108 coolant channels, about 3000 fuel rod compacts, and about 22 million coated fuel particles. Extensive operation and test data on refractory coated fuel particles confirm their performance up to and beyond the maximum temperatures that are expected to occur in design basis accidents.
FIGURE 1: PC-MHR HEAT TRANSPORT SYSTEM
Plutonium utilization is not a new application for high temperature helium-cooled reactors. Six irradiation tests have been conducted using high-enriched (up to 88% plutonium-239) plutonium coated particle fuel. Five of these tests were performed as part of the Dragon Reactor Project (United Kingdom) in the late 1960s, and the sixth was performed by General Atomics in the Peach Bottom gas cooled reactor in the early 1970s. For each of these tests, the fuel consisted of TRISO coated plutonium fuel kernels. The Dragon tests consisted of loose particles in graphite holders. The fuel for the Dragon tests was fabricated by Belgonucleaire in Europe. For the Peach Bottom test, standard fuel rod compacts were fabricated at Oak Ridge National Laboratory. Test conditions included irradiation at temperature up to 1450°C, peak fuel burnup up to 747,000 MWd/t, and fast neutron fluence up to $2.2 \times 10^{25} \text{n/m}^2 (E > 0.18 \text{Mev})$. These temperature and burnup conditions are expected to envelope those experienced by the coated fuel particles in a PC-MHR. For all of these tests, the plutonium fuel performed well, establishing the feasibility of high burnup, TRISO-coated, plutonium fuel for the MHR.

The reactor core consists of 840 hexagonal prismatic fuel elements stacked in an annular array of 84 columns, 10 elements high, as shown in Figures 3 and 4. One half of the fuel elements is replaced annually. Because the PC-MHR uses no fertile fuel material, excess reactivity control and negative temperature feedback is provided through use of other materials. For the PC-MHR these functions are provided by use of erbium oxide poison rods located in selected fuel holes in the graphite fuel elements. Use of graphite as moderator and structural material in the PC-MHR results in a thermal neutron spectrum that is relatively hard compared to typical light water reactors, i.e., the peak and average neutron fluxes are at higher energy levels. The harder spectrum is such that the 0.3 eV plutonium-239 thermal absorption resonance lies within the thermal flux spectrum, which enhances the plutonium-239 consumption rate relative to other nuclides. Furthermore, if the core heats up above normal operating temperatures, which would occur during certain design basis events, the thermal flux would shift into the strong erbium absorption resonance and then into the large, 1.0 eV plutonium-240 capture resonance, providing negative reactivity feedback to terminate the transient.

2.2 PC-MHR Safety Design

The PC-MHR features the same unique safety characteristics of previous uranium-fueled, steam cycle MHR designs. The safety design of the PC-MHR is based on retaining virtually all fission products within the fuel particle coatings under all accident conditions. The integrity of the particle coatings as a barrier to radionuclides is maintained by controlling core heat generation, assuring adequate means of heat removal, and limiting air/water chemical reactions with the core graphite or the fuel particles. The safety design objectives of the MHR are met through a combination of inherent safety features and design selections that take maximum advantage of these features. Some of these features include: (1) helium coolant, which is single phase and inert with no reactivity effects; (2) graphite core, which provides high heat capacity, slow thermal response, and structural stability at very high temperatures; (3) refractory coated plutonium oxide particle fuel, which allows extremely high burnup and retains fission products at temperatures much higher than normal operation; (4) negative temperature coefficient of reactivity, which inherently shuts down the core above normal operating temperatures; and (5) an annular, low power density core in an uninsulated steel reactor vessel surrounded by a reactor cavity cooling system, which enable passive heat transfer to the ultimate heat sink while maintaining fuel temperatures below damage limits.
FIGURE 2: PC-MHR FUEL ELEMENT COMPONENTS

- Fuel Particle
  - TRISO coated particle
  - Pu kernel
  - Porous carbon buffer
  - Inner isotropic pyrolytic carbon
  - Silicon carbide barrier coating
  - Outer isotropic pyrolytic carbon

- Fuel Compact
  - TRISO coating
  - 1.94 in. height
  - 0.49 in. diameter

- Prismatic Fuel Element
  - 31.4 in. height

[Diagram of fuel particle, fuel compact, and prismatic fuel element]
• 36 Control Rods (in core & outer reflector)
• 12 Reserve Shutdown Channels (in core)
INNER REPLACEABLE REFLECTOR

FUEL ELEMENTS

(ACTIVE CORE BOUNDARY)

OUTER REPLACEABLE REFLECTORS

UPPER REPLACEABLE REFLECTOR

LOWER REPLACEABLE REFLECTOR

FIGURE 4: PC-MHR REACTOR CORE ELEVATION
These safety design features result in a reactor that can withstand loss of coolant circulation or even loss of coolant inventory and maintain fuel temperatures below damage limits. No credible severe accident scenario involving a large release of radionuclides from the core has been identified for the PC-MHR. Extensive analyses of a spectrum of design basis events and beyond design basis events confirm that the safety characteristics of the PC-MHR are the same as those of earlier uranium-fueled MHR designs. Accordingly, it is expected that regulatory requirements will be met with margin and that offsite doses resulting from design basis accidents will be less than the threshold at which protective actions are required for the offsite population.

3. PLUTONIUM DESTRUCTION CAPABILITY

Figure 5 presents a comparison of the plutonium destruction capability of various plutonium disposition options. No recycle of spent fuel is assumed for the reactor options. It is clear from this figure that the vitrification option, in addition to recovering none of the energy content of the weapons grade plutonium, does nothing to diminish the usefulness of the material in weapons applications. Furthermore, options that reduce the weapons grade material to a discharge equivalent to that contained in commercial reactor spent fuel (the "spent fuel standard" referred to in the U.S. National Academy of Sciences report on disposition of excess weapons grade plutonium) recover the energy content from only 25-30% of the plutonium and leave a discharged isotopic mixture that can be (and has been) used in a nuclear weapons application.

Figure 5 indicates that the PC-MHR is the most effective option for destroying weapons grade plutonium without recycle and maximizing the energy content recovery from the material. The PC-MHR is capable of consuming more than 90% of the

![Figure 5: Plutonium Destruction Capability of Various Disposition Options](image-url)
initially charged plutonium-239 and more than 65% of the initially charged total plutonium in a single pass through the reactor. The discharged fuel is less than 30% plutonium-239 and contains large quantities of plutonium-240 and -242, which significantly reduces its usefulness in weapons applications. Buildup of actinides other than the higher isotopes of plutonium in the spent fuel is relatively small - equivalent to about 2% of the total actinide discharge, predominately americium-241 and -243.

Depending on the specifics of the fuel cycle chosen, a four module PC-MHR plant can process 0.9 to 1.1 metric tonnes of weapons grade plutonium annually to the level of destruction shown in Figure 5. Over a nominal 40 year design lifetime, a four module plant would process approximately 36 to 44 metric tonnes of material. The plutonium processing rate is directly proportional to the number of reactor modules deployed and to the thermal power rating of each module.

The plutonium destruction capability of the PC-MHR is not limited to the consumption of weapons grade plutonium. Evaluations by General Atomics show that the high levels of weapons grade plutonium destruction made possible by the high burnup capability of the coated particle fuel can also be achieved by using commercial reactor grade plutonium as the feed material. Therefore, the PC-MHR offers a relatively near term option for solving a problem that has been recently characterized by the U.S. National Academy of Sciences as requiring long term steps to resolve - reducing the proliferation risks posed not only by excess weapons grade materials but also by the large and growing inventory of plutonium in commercial reactor spent fuel.

4. RESISTANCE TO DIVERSION AND PROLIFERATION

The PC-MHR fuel cycle offers a very high level of resistance to diversion and proliferation. The plutonium content of each fresh fuel element is very low (less than one kilogram), and the plutonium is much more difficult to recover from fresh PC-MHR fuel than from fresh MOX fuel assemblies. It is very difficult to divert a sufficient quantity of PC-MHR spent fuel to obtain a weapons-useful quantity of plutonium; the technology for separating plutonium from MHR spent fuel has not been developed; and the isotopic discharge of the spent fuel makes it unattractive for weapons applications.

Diversion of PC-MHR spent fuel is difficult for all the reasons that apply to any reactor spent fuel (high radioactivity, large mass, high internal heat generation rate, extensive safeguards and security, etc.), but also for one reason that is unique to the PC-MHR: low plutonium content per fuel element. While the plutonium content in a single LWR MOX spent fuel assembly is of sufficient quantity to use in a weapons application, one would have to divert up to seventeen truckloads of PC-MHR spent fuel elements (each element weighing about 115 kilograms) to obtain a similar quantity of discharged plutonium. The diversion of up to seventeen truckloads of spent fuel shipping casks is a considerably more daunting and readily detectable task than diversion of one truckload of LWR spent fuel.

Even if such a large quantity (both mass and volume) were diverted, the technology for separating plutonium from MHR spent fuel has not been developed, in contrast to the LWR, for which plutonium separation technology is well established and widely known. If a process for separation of plutonium from PC-MHR spent fuel were developed, the amount of equipment required would be much more extensive than for other fuels, and the number and complexity of basic processing steps would be much higher.
Finally, although any mixture of plutonium isotopes can, in principle, be used to make a nuclear device, the plutonium discharged from the PC-MHR, being particularly high in plutonium-240 and -242 content, is much more difficult to use in weapons applications than the discharge from reactors that do not achieve as high a burnup level. The increased difficulty in weapons design, construction, and delivery results from the higher critical mass required, the higher spontaneous neutron emission rate (which increases yield uncertainty), the higher radiation hazard (which makes it more difficult to protect assembly personnel from excessive radiation exposure), and the higher decay heat generation rate from the discharged plutonium (which makes it more difficult to design and assemble a device and to maintain the tight mechanical tolerances required). In addition, as a result of the relatively short half life of plutonium-241 (14.4 years), which constitutes approximately 33 to 38% of the PC-MHR plutonium discharge, the required critical mass will increase with time after separation, further complicating the nuclear design of a device.

The diversion and proliferation resistance of the PC-MHR makes it well suited for international deployment for plutonium disposition. The processes involved in fabrication of PC-MHR fuel, reactor operation, and spent fuel disposition lend themselves to full implementation of IAEA safeguards controls. Discussions were begun in June between General Atomics and the Russian Ministry for Atomic Energy (MINATOM) regarding replacement of the plutonium production reactors at Tomsk with PC-MHRs.

5. SPENT FUEL DISPOSITION

The low plutonium content per spent fuel element and the undesirable mixture of plutonium isotopes in the spent fuel leave little need or incentive for reprocessing or recycle of the PC-MHR spent fuel. Evaluations by both General Atomics and Oak Ridge National Laboratory in the U.S. indicate that the refractory spent fuel elements are an excellent waste package that is suitable for direct disposal, in whole element form inserted into a multipurpose canister, in a geologic repository. The TRISO coated particle fuel has the benefit of long term containment of radionuclides without relying on the performance of the waste package. Quantitative assessments show that the TRISO coating should maintain its integrity for a million years or more in a repository environment. In contrast, the expected lifetime of zircalloy cladding in a repository, even under dry conditions, is less than 1000 years.

6. CONCLUSIONS

The PC-MHR is an effective system to maximize the level of destruction of excess weapons grade plutonium without recycle. The plant produces nuclear electricity at high levels of efficiency while providing a unique level of nuclear safety. The PC-MHR fuel cycle has high resistance to diversion and proliferation, making the system particularly well suited for international deployment for plutonium disposition.
ADVANTAGES AND LIMITATIONS OF THORIUM FUELLED ENERGY AMPLIFIERS

J. MAGILL, C. O’CARROLL, P. GERONTOPOULOS, K. RICHTER, J. VAN GEEL
European Commission, Joint Research Centre, Institute for Transuranium Elements, Karlsruhe, Germany

Abstract

The recently proposed energy amplifier reactor concept, based on the use of an accelerator driven sub-critical assembly with thorium as the breeding fuel, offers potentially significant advantages over conventional reactors in the nuclear fuel cycle. The main advantages stated are, in addition to providing a reactor which is essentially sub-critical, that much less transuranic actinide waste is generated and the risk of nuclear proliferation is substantially reduced. In this paper, we analyse the proliferation and radiotoxicity problems associated with the various energy amplifiers using $^{232}$Th and $^{238}$U as breeding fuels. It is shown that there are major advantages to be obtained in radiotoxicity using $^{232}$Th especially if the bred $^{233}$U and additional uranium isotopes are recycled. In addition, the fuel can be rendered proliferation resistant through mixing with the isotopic denaturant $^{238}$U. This is in contrast to uranium fuelled reactors where there is no natural denaturant for plutonium isotopes.

It is also shown, however, that through the use of this isotopic denaturant for proliferation resistance of thorium fuel, a new source of radiotoxicity arises. The proliferation and radiotoxicity problems are now coupled together and this leads to limitation in the overall advantages to be gained. This is the proliferation / toxicity dilemma of a thorium cycle. Nevertheless, depending on which aspects have priority i.e. proliferation resistance, radiotoxicity of the spent fuel, or combined proliferation - radiotoxicity, a thorium fuelled energy amplifier can have clear advantages over conventional reactors in the nuclear fuel cycle.

1. INTRODUCTION

In the Energy Amplifier (EA) concept [1,2] the input energy to the amplifier is provided by high energy charged particles produced by a particle beam accelerator. The charged particles, typically protons of energy 1GeV, collide with a heavy target (Pb, U, or Th) and each proton produces approximately 50 spallation neutrons of average energy 20MeV. The amplifier consists of sub-critical assembly of $^{232}$Th/$^{233}$U with, for example, $k_{eff} = 0.95$ such that a further neutron multiplication of $1/(1-k_{eff}) = 20$ can be expected. Depending on the moderator/coolant used, these neutrons can be completely thermalised (graphite moderator), partially thermalised (pressurised water moderator) or remain fast (lead coolant) and result in thermal, pressurised water or fast energy amplifiers denoted by T-EA, PW-EA, and F-EA respectively.

In contrast to conventional critical reactors (where for thermal reactors typically 3% fissile material is required in the fresh fuel to give $k_{eff} = 1$), steady state operation of the T-EA and PW-EA results in breeding equilibrium (with typically 1% fissile material) with each fissioned nucleus by a fuel nucleus bred from the fertile $^{232}$Th. Refuelling is required only to overcome fission product poisoning and / or to avoid material damage. This is interesting with respect to the F-EA where, because of the much smaller fission product poisoning, very high
burnups can, in principle, be achieved. The fuel fabrication step may also be avoided by placing the natural $^{232}\text{Th}$ in a blanket around the core until a sufficient concentration of fissile $^{233}\text{U}$ has been bred.

Another interesting aspect of the EA is that the driving neutrons are independent of the sub-critical assembly. From the neutron kinetics point of view, these neutrons can be considered as delayed in that they do not appear immediately from the fission process (hence they do not appear in the exponential term for neutron multiplication [3]). In a critical reactor it is the 0.7% (for $^{235}\text{U}$) delayed neutrons which make control of the reactor possible by reducing $k_{\text{eff}}$ for prompt neutron multiplication to 0.993. Obviously reactivity insertions which give a $\delta k_{\text{eff}} \geq 0.007$ will lead to rapid neutron multiplication. In an EA with $k_{\text{eff}} = 0.95$, a $\delta k_{\text{eff}} \geq 0.5$ is required to produce the same effect. Conversely the EA can tolerate much greater reactivity excursions than critical reactors. For this reason the possibility of a reactivity induced accident is much reduced. As a consequence, this may lead to less demanding safety requirements, fewer studies of hypothetical accident scenarios, and/or faster licencing procedures.

There are, however, two additional problems associated with conventional nuclear fuel cycles i.e. proliferation and waste disposal. In the following sections we consider these aspects in detail.

2. RADIOTOXICITY OF THE SPENT FUEL

The radiotoxicity of a nuclide is determined by its the Annual Limit of Intake or ALI value. These ALI values incorporate dose limits, radiation weighting factors, tissue weighting factors, metabolic and biokinetic information [4]. The ALI values for ingestion and inhalation for radionuclides have been evaluated [4]. The radiotoxicity of a nuclide is given by

$$\text{Radiotoxicity} = (0.02\text{Sv}) \frac{A}{\text{ALI}} \tag{1}$$

where $A$ is the activity of the nuclide expressed in Bq. The factor 0.02Sv is the yearly limit of occupational exposure with a maximum permissible exposure of 0.1Sv over a five year period. In the following, results are given relative to the ALI values for ingestion (results for inhalation are greater numerically but follow the same trends). This study focuses on the potential radiotoxicity of spent fuel without considering any specific accident scenarios or treatment of the mobility and migration of nuclides in the geo- and biospheres. For example, Np is produced in small amounts, however, it is soluble in water making it more mobile and potentially more dangerous than other actinides. Note that, in the thorium fuel cycle, Np is produced in amounts one order of magnitude less than in the uranium cycle.

The actinide inventory in the thorium spent fuel from the T-EA, PW-EA and F-EA's have been taken from Ref [2]. The inventory for the uranium spent fuel has been calculated using RADONN [5]. In fig. 1, the total actinide radiotoxicity resulting from the uranium fuel in a T-EA with a burnup of 40GWD/t at discharge is given. The decay evaluations and radiotoxicity calculations were made using RADONN [5] developed at ITU.

The fission products pose a problem in the short term as they dominate the radiotoxicity for the first few hundred years. There are also potentially dangerous long lived fission products such as $^{99}\text{Tc}$ and $^{129}\text{I}$. There is a difference in the inventory of fission products produced by $^{233}\text{U}$ and $^{235}\text{U}$ fission due to the slight mass difference. However, the total radiotoxicities hardly differ and are not displayed.
In the figures 1-9, reference is made specifically to T-EA, PW-EA and F-EA reactors. The radiotoxicity results, however, also apply to conventional critical reactors with similar neutron spectra i.e. thermal (graphite moderator), partially thermalised (pressurised water), and fast reactors. The radiotoxicities of the actinides and their daughter products are grouped together according to their chemical origin. Hence in fig.1 the label "Pu" implies the four plutonium isotopes $^{239}$Pu, $^{240}$Pu, $^{242}$Pu, $^{243}$Pu (which are present at discharge) and all their daughter products. This representation has the advantage that one can see clearly the effects of recycling particular isotopes from the waste. At a cooling time of 100y, the "Pu" contribution to the radiotoxicity is dominated by $^{241}$Am (433y) which is the daughter of $^{241}$Pu (14.4y). Final decay products of $^{241}$Pu are $^{237}$Np (2x10$^6$y) and $^{229}$Th (7.7x10$^3$y). At 10$^4$y the "Pu" is dominated by $^{240}$Pu (6.6x10$^3$y) and daughter $^{236}$U (2.3x10$^7$y). At 10$^5$y the main contribution comes from parent $^{239}$Pu (2.4x10$^4$y) with daughter $^{235}$U (7x10$^8$y) and $^{242}$Pu (3.7x10$^5$y).

Initially the "Cm" curve is dominated by $^{244}$Cm (18.1y) with daughters $^{240}$Pu (6.6x10$^3$y) and $^{236}$U (2.3x10$^7$y). $^{245}$Cm (8.5x10$^3$y) decays through $^{241}$Pu (14.4y) and $^{241}$Am (433y) to $^{237}$Np (2.1x10$^6$y).

At discharge the "Am" consists primarily of $^{243}$Am (7.4x10$^3$y, decay products $^{239}$Np (2.4d), $^{239}$Pu (2.4x10$^4$y), and $^{235}$U (7x10$^8$y)) and a small amount of $^{244}$Am (10h, decay products $^{244}$Cm (18.1y), $^{240}$Pu (6.6x10$^3$y), and $^{236}$U (2.3x10$^7$y)).

Although the "U" consists initially mainly of $^{238}$U (4.5x10$^9$y), there is a small quantity of $^{239}$U present. This $^{239}$U decays almost immediately to $^{239}$Np (2.4d) and then to $^{239}$Pu (2.4x10$^4$y) and gives a radiotoxicity contribution which is approximately twice that of $^{238}$U. Beyond 10$^5$y the toxicity arises essentially from $^{210}$Po a daughter of $^{210}$U. Hence the radiotoxicity of natural $^{238}$U is approximately one third of the value given by "U" over 10$^4$y.

In fig. 2, 3, and 4, the ingestion radiotoxicities of spent thorium fuel with a burnup of 40GWd/t from T-EA, PW-EA, and F-EA's are shown. In the PW-EA the light water moderator results in a thermal neutron spectrum with considerable epithermal and fast neutron components. In the F-EA, use of a lead moderator/coolant results in a primarily fast spectrum. Hence in the PW-EA and F-EA's the higher energy neutron (n,2n) reactions with the $^{232}$Th are possible and give rise to $^{232}$U and $^{231}$Pa. These isotopes play a key role in a thorium fuel cycle.

In these figures, the label "U" represents the five uranium isotopes $^{232}$U, $^{233}$U, $^{234}$U, $^{235}$U, and $^{236}$U plus all their daughter products. In fig.2, the main contribution comes initially from $^{233}$U (1.6x10$^5$y, with daughter $^{229}$Th (7.3x10$^3$y)). $^{238}$Pu (88y, daughter $^{234}$U (2.5x10$^5$y)), and $^{232}$U (70y, daughters $^{228}$Th (19.1y), through to $^{208}$Pb). In figs. 3 and 4, both $^{232}$U and $^{233}$U dominate. At 10$^5$y the toxicity is dominated by $^{229}$Th a daughter of $^{233}$U.

In both the T-EA and PW-EA's the main contribution to "Pu" is from $^{238}$Pu and at later times $^{239}$Pu. In the F-EA there is no plutonium contribution. In the T-EA (fig.2) the "Pa" radiotoxicity is determined by $^{233}$Pa (27d, daughter $^{233}$U (1.6x10$^5$y)) and the "Pa" and "U" toxicities show similar behaviour. In both the PW-EA and F-EA, the isotope $^{231}$Pa (3.3x10$^4$y), produced by higher energy neutrons in (n,2n) reactions with $^{232}$Th, contributes to "Pa".

The "Th" radiotoxicity is determined by $^{232}$Th (7.3x10$^3$y) and daughter $^{228}$Ra in all cases. The "Th" curve may be considered a reference level for one tonne of natural thorium. In both the T-EA and F-EA's, $^{237}$Np is produced in small quantities.

In fig. 5 a comparison is made of the ingestion radiotoxicities of spent uranium and thorium fuels from a T-EA: (burnup 40GWd/t). Clearly, in the first 30,000 years the radiotoxicity of the thorium waste is approximately a factor of 30 less than for the uranium waste. From 30,000 to 10$^6$ years the radiotoxicity of the thorium waste is higher than that of uranium by approximately a factor of five. The toxicity of the spent thorium fuel in a PW-EA is higher than from a T-EA initially due to the higher concentration of $^{232}$U from (n, 2n) reactions. The radiotoxicity of thorium from a F-EA is a factor ten higher than that from a T-EA due the fact that in breeding equilibrium the concentration of $^{233}$U is a factor ten higher.
The radiotoxicity of spent thorium fuel from an EA can be reduced further by recycling the uranium isotopes making use of the fact that the $^{233}\text{U}$ can be used to seed thorium for fresh fuel. In fig. 6, the effect of recycling 99% of the uranium is shown. It has also been assumed that after discharge enough time (some months) has elapsed before recycling to allow most of the $^{233}\text{Pa}$ to decay to $^{233}\text{U}$. From the figure it can be seen that over the first 30,000 years the radiotoxicity of the thorium waste is now a factor of 200-700 lower than that of the spent uranium fuel.

In fig. 7 a comparison is made of the radiotoxicities of spent thorium fuel and waste resulting from recycling. If only the uranium is recycled (99% including the $^{233}\text{Pa}$ which has decayed), the radiotoxicity is reduced only by a factor 2-10 over the first 30,000 years. The remaining radiotoxicity is primarily due to the isotope $^{231}\text{Pa}$ which is not separated. Only when this $^{231}\text{Pa}$ is recycled in addition can major reductions in radiotoxicity (100 - 1000) result.

In fig. 8, the effect of recycling spent thorium fuel from a F-EA is shown. Recycling 99% of the uranium leads to a reduction in radiotoxicity of 5-500 over the first 30,000 years. If in addition the $^{231}\text{Pa}$ is recycled the reduction can be increased to 60-1000.

In fig. 9 the effect of burnup on the radiotoxicity is given for spent thorium fuel in a F-EA. It can be seen that after $10^3$ years the radiotoxicity (of actinides and daughters) per unit mass is independent of burnup. The differences at $10^2$ years are accounted for by the isotope $^{232}\text{U}$, the concentration of which increases with burnup. The radiotoxicity per unit mass can also be divided by the burnup to give the radiotoxicity per unit of thermal energy obtained by fission and this is also shown in fig. 9. Hence, taking the fuel to extended burnup does not lead to a dramatic decrease in the radiotoxicity per unit energy produced.

### 3. PROLIFERATION ASPECTS OF A THORIUM FUEL CYCLE

From the previous section it has been shown that considerable advantages regarding radiotoxicity of the spent fuel can be obtained through the use of thorium ($^{232}\text{Th}$) rather than natural uranium as a breeder fuel in the energy amplifier. Even without recycling, the waste from the thorium fuel in a T-EA is approximately a factor 30 less toxic than the waste from uranium over the first 30,000 years cooling time. By recycling the uranium isotopes in the T-EA, and in addition the $^{231}\text{Pa}$ in the PW-EA and F-EA, this factor can be increased by another order of magnitude. There is, however, a problem here since the recycled uranium from the spent thorium fuel could be diverted and used to construct a nuclear explosive. The highly fissile $^{233}\text{U}$ produced by irradiation of $^{232}\text{Th}$ is present in the fuel together with other uranium isotopes. The reactor grade uranium does not have the neutron reproduction characteristics of pure $^{233}\text{U}$, nevertheless, criticality can be achieved.

In contrast to reactor grade plutonium associated with the uranium cycle, where sophisticated engineering is required for the implosive assembly [6,7], the reactor grade uranium from the thorium cycle could be assembled by a relatively simple gun type device. Because of the high neutron background from the plutonium isotopes, a gun type assembly of such material will result in predetonation. An estimate of the neutron background threshold beyond which an implosive assembly is required is $10^4$ neutrons/kg/s as shown in fig. 10 (in the Trinity device [8], the $^{240}\text{Pu}$ content was smaller than the six percent value which currently defines weapons grade Pu. Here we take a reference value of one percent to fix a value beyond which an implosive assembly is required - this results in $1.2 \times 10^4$ neutrons/kg/s). Also shown in fig. 10 are the neutron background rates for the recycled uranium isotopes in spent thorium fuel from the T-EA, PW-EA, and F-EAs at different burnups. Clearly, the reactor grade uranium has a low neutron background allowing critical assembly with a gun type device. (An alternative proliferation pathway [9] is to chemically separate the $^{233}\text{Pa}$ from the waste. Since
this decays to $^{233}$U with a half life of 27 days potentially pure, highly fissile, material can be obtained. It would, however, require an entire charge of a T-EA i.e. 5 tonnes to obtain enough $^{233}$U for one critical mass (approximately 5 kg [10]).

A potential deterrent to diversion is, however, the gamma activity of the material. The major source of this gamma activity is $^{208}$Tl, a daughter product of $^{232}$U, which emits highly penetrating 2.6MeV gamma photons. The quantity of $^{232}$U in the uranium recycled from the spent thorium fuel from a T-EA, PW-EA, and F-EA as a function of burnup is shown in table 1. Even at burnups of 40GWd/t, the PW-EA has a high concentration of $^{232}$U. With a F-EA one has to reach burnups in excess of 160GWd/t to attain similar levels. As to be expected from the T-EA, the concentrations of $^{232}$U are much less than for the PW-EA.

<table>
<thead>
<tr>
<th>Burnup</th>
<th>T-EA</th>
<th>PW-EA</th>
<th>F-EA</th>
</tr>
</thead>
<tbody>
<tr>
<td>40GWd/t</td>
<td>200ppm</td>
<td>3100ppm</td>
<td>500ppm</td>
</tr>
<tr>
<td>80GWd/t</td>
<td>200ppm</td>
<td>5000ppm</td>
<td>900ppm</td>
</tr>
<tr>
<td>160GWd/t</td>
<td>-</td>
<td>-</td>
<td>2200ppm</td>
</tr>
</tbody>
</table>

The dose rate from 5kg (approximate critical mass) $^{233}$U containing 100, 1000, and 10,000ppm $^{233}$U at various times after chemical separation is shown in fig.11. Also shown is the LD-50 which gives a 50% chance of death following one hour exposure. Clearly the material is strongly gamma active, but only in the case of high concentrations of $^{232}$U of 10,000ppm does the activity lead to significant barrier to diversion. It should be noted that the concentration of $^{232}$U can be increased in the PW-EA and the F-EA by recycling the isotope $^{231}$Pa [11]. The presence of $^{232}$U in the material will, of course, complicate the uranium recycling procedure. In table 2 we give the lead thicknesses necessary to reduce the gamma dose rate at 1m to 2.5μSv/hr from 1kg $^{233}$U at various times after chemical purification.

Table 2. Lead shield thickness (cm) necessary to reduce the gamma dose rate at 1m distance to 2.5μSv/hr from 1kg $^{233}$U with various concentrations of $^{232}$U following chemical purification.

<table>
<thead>
<tr>
<th>$^{232}$U/$^{233}$U</th>
<th>10</th>
<th>30</th>
<th>100</th>
<th>400</th>
<th>4000</th>
</tr>
</thead>
<tbody>
<tr>
<td>100ppm</td>
<td>6.75</td>
<td>10.4</td>
<td>13.3</td>
<td>16.4</td>
<td>18.4</td>
</tr>
<tr>
<td>1000ppm</td>
<td>12.1</td>
<td>15.5</td>
<td>18.4</td>
<td>21.5</td>
<td>23.5</td>
</tr>
<tr>
<td>10000ppm</td>
<td>17.2</td>
<td>20.8</td>
<td>23.5</td>
<td>26.0</td>
<td>28.1</td>
</tr>
</tbody>
</table>

It should be clear that if recycling is to be part of the thorium fuel cycle then there is a potential proliferation problem associated with the recycled reactor grade uranium. In contrast to reactor grade plutonium: from the uranium cycle, the reactor grade uranium can be isotopically denatured with the isotope $^{238}$U. In table 3, the critical masses of mixtures of $^{233}$U and $^{238}$U are given. The values here are relevant to the F-EA since the reactor uranium consists almost entirely of $^{233}$U. In the calculations, nuclear data for $^{239}$Pu have been used to simulate the $^{233}$U. To estimate the critical mass of the mixture the one group relations given in refs. [6,7,10] have been used. Clearly a mixing ratio of approximately 9 (parts $^{238}$U) to 1 (part $^{233}$U) will require a total mass of the explosive device of approximately one tonne (between 940kg for the bare sphere and 1200kg for the core plus reflector) - a value which is completely impractical. On the other hand, from table 3, a 1:1 mixing ratio (which implies 10% $^{238}$U in the
fuel) has a critical mass of only 14kg with a reflector. A fifteen tonne F-EA fuel inventory [2], which contains 1500kg reactor grade uranium (total power = 2GWth), is sufficient for 83 (without reflector) to 300 (with reflector) critical masses. Clearly, to safeguard this uranium, a mixing ratio between 4:1 and 9:1 is required. With a lower limit of 4:1 there is 40% $^{238}\text{U}$ in the fuel and this will be the dominant factor in the radiotoxicity.

Table 3. Approximate critical masses of isotopically denatured $^{233}\text{U}$ for various mixing ratios. Data for $^{239}\text{Pu}$ have been used to simulate $^{233}\text{U}$.

<table>
<thead>
<tr>
<th>%$^{233}\text{U}(^{238}\text{U},^{233}\text{U})$</th>
<th>$^{233}\text{U}+^{238}\text{U}$ (kg)</th>
<th>$^{233}\text{U}+^{238}\text{U}$ (kg)</th>
<th>Total mass (kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10% (9:1)</td>
<td>940</td>
<td>230</td>
<td>1200</td>
</tr>
<tr>
<td>20% (4:1)</td>
<td>240</td>
<td>61</td>
<td>560</td>
</tr>
<tr>
<td>25% (3:1)</td>
<td>180</td>
<td>46</td>
<td>480</td>
</tr>
<tr>
<td>33% (2:1)</td>
<td>110</td>
<td>28</td>
<td>390</td>
</tr>
<tr>
<td>50% (1:1)</td>
<td>54</td>
<td>14</td>
<td>300</td>
</tr>
<tr>
<td>100%</td>
<td>18</td>
<td>5</td>
<td>210</td>
</tr>
</tbody>
</table>

* critical mass with/without a 10cm uranium reflector

In contrast to the F-EA, reactor grade uranium from a T-EA or a PW-EA contains significant quantities of the isotope $^{234}\text{U}$. Since the fission properties of $^{234}\text{U}$ are similar to $^{238}\text{U}$, this acts also as an isotopic denaturant. From table 3 it can be seen that a 1:1 mixture of $^{233}\text{U}$ and $^{238}\text{U}$ has a critical mass similar to that of $^{235}\text{U}$. Hence the nuclear data for $^{235}\text{U}$ can be used to simulate the reactor grade uranium from a T-EA or a PW-EA.

In table 4 the critical masses of mixtures of reactor grade uranium and $^{238}\text{U}$ are given. In these calculations nuclear data for $^{235}\text{U}$ have been used to simulate the reactor grade uranium. Here a mixing ratio of approximately 4 (parts $^{238}\text{U}$) to 1 (part reactor uranium) will result in a total mass of the explosive device of approximately one tonne (990kg without reflector, 1200kg with reflector), again a value which is impractical. With a mixing ratio of 1:1, which implies approximately 2% $^{238}\text{U}$ (an amount equal to that of $^{233}\text{U}+^{234}\text{U}$), 48kg reactor grade uranium is required for a critical mass (with reflector). A small five tonne T-EA or PW-EA fuel inventory (total power = 250MWth) contains approximately 1.5-2% or 75-100kg of reactor uranium. Hence, the entire core is sufficient for about five critical masses and diversion of such material should thus be easily detected. This 1:1 mixing ratio, however, must be regarded as a lower limit for proliferation resistance. From the radiotoxicity, the 2% $^{238}\text{U}$ used to denature the reactor uranium will result in a radiotoxicity a factor 50 less than that resulting from the use of $^{238}\text{U}$ as breeder fuel. It should be noted that even after accounting for the different power ratings of the fuels (for F-EA 160MWt, for T-EA 50MWt) approximately five times as many critical masses can be obtained from non-denatured F-EA fuel than from T-EA fuel for the same power produced.

In summary, the proliferation problem associated with the reactor grade uranium can be overcome by denaturing with $^{238}\text{U}$. However, by introducing the denaturant $^{238}\text{U}$ into the fuel, we limit the radiotoxicity advantages associated with the recycled thorium fuel. *This is the proliferation / toxicity dilemma of the thorium based fuel cycle.* Denaturing the reactor grade uranium with an equal quantity of $^{238}\text{U}$ (this leads to a $^{238}\text{U}$ concentration of ≈ 2%) should be regarded as a lower limit for non-proliferation. The resulting radiotoxicity is a factor 50 lower than that obtained by using $^{238}\text{U}$ as the breeder fuel in a T-EA or a PW-EA. (Notice here that since $^{232}\text{Th}$ "burns" at a faster rate $^{238}\text{U}$ ($\sigma_f^{232}\text{Th} = 7.4b, \sigma_f^{238}\text{U} = 2.7b$), a burnup of 40GWd/t for $^{232}\text{Th}$ fuel corresponds to a burnup of 15GWd/t for the $^{238}\text{U}$. The toxicity of the
Table 4. Approximate critical masses of isotopically denatured reactor\(U\) for various mixing ratios. Data for \(^{235}\)U have been used to simulate reactor\(U\).

<table>
<thead>
<tr>
<th>%react(U) ((^{238})U:react(U))</th>
<th>reactor(U+^{238})U (kg)</th>
<th>reactor(U+^{238})U (kg)</th>
<th>Total mass (kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>20% (4:1)</td>
<td>990</td>
<td>250</td>
<td>1200</td>
</tr>
<tr>
<td>25% (3:1)</td>
<td>650</td>
<td>160</td>
<td>930</td>
</tr>
<tr>
<td>33% (2:1)</td>
<td>400</td>
<td>100</td>
<td>710</td>
</tr>
<tr>
<td>50% (1:1)</td>
<td>190</td>
<td>48</td>
<td>500</td>
</tr>
<tr>
<td>100%</td>
<td>62</td>
<td>16</td>
<td>320</td>
</tr>
</tbody>
</table>

* critical mass with/without a 10cm uranium reflector

\(^{238}\)U waste is dominated by \(^{240}\)Pu and \(^{239}\)Pu and the concentrations of these isotopes at 15GWd/t is approximately 75\% of the values at 40GWd/t - referred to the \(^{238}\)U burnup.

With the F-EA the situation is more extreme. To isotopically denature one requires between 4 and 9 times as much \(^{238}\)U to denature and this will result in a radiotoxicity approximately 40-90\% of that obtained by using \(^{238}\)U as the breeder fuel. Clearly, isotopically denaturing the thorium breeder fuel in the F-EA does not lead to any significant advantages in radiotoxicity.

In addition, isotopic denaturing leads to an increase in the absolute amount of \(^{233}\)U required and hence to the gamma activity of the material required to construct an explosive device. It can be seen from table 4 that, with a mixing ratio of 4:1, the mass of fissile material (reactor uranium plus denaturant \(^{238}\)U) for a weapon is approximately 250kg. Approximately one fifth of this is \(^{233}\)U i.e. 50kg. Hence the dose rate from the \(^{232}\)U will be ten times higher than those given in fig. 11. In the case of a PW-EA recycled uranium, one hour exposure to this material will lead to a lethal dose 100 days after chemical separation. The T-EA recycled uranium will be a factor ten less gamma active.

4. CONCLUSIONS

A comparison of fuels in a thermal energy amplifier at a burnup of 40GWd/t shows that spent thorium fuel is considerably less radiotoxic than spent uranium fuel. In the first 1000y, the toxicity reduction is approximately a factor 50, and from \(10^{3}-10^{4}\)y the reduction is a factor 10. Use of thorium breeder fuel in both the PW-EA and F-EA is less favourable with regard to radiotoxicity. In the case of the F-EA the radiotoxicity of the spent fuel after 40GWd/t is a factor ten higher than that from the T-EA due to the much higher levels of fissile \(^{233}\)U required. This can be partly compensated by going to higher burnups (since fission product poisoning is less severe). However even with a burnup of 160GWd/t one can only recover a factor four in radiotoxicity (fig. 9). We conclude that the use of thorium breeder fuel in combination with a T-EA will lead to a potentially significant decrease in radiotoxicity even in a once through, non-recycling scenario.

The radiotoxicity can be further decreased by recycling the uranium isotopes. These uranium isotopes can then be use to seed thorium and produce fresh fuel. From the T-EA, the recycled waste has a radiotoxicity at least a factor 100 less than the (non-recycled) uranium waste over the first 10,000y. It (thereafter it is about one order of magnitude less toxic). With the PW-EA and F-EA, recycling the uranium isotopes does not lead to as significant reduction.
in the radiotoxicity as in the case of the T-EA. One must, in addition, recycle the isotope $^{231}$Pa before major reductions in radiotoxicity can be achieved.

The main disadvantage associated with recycling the uranium isotopes is that this reactor grade uranium is fissile and constitutes a proliferation problem. In contrast to reactor grade plutonium associated with the uranium cycle, where sophisticated engineering is required for the implosive assembly, the reactor grade uranium from the thorium cycle could be assembled in a relatively simple gun type device. Because of the high neutron background from the plutonium isotopes, a gun type assembly of such material will result in predetonation.

A possible solution to safeguard the reactor grade uranium is to denature with a small amount of $^{238}$U. A mixing ratio of $^{238}$U: reactor U of approximately 4:1 for a T-EA and PW-EA, and 9:1 for a F-EA, would result in a total mass of the order of one tonne (fissile material plus reflector) and ensure the impracticability of such material for construction of a nuclear explosive. In addition, if the material resulted from a PW-EA, one hour exposure would result in a lethal radiation dose. Clearly, however, the use of these level of denaturant will contribute a major source of radiotoxicity. A mixing ratio of 4:1 results in $\approx 5\%$ $^{238}$U in the thorium fuel will give a radiotoxicity reduction of only a factor 20 over that of the value of a uranium fuel in a T-EA. In the F-EA, there is radiotoxicity reduction is less. Hence, although the proliferation resistance of the recycled uranium is essentially guaranteed, a new source of radiotoxicity has been introduced. This we call the proliferation / toxicity dilemma of the thorium based fuel cycle.

It must be emphasised, however, that in contrast to Pu isotopes for which there is no natural isotopic denaturant, $^{238}$U can be used to denature the reactor uranium produced with thorium breeder fuel. If proliferation is the major issue, then an essentially proliferation resistant can be designed. If waste disposal is the main issue, orders of magnitude reductions in radiotoxicity can be obtained. But through the proliferation / toxicity dilemma, it has been shown that we cannot have both proliferation resistance and negligible toxicity simultaneously. For the combined proliferation and toxicity, the best that can be done is to denature the reactor uranium in a 1:1 mixing ration with $^{238}$U (thereby ensuring relative proliferation resistance) and reducing the radiotoxicity by a factor $\approx 50$ (which arises from the $\approx 2\%$ denaturant used).

Also, depending on which energy amplifier is used, different levels of the isotope $^{232}$U will be produced. Through its $^{208}$Tl daughter, which is strongly gamma active, one will require shielding in any recycling / fabrication stages. However this gamma activity will also allow one to monitor movements of the material and possible diversion.

We conclude that a thorium based fuel cycle in combination with the family of energy amplifiers does offer a variety of options for improvements in the nuclear fuel cycle with regard to proliferation and/or radiotoxicity. Further studies are required to clarify the potential advantages of accelerator driven sub-critical systems over critical systems from the points of view of safety, optimisation of power production, and transmutation of particularly hazardous short and long lived fission products.
Figure 1. Ingestion radiotoxicity of spent uranium fuel (burnup 40GWd/t) from a Thermal Energy Amplifier (T-EA). Actinides and their daughters are grouped (" ") according to the chemical nature at discharge.

Figure 2. Ingestion radiotoxicity of spent thorium fuel (burnup 40GWd/t) from a Thermal Energy Amplifier (T-EA). Actinides and their daughters are grouped (" ") according to their chemical nature at discharge.
Figure 3. Ingestion radiotoxicity of spent thorium fuel (burnup 40GWD/t) from a Pressurised Water Energy Amplifier (PW-EA). Actinides and their daughters are grouped (" ") according to the chemical nature at discharge.

Figure 4. Ingestion radiotoxicity of spent thorium fuel (burnup 40GWD/t) from a Fast Energy Amplifier (F-EA). Actinides and their daughters are grouped (" ") according to their chemical nature at discharge.
Figure 5. Comparison of the ingestion radiotoxocities of spent uranium and thorium fuels (burnup 40GWd/t) from a T-EA. The radiotoxocities of spent thorium fuel from a PW-EA and F-EA are also shown.

Figure 6. Comparison of the ingestion radiotoxocities of spent uranium and thorium fuels and thorium waste from a T-EA (burnup 40GWd/t). The thorium waste results from the removal of 99% uranium and protactinium from the spent fuel.
Figure 7. Comparison of the ingestion radiotoxicities of spent thorium fuel and waste from a PW-EA (burnup 40GWd/t). The thorium waste results from the removal of 99% uranium and protactinium from the spent fuel. The radiotoxicity of spent T-EA uranium fuel is shown for reference.

Figure 8. Comparison of the ingestion radiotoxicities of spent thorium fuel and waste from a F-EA (burnup 40GWd/t). The thorium waste results from the removal of 99% uranium and protactinium from the spent fuel. The radiotoxicity of spent T-EA uranium fuel is shown for reference.
Figure 9. Ingestion radiotoxicity of spent thorium fuel from a F-EA at burnups of 40, 80 and 160GWd/t.

Figure 10. Neutron emission rates for spent thorium fuel from the T-EA, PW-EA, and F-EA at different burnups. The reference value for an implosive assembly is given by a 1% mixture of $^{240}\text{Pu}$ in $^{239}\text{Pu}$. 
Figure 11. Dose rate as a function of ageing time from 5kg of recycled $^{233}U$ contaminated with $^{232}U$.

REFERENCES


MIXED PLUTONIUM-THORIUM FUEL USE IN FAST BREEDING REACTORS - ECOLOGICAL WAY OF THE ACCUMULATED PLUTONIUM BURNING

E.Ya. SMETANIN, V.B. PAVLOVICH, G.N. KAZANTSEV, I.Ya. OVCHINNIKOV
Institute of Physics and Power Engineering, Obninsk

B.Ya. ZILBERMAN, L.V. SYTNIK
Radium Institute, St. Petersburg

Russian Federation

Abstract

The variant of Pu-Th fuel is considered for burning plutonium accumulated in the fast reactor. The possibility of its extraction reprocessing followed by use of fuel components in reactors is addressed.

A wide range of fuel compositions is used at different nuclear reactors. At present the composition of uranium dioxide with the required enrichment is used mostly. Compositions with metal uranium, uranium alloys are known. Tests with uranium carbide and uranium nitride fuel have been performed.

It is a natural question about Thorium application in nuclear power, and certainly, it may be used in the mixture with plutonium oxide. Such application solves the problem of civil and military plutonium disposition with simultaneous production of the Uranium-233 isotope. Due to long irradiation, for example in a fast reactor core, the isotope being formed will contain near 100 ppm of U-232 isotope, that excludes even an idea to use it as a military material.

Plutonium-thorium oxide fuel can be manufactured by the technology in many aspects similar to the MOX-fuel manufacturing one. In the case of this composition homogeneous precipitation of the oxides' mixture from aqueous solutions with following calcination, pellets' pressing and annealing allow to obtain fuel elements of sufficient strength, radiation resistance and heat conductivity.
One of the main questions of such a composition application in nuclear power is a possibility to reprocess it after burning in the reactor.

It is necessary to analyze carefully possibilities of such a fuel chemical dissolving with possible variants of the preparatory operations.

Does the effect of formation of plutonium oxides solid solutions in thorium oxide take place (as in the case with MOX-fuel)? How does direct dissolving occur in the nitric acid? Do hydrofluoric acid additions give considerable acceleration? Do they provide complete fuel breakdown? Is there an effect of the catalyst use? In any case problems of such a fuel breakdown can be solved.

In our opinion it is necessary to pay attention to the development of fuel compositions of PuO$_2$ + Th kind, where PuO$_2$ particles with shielding facing, for example Nb, are distributed uniformly in the metal thorium. It allows to lower the working temperatures considerably and to simplify the spent fuel elements reprocessing.

Besides it is proposed to use PuO$_2$ + ZrC fuel compositions with achievement of the required content of PuO$_2$ in the products by means of a porous workpiece from zirconium carbide impregnation by plutonium nitrate, repeated many times. Content of zirconium carbide is up to 60 - 70%; the workpiece should have the given geometry. Processes of drying and calcination should follow the impregnation.

Under such fuel composition application U-233 won't be produced because of thorium absence. The increased heat conductivity and relatively simple technological procedure of the products manufacturing are also advantages of this composition.

The present fuel composition is more simple during the spent fuel elements reprocessing.

Fuel components' separation and their purification from main fission products are complicated tasks, but they can be solved by modern methods. The first task is to extract U-233 by tributyl phosphate solutions of comparatively low concentration (5 - 10 %), under this process plutonium must be stabilized in aqueous solution of 3-valent form.
The following operations, depending on the chosen decision, may provide joint thorium-plutonium extraction by extraction methods with their purification from the fission products. The obtained solution after the components' ratio correction (addition to compensate for an amount of the burnt plutonium) may be sent to the fuel manufacturing operations.

Taking into account thorium nuclear-physical properties (Th-228 isotope appearance) it is also possible to separate these components at one of the stages of the extraction process. In this case plutonium can be directly sent to recycling with a fresh lot of thorium, and the extracted thorium can be stored during several years.

The most reasonable is to recycle the two products with remote manufacturing of fuel elements with biological shield, which provides permissible control radiation levels at working places. While civil plutonium handling it is necessary condition, and thorium addition can only impact on the biological shield thickness. The obtained fuel elements, assemblies must be stored at a factory and plants, and transported in shielded containers. Such an end of the technological process allows to decrease requirements on the fuel components absolute purification from the fission products at least up to the value of plutonium and thorium own activity.

Technical-economical parameters that follow from plutonium-thorium cycle realization:

1. Civil and military plutonium burning is provided, depending on a reactor capacity. In the case of a necessity military plutonium conversion to the civil one may be promoted.

2. Thorium that is widely distributed in nature is involved in the fuel cycle; the technology of its extraction, purification and manufacturing of products from this material is practically developed.

3. Uranium-233 isotope is produced, the most safe thermal neutrons reactor may be created on its basis. In the fast reactor core considerable amounts of
this isotope with the increased content of U-232 are produced, and in the blanket area more pure uranium-233 is produced.

In the technology of fuel elements and assemblies development for the uranium-233 — thorium thermal reactor it should be "fresh"-extracted or "fresh"-purified uranium-233 usage foreseen. In this case the biological shield thickness of the technological equipment may be decreased. Such a fuel storage and transportation should be provided with biological shield of warehouses and containers, remote devices for loading into reactors.

4. Plutonium burning in fast reactors and uranium-233 production will cause reduction of the transuranic elements production — neptunium, americium, curium, and will simplify handling with high radioactive wastes of these reactors spent fuel reprocessing.
SESSION 3: CANDU AND LWR
ANNIHILATION OF PLUTONIUM IN CANDU REACTORS

A.R. DASTUR, D.A. MENELEY
AECL CANDU, Mississauga,

R.A. VERRALL
AECL Research,
Chalk River
Ontario, Canada

Abstract

In this paper we show that high neutron economy and a well-thermalized neutron spectrum provide major advantages in plutonium annihilation with thermal reactors. High neutron economy reduces core fissile requirements and consequently the plutonium level in discharged fuel, eliminating reprocessing and recycling cost. The well-thermalized spectrum reduces Pu240 formation by increasing the fission to capture cross section ratio of Pu239. The CANDU reactor which achieves high neutron economy by using heavy water as moderator/coolant and by eliminating reactivity suppression through on-power refuelling, can, in the absence of U238, annihilate 90% of the initial fissile inventory in a once-through (non-reprocessing) fuel cycle. Recycling the spent fuel (without refabrication) is available as an option to consider eventual storage without reprocessing. The absence of U238 has a limited effect on reactor dynamics and can be addressed with the use of current reactor technology. A program has commenced at AECL to select a neutronically inert material to contain the plutonium. Candidates include MgAl₂O₄, ZrO₂, SiC, Si, BeO, and others. For an initial screening process (prior to in-reactor testing), heavy doses of 70 Mev iodine ions are being applied to samples of these materials, as a simulation of in-reactor fission fragment damage. The main objective of this screening process is to find a candidate that does not swell significantly. Alumina has been included in these tests as a reference because it is known that alumina swells significantly due to a transformation to the amorphous phase. Test results to date, and of other tests worldwide, are presented.

1. INTRODUCTION

The end of the cold war has led to the issue of the safe disposition of plutonium from dismantled weapons. Several options are being studied, which include burning plutonium as Mixed Oxide (MOX) fuel in thermal and fast reactors. The main shortcoming of this approach is the production of plutonium from uranium during the irradiation of MOX and, subsequently, the large fissile plutonium content of the discharged fuel. The latter is especially true of reactors which operate with a relatively high fissile inventory due to poor neutron economy.

A more definitive approach is to annihilate the plutonium to negligible levels in the discharged fuel. This option involves burning plutonium in the absence of uranium to prevent plutonium formation. In this approach the level of plutonium in the discharged fuel is more strongly dependent on neutron economy.

In this paper we present the CANDU as a candidate for plutonium annihilation, describe the basis for the CANDU advantage, illustrate the advantage by giving the results of studies and identify areas of development.
2. FISSION REQUIREMENTS

The CANDU reactor with its excellent neutron economy can operate on core fissile plutonium inventories as low as 50 kg (in the absence of uranium) and consequently with thermal neutron flux levels in excess of \(10^{15}\) n/(cm\(^2\)-s). On-power fuel management studies with CANDU fuel consisting of weapons grade plutonium, illustrate that plutonium annihilation rates can be maximized by progressively shifting the fuel into higher neutron flux regions as the fissile content of the fuel depletes. Fissile content drops by 90% in a single pass through a CANDU fuel channel. A second pass of the fuel is needed to reduce the fissile content of the discharged fuel to negligible levels. Complete annihilation of the initial fissile inventory is, therefore, achievable without having to reprocess and refabricate the discharged fuel.

This ability of CANDU is related to the basic design features of using heavy water as moderator and a fuel handling system that provides an on-power refuelling capability. Both design features lead to low neutron absorption rates in the CANDU lattice due to the low neutron absorption cross section of the heavy water and the absence of burnable poisons for reactivity suppression. As a result, CANDU can be operated with a variety of low grade (low fissile content) fuels as these provide sufficient reactivity in a CANDU lattice.

The extent of CANDU's fissile economy is illustrated in Table 1 where the fissile inventory for several reactor concepts is compared for 1 GW(e) cores. The inventory needed for CANDU is between 2 and 4 times lower. This is the case for a reactor that is fuelled with natural uranium.

<table>
<thead>
<tr>
<th></th>
<th>FBR</th>
<th>PWR</th>
<th>CANDU</th>
<th>CANDU AB</th>
</tr>
</thead>
<tbody>
<tr>
<td>Inventory (te)</td>
<td>3 to 4</td>
<td>2 to 3</td>
<td>1</td>
<td>0.05</td>
</tr>
</tbody>
</table>

For plutonium annihilation, the fuel consists of a mixture of the plutonium isotopes in a neutronically inert matrix. (Several materials for the matrix have been proposed). The absence of U238 eliminates the main source of the plutonium isotopes. It also eliminates the main neutron absorber of the lattice resulting in a remarkable improvement in neutron economy. The loss of the neutron absorption in U238, which comprises 30 to 40% of the neutron absorption in the CANDU lattice, reduces the fissile requirement of the CANDU AB (Actinide Burner) to 50 kg for a 1 GW(e) core. (Table 1).

A lower fissile inventory requires a correspondingly higher operating neutron flux level to produce the rated power. The higher operating neutron flux level is the basis for the CANDU superiority in reducing the accumulation of the non-fissile minor actinides. In particular, the on-power refuelling system is used to move the fuel into regions of higher flux as its fissile content depletes during irradiation. Through proper fuel management of this type the last traces of plutonium can be eliminated before the fuel is discharged.

In order to appreciate the extent of the CANDU flux advantage, the thermal neutron flux levels in the fuel for the reactor concepts of Table 1 are compared in Table 2. Note that the flux level in the CANDU AB is comparable to that in some FBRs.

104
Table 2  NEUTRON FLUX LEVEL IN FUEL

<table>
<thead>
<tr>
<th>Reactor Type</th>
<th>Neutron Flux (n/cm².s))</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Thermal (0.625 ev)</td>
</tr>
<tr>
<td>FBR</td>
<td>-</td>
</tr>
<tr>
<td>PWR</td>
<td>8.0E+13</td>
</tr>
<tr>
<td>CANDU</td>
<td>1.4E+14</td>
</tr>
<tr>
<td>CANDU AB</td>
<td>5.0E+15</td>
</tr>
</tbody>
</table>

To understand the annihilation process, the capture and fission cross sections of the plutonium isotopes in thermal and fast energy spectra must be considered, (Table 3). In a fast spectrum, all the plutonium isotopes are fissionable, but the fission cross section is relatively small; about 3 barns for 0.8 mev neutrons. In a thermal spectrum, only the isotopes with odd mass numbers are fissionable, but the fission cross sections are large; several hundred barns.

Table 3  NEUTRON CROSS SECTIONS
(barns)

<table>
<thead>
<tr>
<th>ISOTOPE</th>
<th>THERMAL</th>
<th>FAST</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>(\sigma_c)</td>
<td>(\sigma_f)</td>
</tr>
<tr>
<td>Pu 238</td>
<td>540</td>
<td>18</td>
</tr>
<tr>
<td>Pu 239</td>
<td>269</td>
<td>748</td>
</tr>
<tr>
<td>Pu 240</td>
<td>290</td>
<td></td>
</tr>
<tr>
<td>Pu 241</td>
<td>358</td>
<td>1011</td>
</tr>
<tr>
<td>Pu 242</td>
<td>19</td>
<td></td>
</tr>
<tr>
<td>Pu 243</td>
<td>87</td>
<td>196</td>
</tr>
</tbody>
</table>

The annihilation process in CANDU involves first, neutron capture to change from even to odd and then fission by thermal neutrons of the odd numbered isotopes. The high thermal neutron flux level provides an appreciable annihilation rate in spite of the neutron capture required first. Reaction rates (per nuclide) of the plutonium isotopes in CANDU and FBRs of current design are compared in Table 4. The reaction rates in CANDU are between one and two orders of magnitude higher.
Table 4

COMPARISON OF REACTION RATES

(Relative Units)

<table>
<thead>
<tr>
<th>ISOTOPE</th>
<th>FBR</th>
<th></th>
<th>PWR</th>
<th></th>
<th>CANDU AB</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>capture</td>
<td>fission</td>
<td>capture</td>
<td>fission</td>
<td>capture</td>
</tr>
<tr>
<td>Pu238</td>
<td>1.25E+16</td>
<td>4.32E+16</td>
<td>1.44E+15</td>
<td>2.70E+18</td>
<td>9.00E+16</td>
</tr>
<tr>
<td>Pu239</td>
<td>1.00E+14</td>
<td>1.00E+16</td>
<td>2.15E+16</td>
<td>5.98E+16</td>
<td>1.35E+18</td>
</tr>
<tr>
<td>Pu240</td>
<td>1.00E+16</td>
<td>2.32E+16</td>
<td>8.09E+16</td>
<td>1.79E+18</td>
<td>5.06E+18</td>
</tr>
<tr>
<td>Pu241</td>
<td>1.00E+16</td>
<td>2.86E+16</td>
<td>1.52E+15</td>
<td>9.50E+16</td>
<td></td>
</tr>
<tr>
<td>Pu242</td>
<td>7.50E+15</td>
<td>1.57E+16</td>
<td>4.35E+17</td>
<td>9.80E+17</td>
<td></td>
</tr>
<tr>
<td>Pu243</td>
<td>6.96E+15</td>
<td>1.57E+16</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

3. FUELLING CANDU WITH PLUTONIUM

The absence of U238 has a major impact on the fuel management strategy that is used in the annihilation option. As formation of Pu239 is eliminated, the reactivity of the lattice drops rapidly with fuel burnup. The refuelling rate required to maintain criticality is significantly higher compared with the reactor that burns natural uranium. The refuelling rate is kept within the capability of the Fuel Handling System by adjustment of the initial fissile content of the bundles. An increase in fissile content to reduce the refuelling rate, however, has detrimental effects. It requires reactivity suppression which raises the fissile content of the discharged fuel. It hardens the neutron spectrum and increases Pu240 formation. It reduces the neutron flux level which increases the accumulation of non-fissile actinides.

The fuel management strategy used is, therefore, a balance between the fissile content and the refuelling rate to obtain the optimum benefit from the fuel handling capability that is available. The optimization includes the option of shuffling the fuel between bundle positions and/or between channels.

The lattice reactivity drop with irradiation for an initial Pu239 content of 76 g in each CANDU fuel bundle is shown in Figure 1. The excess reactivity averages to 4.5% over 150 days of irradiation. This is the required value to sustain reactor operation. The rate of fuelling the reactor to achieve this fuel lifetime is within the capability of current fuel handling technology.

Table 5

BUNDEL PLUTONIUM CONTENT

<table>
<thead>
<tr>
<th>ISOTOPE</th>
<th>CONTENT</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu239</td>
<td>68.5</td>
</tr>
<tr>
<td>Pu240</td>
<td>4.23</td>
</tr>
<tr>
<td>Pu241</td>
<td>0.26</td>
</tr>
<tr>
<td>Pu242</td>
<td>0.02</td>
</tr>
</tbody>
</table>
With a bundle plutonium inventory of 73 g, (isotopic mix shown in Table 5), the Pu239 content at discharge from a once-through (no shuffling) cycle is reduced by 87%. The isotopic mix of the discharged bundle is shown in Table 6.

Table 6  
ISOTOPIC MIX OF DISCHARGED BUNDLE

<table>
<thead>
<tr>
<th>Isotope</th>
<th>g</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu239</td>
<td>9.2</td>
</tr>
<tr>
<td>Pu240</td>
<td>15.2</td>
</tr>
<tr>
<td>Pu241</td>
<td>3.3</td>
</tr>
<tr>
<td>Pu242</td>
<td>2.7</td>
</tr>
</tbody>
</table>

Using the once-through (no shuffling) cycle in a 480 channel (950 MW(e)) CANDU, the annual reduction of Pu 239 is 1.5 te. The fuelling rate required for this cycle is six fuel channels per FPD. The bundle fissile content is low enough to refuel the channel completely. It is estimated that the Reactor Regulating System is capable of handling this rate of refuelling.

Figure 1. Lattice Reactivity Depletion
3.1 The Once-Through Cycle

The plutonium remaining in the discharged fuel can be recycled after reprocessing and refabrication into bundles. This is a major cost component of the annihilation process.

The once-through fuel cycle minimizes cost by avoiding reprocessing and refabrication of the fuel. The key requirement for this cycle is to achieve a bundle refuelling rate that maintains the fissile content of the core close to the minimum value of 50 kg. There are two approaches envisaged to achieve this. One involves a redesign of the Fuel Handling System. This is the less desirable of the two approaches as it uses technology that is currently unproven.

In the second approach, the fuel bundle is passed through the reactor once in the outer (low flux) channels that are located towards the core edge and then again through the central channels where the neutron flux is higher. The higher flux compensates for the lower fissile content during the second pass and minimizes the reduction in power density. This type of fuel shuffling is required to annihilate the plutonium remaining in the bundle at the end of the first pass through the channel. This approach exploits the on-power refuelling feature of CANDU. One additional pass (or at most two) is sufficient to reduce the plutonium content of the discharged bundle to a level that is comparable to the losses experienced in fuel reprocessing.

The additional pass would entail reservation of specific fuel channels in the core for the irradiation of second pass bundles and an increase in the fuelling machine duty. It is expected that some modifications will be required to the design of the Fuel Handling System to achieve this capability. However, the design concept of the system will remain unchanged.

4. REACTOR DYNAMICS AND CONTROL

There are two major reactivity feedback effects that are attributed to U238 in CANDU. Coolant voiding from a fuel channel reduces resonance absorption in U238 and increases the fast fission rate. This produces positive reactivity. U238 is the main contributor to the negative feedback from doppler broadening of the cross section resonances on fuel temperature increase. As expected, both effects are reduced in the absence of U238.

<table>
<thead>
<tr>
<th>Table 7  REACTIVITY COEFFICIENTS FOR A CANDU LATTICE</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bundle Plutonium Content</td>
</tr>
<tr>
<td>---------------------------</td>
</tr>
<tr>
<td>76 g</td>
</tr>
<tr>
<td>Fuel Temperature Coefficient (micro-k/Deg C)</td>
</tr>
<tr>
<td>Coolant Void Reactivity (milli-k)</td>
</tr>
</tbody>
</table>
The coolant void reactivity coefficient is near-zero. The fuel temperature reactivity coefficient is negative and near zero, (Table 7). The power coefficient of reactivity is negative or zero throughout the fuel life. As the power coefficient of reactivity is near zero for the currently operating CANDU reactors, there is no implication on the Reactor Regulating System of using this type of fuel.

Table 8

EFFECT OF 100 °C TEMPERATURE RISE ON PLUTONIUM REACTION RATES
(x 1000)

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Change in Neutron Yield</th>
<th>Change in Neutron Absorption</th>
<th>Difference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu239</td>
<td>-0.15</td>
<td>-0.074</td>
<td>-0.076</td>
</tr>
<tr>
<td>Pu240</td>
<td>0.0</td>
<td>0.135</td>
<td>-0.135</td>
</tr>
<tr>
<td>Pu241</td>
<td>-0.029</td>
<td>-0.0012</td>
<td>-0.028</td>
</tr>
<tr>
<td>Total</td>
<td>-0.179</td>
<td>0.060</td>
<td>-0.239</td>
</tr>
</tbody>
</table>

The negative value of the fuel temperature coefficient in the absence of U238 warrants some explanation. The plutonium isotopes have a negative contribution to the lattice fuel temperature coefficient. This behaviour is due to the shift, on fuel temperature rise, in the temperature of the thermal neutron spectrum and the resulting change in the cross sections of Pu239 and of Pu241.

Table 8 shows the behaviour of the reaction rates of plutonium isotopes with fuel temperature rise. Pu239 and Pu241 reaction rates decrease with fuel temperature. But the drop in neutron yield is larger than the drop in neutron absorption.

There is a significant reduction in the delayed neutron fraction relative to the natural uranium CANDU. The value for this lattice is between 3.0 and 3.8 mk compared with 6 mk. However, the neutron generation time for the lattice is significantly longer (2 ms) and the discontinuity between the sub-prompt and super-prompt neutron transient is relatively small.

5. DEVELOPMENT REQUIREMENTS

This study has, as of yet, not uncovered any neutronic issues that would prevent the use a plutonium-based non-fertile fuel in CANDU. It is expected that with current fuel handling technology, the fuelling strategies that are envisaged for the use of this type of fuel can be implemented. This makes this plutonium annihilation strategy feasible with the current CANDU concept.

The main task is to develop a neutronically inert matrix that is compatible with the actinide mix in a reactor environment at power densities that are currently achievable. If the CANDU is to operate at current power densities, the fast neutron flux level, which affects the life of the CANDU plant remains unchanged.
6. FUEL DESIGN

Selection of Fuel Inert Matrix Material or Carrier Material

The objective is to find a suitable inert matrix material or carrier material to hold the Pu. Its function is to dilute the Pu, since pure Pu or PuO₂ would be much too highly fissile, and to not produce new Pu during the irradiation (as UO₂ does). The following sections identify the properties required for such a carrier material, list and assess a selection of materials that appear the most promising, and discuss work begun at CRL and elsewhere on this program.

6.1. Properties Required For a Carrier Material

**Neutronics:** All elements in the carrier material must have low neutron absorption.

**Melting Temperature:** A high melting temperature is required so that fuel operating temperatures are below approximately 0.5 of the melting temperature (Kelvin). This ensures that thermally activated processes, such as fission gas diffusion, are acceptably slow. A high melting temperature also ensures that there is an acceptable safety margin between fuel operating temperature and melting temperature, to accommodate potential off-normal excursions.

**Compatibility with Coolant and Clad:** Chemical compatibility with water at coolant temperatures is a requirement. Compatibility with the clad is also required, although the clad need not necessarily be zirconium or a zirconium alloy.

**Phase Stability:** Phase transformations as the fuel cycles up and down in temperature are unacceptable if there is a volume change associated with the transformation or if the material loses structural integrity. Similarly, a loss of crystallinity, thus forming an amorphous phase, as the fuel is irradiated, is unacceptable if it leads to similar problems. This latter point is one of the aims of the test program outlined below.

**Thermal Conductivity:** High thermal conductivity is advantageous, since this allows low fuel operating temperature (for a given element power rating). This provides increased safety margins in the carrier material. Thermal conductivity at least as high as, and preferably higher, than UO₂ is required.

**Solid Solubility with Pu:** In order to disperse the Pu on an atomic scale in the carrier material, the carrier should be able to dissolve 1-3% Pu. In contrast, MOX fuel with small particles of PuO₂ in a UO₂ matrix have been used successfully in LWR power reactors, but reactor operators have had to be conservative in the burnup limits of the fuel, until the effect of the particulate nature of the fuel under irradiation is established. Note, however, that the UO₂ matrix has ability to dissolve PuO₂; it is only that the fabrication processes used for the production of (U,Pu)O₂ have not been sufficiently advanced to produce a perfect solid solution. Similarly, it may be acceptable to produce a dispersion of fine particles of PuO₂ in the candidate carrier materials considered here.

**Irradiation Properties:** Good irradiation properties would be required, and demonstrated by extensive in-reactor testing.

6.2. Candidates

A list of candidate carrier materials for the Pu includes ZrO₂, BeO, MgAl₂O₄, CeO₂, SiC and ThO₂. The last material listed, ThO₂, is included because no new Pu is produced, even though
its irradiation produces new fissile material, $^{233}\text{U}$. Thus, use of $\text{ThO}_2$ to hold the Pu would achieve the goal of effectively destroying Pu. If, however, a subsidiary goal were to not produce any new fissile material, $\text{ThO}_2$ would not be an acceptable candidate.

Since there is a considerable base of irradiation experience on $\text{ThO}_2$, it would clearly be the simplest of the candidates to qualify for use as a carrier material for Pu. It will not be further discussed in the following.

Other candidates considered were $\text{Al}_2\text{O}_3$ and silicide dispersion fuels. Alumina ($\text{Al}_2\text{O}_3$) was rejected because it has long been known that it becomes amorphous with a volume change of 30% when used as a fuel. Silicide dispersion fuels were rejected because the Al matrix of the dispersion fuels has too low a melting temperature, $600^\circ\text{C}$, for safety considerations.

Alloys of Pu and metals were considered, for example Zr alloyed with Pu. An apparently promising candidate is Si, as it is cheap, abundant, and has good thermal properties. Also, there is irradiation experience with uranium silicides (larger amounts of uranium that required of plutonium for this purpose). This candidate is retained as a representative of metallic alloys and silicides.

### 6.3. Properties of the Candidate Materials

Melting temperatures, thermal conductivities, and heat capacities of the candidate materials are given in Table 9.

#### Table 9

**MELTING TEMPERATURES AND THERMAL CONDUCTIVITIES OF CARRIER MATERIALS**

<table>
<thead>
<tr>
<th>MATERIAL</th>
<th>Melting Temperature ($^\circ\text{C}$)</th>
<th>Thermal Conductivity (W/m·K)</th>
<th>Heat Capacity$^1$ (J/cm$^3$·K)</th>
<th>Heat Capacity$^1$ (J/g·K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\text{ZrO}_2$</td>
<td>2715</td>
<td>1.9</td>
<td>2.3</td>
<td>2.55</td>
</tr>
<tr>
<td>$\text{BeO}$</td>
<td>2530</td>
<td>220.0</td>
<td>20.0</td>
<td>3.07</td>
</tr>
<tr>
<td>$\text{MgAl}_2\text{O}_4$</td>
<td>2135</td>
<td>10.7</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\text{CeO}_2$</td>
<td>2600</td>
<td>10.9</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\text{SiC}$</td>
<td>2700</td>
<td>85.0</td>
<td>2.15</td>
<td>0.67</td>
</tr>
<tr>
<td>$\text{Si}$</td>
<td>1410</td>
<td>108.0</td>
<td>1.65</td>
<td>0.71</td>
</tr>
<tr>
<td>$\text{UO}_2$</td>
<td>2878</td>
<td>8.75</td>
<td>3.21</td>
<td>2.58</td>
</tr>
</tbody>
</table>

Notes:
The heat capacity per unit volume is more relevant, for calculating temperature increases for a given input of heat, than per unit weight because the volume of the fuel element will be constant whatever the carrier material.
**Zirconia** has a high melting temperature and good neutronic properties (absorption cross section of 0.184 B for thermal neutrons) as long as hafnium, a persistent impurity is carefully removed. Commercial grade Zr contains 1-3 % Hf, but nuclear grade Zr is essentially pure (~50 ppm Hf).

Thermal conductivity increases with temperature, in contrast to most materials. However, it is probably too low, being even lower than for UO₂, which has thermal conductivity values of 9 W/m-K at 100°C and 3 W/m-K at 1000°C.

It is insoluble in water, and should be compatible with Zircaloy sheath.

Phase stability is a potential issue. Zirconia undergoes a phase transformations with large associated volume changes at 900-1000°C (monoclinic to tetragonal). This phase transformation causes cracking and would eventually disintegrate the fuel, after repeated temperature cycling through the phase transition temperature. However, additions of yttrium, calcium, magnesium or possibly other materials will stabilize the high temperature cubic phase of zirconia. (This cubic phase is the fluorite structure, the same as UO₂, PuO₂, and ThO₂.) Approximately, 8 at.% of the additive is required for full stabilization, but smaller amounts provide partial stabilization. The fracture toughness that this stabilization provides makes this material an intensely studied material in the general ceramics field. Thus, the database is good. The thermal neutron absorption cross sections of these additives are not large (Y, 1.28 barns; Ca, 0.43 barns; Mg, 0.63 barns), so while providing an extra degree of complexity in fabrication and possibly in irradiation, maintains this material as a good candidate.

Thus, zirconia's poor thermal conductivity is its most serious drawback. The fuel would have to run at lower powers than urania. Other, more acceptable candidates are sought.

The very high thermal conductivity of BeO, combined with a high melting temperature, are strong advantages for this candidate. The conductivity does decrease rapidly with temperature, but is still high at 1000°C and the fuel operating temperature is determined more by its low temperature conductivity (above coolant temperature) than its high temperature value. It has very low neutronic absorption (0.008 barns for thermal neutrons) and is a neutron moderator. It does not undergo phase transitions (hexagonal over its whole temperature).

A potential drawback to BeO is its toxicity. The hazard is highest when using powders, as fabrication would probably require. However, since Pu is toxic already, the extra toxicity of BeO should be acceptable. Unknown is whether the material would form a solid solution with Pu. The toxicity of BeO would make laboratory testing more difficult.

Spinel (MgAl₂O₄) is a candidate suggested by the Europeans. The French, supported by the Transuranium Institute at Karlsruhe, Germany) are actively considering it for an inert matrix (carrier) material for both LWR and Fast Breeder Reactors, and irradiations are underway in the fast reactor PHENIX. An advantage is its perceived resistance to swelling in-reactor. The argument is that for swelling to nucleate, a number of vacancies must congregate together. Furthermore, there must be Mg, Al, and O vacancies in stoichiometric proportions, i.e., in ratios of 1:2:4. Since the probability that this will happen randomly is small, the material is resistant to swelling. However, its thermal conductivity is only marginally higher than that for UO₂. Melting point is sufficiently high, 2135°C. Thus, it remains a promising candidate as long as thermal conductivity is not an overriding concern.

CeO₂ is considered¹ because its crystal structure is identical to UO₂. Since UO₂ has good properties, except for thermal conductivity, and, especially, good irradiation performance, it is

---

¹ suggested by Hj. Matzke, Transuranium Institute, FRG.
thought that this material could have similar good properties. Its thermal conductivity is about 10.9 W/m-K at 100°C, slightly higher than that for UO$_2$, and its melting temperature is very high, 2600°C. Because of its close similarity with UO$_2$, this is a very promising candidate.

Silicon is abundant, cheap, and has good thermal properties. Also, there is irradiation experience with uranium silicides (with larger amounts of uranium than of plutonium required for this purpose). Silicon is generally relatively inert, although it is attacked by dilute alkalis. Most acids do not affect it. Presumably, control of the coolant pH, together with the use of a Zr sheath would neutralize concerns about attack by the coolant.

The use of silicide fuels (U$_3$Si and U$_3$Si$_2$) for CANDU was investigated in the 1970’s by AECL. They were not selected because they softened and swelled above a certain temperature, which depended on the burnup, and there was a reactor control problem, associated with the low heat capacity. These concerns would need to be re-addressed for the purer silicon-plutonium fuel.

Silicon carbide is a material with a considerable industrial base of use, and, thus also, a large data base on properties, especially compared to other candidates considered above. It has a very high thermal conductivity and a high melting temperature. Both elements, Si and C, are low neutron absorbers, and C will also act as a neutron moderator. Although compatible with water at low temperatures, it must be tested for compatibility with water at reactor coolant temperatures, 300°C and above. Fabrication methods have been developed for industrial application, although these may require modification to incorporate Pu. As long as this material is compatible with water at high temperatures, it appears to be an excellent candidate.

6.4. Irradiation Experience

The only program to test carrier materials in-reactor, with a fissile component to introduce fission damage to the carrier material, was a Bettis program [1-3] in the early 1960’s to select a material for a full-core loading of the Shippingport test reactor. They tested Al$_2$O$_3$, ZrO$_2$, ZrO$_2$+CaO, BeO and ZrSiO$_4$, all containing large amounts of UO$_2$ (>20 wt. %). Only ZrO$_2$ behaved satisfactorily for them: Al$_2$O$_3$ became amorphous after only a very short time in reactor; BeO exhibited high swelling (from fission gas bubble formation) and the BeO matrix material "suffered considerable fission fragment damage". The authors of the Bettis reports suggest that it is the cubic crystal structure of ZrO$_2$ which provides its resistance to damage. This was supported, for example, by the results on the candidate material ZrO$_2$+CaO+UO$_2$, which existed as a two-phase material, one phase cubic and one non-cubic. The cubic phase showed resistance to damage, whereas the non-cubic phase did not. Al$_2$O$_3$, BeO, and ZrSiO$_4$ are all non-cubic and did not test well. However, CeO$_2$, MgAl$_2$O$_4$, and SiC, candidates listed above but not tested by Bettis, all form cubic crystal structures. Therefore, there is reason to expect that one or more of these candidates will be acceptable as a carrier material for Pu in CANDU.

The Bettis results show that ZrO$_2$ might make a good carrier material for our purposes. However, it is the least desirable of the suggested candidates from a thermal conductivity consideration. Therefore, a program of developing a carrier material for Pu in CANDU has begun, focusing first on the candidates CeO$_2$, MgAl$_2$O$_4$, and SiC.

French irradiations of selected candidate materials in PHENIX and a thermal reactor, although not containing fissile material, represent a major investment towards finding a carrier material, although their primary purpose is not to burn Pu, but rather to burn selected undesirable actinide wastes, separated from spent LWR fuel.
6.5. AECL Test Program

To select a carrier material from the above candidates, accelerators can be used for initial screening, as described below. This must be followed by a substantial in-reactor irradiation program (with dissolved fissile material, although not necessarily Pu initially). Simultaneously, an out-reactor properties-determination program must be undertaken to establish that there are no other deleterious properties of the candidate material, such as reaction with water or the selected clad material.

Using the Chalk River Laboratories (AECL) Tandem Accelerator, a beam of 70 MeV iodine ions, incident upon the candidate materials, are used to simulate fission fragments. This will provide a first indication regarding their suitability for use as a fuel. The work has only begun. To date, spinel and alumina (single crystals) have been tested, but only at room temperature. In the alumina specimen, swelling was apparent. The spinel sample is currently being analyzed. Also, a furnace has been built, so that specimens can be tested at temperatures more relevant to operating fuel. SiC samples have been obtained for testing.

SUMMARY

Because of their high neutron economy, CANDU reactors can operate with minimal fissile content. This is especially so in the absence of uranium, i.e. when separated plutonium is used as fuel. The low fissile requirement and the on-power refuelling capability of CANDU can be exploited to achieve high annihilation rates and minimal fissile content in discharged fuel. This minimizes reprocessing and refabrication cost.

The absence of U238 reduces the coolant void reactivity which is the major positive feedback reactivity component in existing CANDU reactors.

The well-thermalized spectrum maintains a negative fuel temperature coefficient by minimizing resonance absorption in Pu239.

These features give CANDU reactors a strategic role in plutonium annihilation.

A program to develop a suitable inert material to hold the plutonium is underway at AECL.
PLUTONIUM BURNING VIA THERMAL FISSION IN UNCONVENTIONAL MATRICES

C. LOMBARDI, A. MAZZOLA*
Department of Nuclear Engineering,
Politecnico di Milano,
Milan, Italy

Abstract

The plutonium coming from dismantled warheads and that already stockpiled from commercial fuel reprocessing have raised many proposals for their burning in a safe and economical manner. This paper concerns the utilization of current PWRs partially fed with a non-fertile oxide-type fuel, the rest of the core being still fed with standard U-235 enriched fuel. The unconventional fuel consists of PuO₂ diluted in an inert matrix, which is highly radiation resistant and scarcely neutron absorbent. The matrix may be ZrO₂, Al₂O₃, MgO or their proper combinations. To verify the reliability and validity of the results, two different computer programmes were been utilized: first single pin cell calculations were performed using the WIMS-E program to assess the physical and neutronic features of the new fuel, then the CASMO-3 program was employed to study the fuel assembly in its actual layout. Commercial PWRs operating in a once-through cycle scheme can transmute more than 97% of Pu-239 and more than 71% of total initially loaded civil or weapons-grade plutonium. The discharged plutonium is in any condition definitely proliferation resistant. The high initial reactivity causes a high initial power peak, and a continuously decreasing power generation during the irradiation. The relatively high thermal conductivity of Al₂O₃ compared to UO₂ one and the use of burnable poisons should allow this new fuel to face this latter effect. A less pronounced power peak in UO₂ rods at EOL has to be tackled. Reactivity coefficients are, in absolute value, slightly lower than the UO₂ ones, but the reactor dynamic behaviour should not be different from that of the operating 20 to 30% MOX-fuelled cores. In conclusion, it seems that the once-through cycle burning of plutonium in PWRs via non-fertile matrices is an interesting solution for both weapons-grade and civil plutonium disposal, worth further investigation.

1. INTRODUCTION

In the recent years, the excess plutonium coming from dismantled warheads has promoted in the nuclear community an extensive debate: several solutions for disposing of weapons-grade plutonium have been so far proposed and discussed from the viewpoints of safeguards, proliferation resistance, environmental safety, technological background, economy and time schedule [1-4]. The result of many published studies seems to be that fission options such as Light Water Reactors (LWRs) with mixed-oxide (MOX) fuels, Liquid Metal-Cooled Reactors (LMRs) with metal fuels, High-Temperature Gas-Cooled Reactors (HTGRs) with coated particle fuels, and Fast Breeding Reactors (FBRs) without the fertile blanket are the most efficient and effective ways to dispose of surplus weapons-grade plutonium. In this framework, it may be worth to take stock of the situation about the present utilization of plutonium in commercial power plants and about its overall role in the fuel cycle.

Plutonium has always been viewed as an advantageous material, suitable as a substitute for uranium-235 and therefore many studies have been carried out aiming at improving the conversion ratio in nuclear reactors so as to generate plutonium at high rate for the
following utilization in FBRs. These reactors are designed to obtain a positive breeding gain, thus allowing a better utilization of uranium ore than in LWRs. In fact, the energy extracted from a given uranium quantity results to be amplified by approximately a factor one hundred.

Nowadays, on the other hand, the present situation points to the need of identifying solutions for plutonium burning in a safe and economical manner. At present, the introduction of FBRs has been shifted into the far future because of economic, technical and political reasons. Besides, the considerable decreasing of uranium prices, the lower than expected uranium demand and the higher availability of natural uranium resources render the concerns about fissile material supplies no longer a critical issue. Moreover, large quantities of plutonium recovered from reprocessed UO₂ fuel have been already stockpiled and over the next decade, under already ratified agreements, significant quantities of weapons-grade plutonium are expected to be available (about 100 metric tonnes). All these considerations, together with proliferation concerns, indicate that the plutonium question might be faced in a different way.

Therefore, it seems possible to affirm that an ever growing plutonium production should not be any longer the goal, and hence the already stored quantities, those continuously produced from operating reactors and those obtained from weapons dismantlement have to be burnt at the highest possible rate with the best economic benefit, avoiding, during their elimination, any further plutonium production. This proposal is primarily referable to weapons-grade plutonium, having to meet the more stringent non proliferation and safety requirements, but has to be considered suitable for civil plutonium as well.

2. A NON-FERTILE FUEL CONCEPT

A variety of different technical solutions for plutonium burning have been so far proposed and investigated. This paper concerns specifically the utilization of conventional PWRs partially fed with a new non-fertile oxide-type fuel. This combination could be an attractive option for disposition of plutonium. The utilization of heavy water reactors is under study as well, and relevant results will be presented in a further paper.

The proposed solution has been suggested by the Authors in 1992 and successively put forward in many occasions [5-8]. It seems that other people share this proposal, particularly in Japan (see here below).

To avoid any plutonium production during the irradiation, uranium-238 is completely absent in this special fuel. The fissile component, in PuO₂ format, is spatially diluted in an inert matrix which is highly resistant to radiation, scarcely neutron absorbent and has good thermo-physical properties. Chemical formats of the non-fertile matrix would be Al₂O₃, ZrO₂, MgO or their proper combinations. If needed, ThO₂ may also be considered to improve the fuel neutronic characteristics, but in this case, for the proliferation resistance of the spent fuel, it may be necessary to add some uranium-238 to dilute the uranium-233 coming from breeding of thorium-232. These fuels have substantially different characteristics than the usual MOX fuels because of the different composition and particularly of the absence of fertile nuclides.

Recently a JAERI research group published the results of a study in which this oxide-type fuel has been considered [9]. It is worth to briefly summarize the main results. The proposed oxide fuels are tailor-made multiphase fuels consisting of mineral compounds, and have been produced by the reaction between plutonium and some additives. From the phase relations of ceramic materials and their chemical properties, it seems that a two-phase mixture of a fluorite-type phase and alumina (suitable for stabilizing the spent fuel) has favourable characteristics for plutonium burning. It also seems that the fluorite-type phase such as thoria and fully stabilized zirconia are acceptable as host phases of plutonium because of the high solid solubility of the actinide elements and fission products, the irradiation stability and the chemical stability. The spent fuels finally obtained
from both systems PuO$_2$-ThO$_2$-Al$_2$O$_3$ and PuO$_2$-ZrO$_2$-Al$_2$O$_3$ will become mineral-like waste forms which would have chemical stability, high neutron emission rate, high heating rate and be quite radioactive, making it unattractive for subsequent use in weapons. The JAERI investigation demonstrates that this solution is technologically viable and could combine a high plutonium burning rate with a chemically stable form of the waste. Whatever the origin of plutonium, several technologies are fit for fuel fabrication: for example co-precipitation of fissile with matrix elements from solutions in a process leading to mixed oxides, and absorption, selective or not, of fissile ionic compounds of suitable porous carriers, followed by heat treatments or similar techniques.

It is worth noticing that the new oxide-type fuel should have a higher thermal conductivity than the standard UC$_2$O$_3$ one. This improves the heat transfer process from the fuel to the coolant, thus leading to lower centre-line temperatures. The fuel temperature may be kept either low to limit fission gas release and then the rod internal pressure and the cladding stress corrosion, or equal to the values of the usual fuel so the new plutonium-type fuel could sustain a higher power generation. Besides, a high thermal conductivity would allow the fuel to have short time constants, which is an important characteristic during transients.

Single pin cell together with preliminary assembly-level neutronic calculations have been recently presented [8] and are here only summarized. This paper describes a further development of the neutronic study by presenting more detailed assembly-level results.

3. REACTOR CORE DESIGN

Reactors involved in plutonium incineration would be PWRs operating in a once-through cycle scheme. The plutonium-poor spent fuel, handled as a high-radioactive waste, could be buried under deep geological formations without further reprocessing.

Three different core configurations may be considered:

1. the core is totally fed with non-fertile fuels;
2. a suitable fraction of the core is loaded with special assemblies containing the new oxide-type fuels, the residual part being still fed with standard UO$_2$ assemblies;
3. the core is entirely loaded with assemblies containing both UO$_2$ and plutonium-oxide fuel rods in proper proportions.

The first solution would be very interesting and attractive because it would allow the larger plutonium consumption rate. Nevertheless, the dynamic characteristics of the inert fuel could lead to considerable modifications of the current control systems design. In fact, the plutonium delayed neutron fraction $\beta$ is lower (0.21% for plutonium-239) than the uranium one (0.65% for uranium-235), and the absence of uranium-238, strong resonance absorber, decreases the fuel temperature coefficient (in absolute value). Moreover, the plutonium nuclear parameters (for example fast and epithermal cross sections) reduce, in absolute term, the void reactivity coefficient and the moderator reactivity coefficient as well.

The second solution can use the large experience of operating MOX-fuelled cores. A suitable arrangement of the special fuel assemblies in the core and a proper reloading strategy might reduce the power peaks and flatten the flux radial profile. Anyhow, the local behaviour during both operational and accidental transients has to be carefully investigated.

The third option seems to be the easiest to study. Some plutonium-type rods (20 to 30%) are inserted in the standard fuel assembly without any change to the present reloading strategy of current PWRs. The overall and local reactor behaviour would be driven by the standard UO$_2$ fuel and only limited perturbation would be induced by the inert fuel.

The partial loading of plutonium fuel, as envisaged in solutions 2 and 3, is also coherent with a scenario in which a plutonium-loaded fraction of the core could burn the plutonium produced in a previous fuel batch by the standard UO$_2$-loaded fraction at the equilibrium cycle.
The third solution was chosen for this analysis. The aim of the study is to investigate whether a high plutonium consumption rate may be achieved in current PWRs without any major modification to the reactor design. The resulting spent fuel should be plutonium-poor and proliferation resistant.

The reference reactor is a 3.2% enriched AP600-type one. The reactor main parameters are summarized in Table I.

<table>
<thead>
<tr>
<th>TABLE I: REFERENCE REACTOR MAIN PARAMETERS</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>pellet diameter</strong></td>
</tr>
<tr>
<td><strong>cladding outer diameter</strong></td>
</tr>
<tr>
<td><strong>pin pitch</strong></td>
</tr>
<tr>
<td><strong>pUO2</strong></td>
</tr>
<tr>
<td><strong>average linear heat rate</strong></td>
</tr>
<tr>
<td><strong>assembly layout</strong></td>
</tr>
<tr>
<td><strong>no. fuel rods / assembly</strong></td>
</tr>
</tbody>
</table>

The reference fuel is the assembly defined in Table I, in which 56 rods over a total of 264 are substituted with the new-fuel rods (see Figure 1). Two plutonium isotopic compositions were considered: civil plutonium and military-grade plutonium. The plutonium compositions are detailed in Table II. The plutonium mass content is an important parameter. Assuming as the reference value the uranium-235 mass contained in a standard 3.2% enriched UO2 fuel (Fissile Uranium, FU), we adopted fissile plutonium masses (plutonium-239+plutonium-241) in each rod equal to 0.71 FU, 1.0 FU and 1.3 FU respectively. This interval was chosen according to the following considerations relevant to civil plutonium: 0.71 FU implies that the total plutonium mass is equal to FU; 1.0 FU enables a direct comparison with the same initial fissile mass; 1.3 FU is one of the values adopted in a MOX fuel.

<table>
<thead>
<tr>
<th>TABLE II: PLUTONIUM ISOTOPIC COMPOSITIONS</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>isotope</strong></td>
</tr>
<tr>
<td>Pu-239</td>
</tr>
<tr>
<td>Pu-240</td>
</tr>
<tr>
<td>Pu-241</td>
</tr>
<tr>
<td>Pu-242</td>
</tr>
</tbody>
</table>

In any condition the remaining empty volume was completely filled with Al2O3, regardless of technological choices, but tending to be conservative in terms of neutron absorptions. A variant envisages the use of burnable poisons under the form of gadolinia (Gd2O3) or erbia (Er2O3).
4. STUDY METHODOLOGY

To verify the results reliability and validity, two different computer programmes were utilized. At first single pin cell calculations were performed by using the WIMS-E program [10] and its 69-groups ‘1986’ library [11] to assess the physical and neutronic characteristics of the new fuel. The results are detailed in Reference [8]. In this paper the CASMO-3 code (version 4.7) [12] was employed to study the fuel assembly in its actual layout.

CASMO is a multigroup 2-dimensional transport program for burnup calculations on fuel assemblies. The program handles a geometry consisting of cylindrical fuel rods of varying composition in a square pitch array with allowance for fuel rods loaded with gadolinium and erbium. Nuclear data are collected in a library containing microscopic cross sections in 40 energy groups (E4LTJB4 library). Neutron energies cover the range 0 to 10 MeV. Since we limit our study to assembly-level fuel depletion calculations, we adopt a reactivity-based cycling model [13] to determine an equilibrium cycle for each of the whole-core compositions under study. A realistic model of a particular reactor involves a detailed description of the neutron and fuel behaviour in position, energy and time as well. In fact, flux and power distributions vary between the fresh and the old fuel and such effects should be taken into account with a detailed analysis covering spatial and energy dependence. However, the essential features of fuel behaviour can be investigated by using a simple model neglecting this detail, assuming the time is the only independent variable. The simplifying assumption is that fuel batches of different exposures combined in the same core contribute to the overall reactivity in proportion to the reactivity in similar sized reactors, operating in a 1-batch cycle, loaded with fuel having an uniform exposure. Then, considering a reactor with $M$ fuel batches, each having a different burnup history, one might introduce the partial infinite multiplication factor of batch $i$ ($k_i^*$) as:

$$k_i^*(\tau^i) = \frac{k_i(\tau^i)}{M}$$

where $k_i$ is the infinite reactivity in a 1-batch operated reactor subject to fuel burnup $\tau^i$. Then, the $M$-batch operated reactor in this model has an overall infinite multiplication factor $\bar{k}$:

$$\bar{k} = \sum_{i=1}^{M} k_i^*(\tau^i) = \frac{1}{M} \sum_{i=1}^{M} k_i(\tau^i)$$

The EOL k-infinity of the equilibrium cycle was assumed equal to 1.066 to take into account both the leakage and additional parasitic absorptions. With this value the standard 3.2% enriched AP600 core reaches a discharge equilibrium burnup of 33.1 MWd/kgHM. Let $\theta$ the cycle time measured in a convenient unit. Effective Full Power Days (EFPD) were chosen to measure the irradiation time instead of the usual MWd/kgHM because of the different heavy metal content in the conventional and the special fuel rods. The above condition on the overall reactivity for the EOL of $n$th cycle can be expressed as:

$$\bar{k}(\theta_n) = \sum_{i=1}^{M} k_i(\tau^i) = 1.066$$

At the beginning of the next cycle (BOCn+1) the longest irradiated batch is removed and a fresh batch is loaded. If we assume convergence, i.e.
\[ \lim_{n \to \infty} \theta_n = \theta_{ss} \]

independent of \( n \), then, for sufficiently large \( n \), the discharge burnup of the equilibrium cycle is \( \theta^* = M \theta_{ss} \). Therefore, the EOC condition becomes:

\[
\frac{1}{M} \sum_{n=1}^{M} k(n \theta_{ss}) = 1.066
\]

which can be used for determining \( \theta^* \).

5. SINGLE PIN CELL CALCULATIONS

In the preliminary phase, detailed in Reference [8], we were interested in assessing some neutronic features of this new fuel, considered as a single rod separated from the assembly. Features of the standard AP600 fuel rod are kept as a reference.

The WIMS-E single pin calculations were carried out on the same fuel compositions here adopted with the exception of a further variant implying the addition of ThO\(_2\) filling 50% of the free volume of the rod. The depletion calculations were done with the assumption of imposing a thermal flux equal to that obtained in standard UO\(_2\) fuel irradiated at constant power. This means to assume in these calculations that the presence of the special rods induces only small perturbation of the flux distribution inside the fuel assembly. The obtained results are here summarized.

1. The new-type fuel seems to have the capability of an almost complete elimination of plutonium-239 (>97%), both for civil and military-grade plutonium. The plutonium-poor spent fuel contains large fractions of plutonium-240 and plutonium-242, thus rendering it completely proliferation resistant.

2. Plutonium fuel rod shows a high initial reactivity, followed by a steep decrease versus EFPD (Figure 2).

3. Temperature and void reactivity coefficients are much lower, in absolute value, than for standard fuel, reaching positive values at EOL for military-grade plutonium (Figures 3, 4 and 5).

4. Thoria bearing fuel behaves in a more similar way to UO\(_2\) fuel, but it shows a lower reactivity (Figure 2). Some concerns about the proliferation potential of the spent fuel exist due to the uranium-233 build-up. The initial reactivity could be increased through a larger initial plutonium inventory, but in this case, as well as in the original one, the fuel reprocessing should be necessary for the best exploitation of all the fissile material. Being this complex strategy out of the scope of our proposal, the thoria fuel will not be further considered in our study.

5. The power generation in the plutonium rods steeply decreases during the fuel life (Figure 6). This aspect was deemed to be better investigated by assembly-level calculations also considering the use of burnable absorbers.

6. ASSEMBLY LEVEL CALCULATIONS

Single pin cell neutronic calculations showed that the proposal seemed viable and promising as far as a useful plutonium burning in thermal reactors is concerned. However, the analysis was to be extended at assembly level in order to take into account the heterogeneity effects of the lattice, which were not included in these calculations. For this scope the CASMO-3 program was applied to the actual assembly geometry shown in Figure 1.
Neutronic calculations refer only to an infinite lattice without any soluble poison in the coolant. Depletion calculations were obtained by keeping constant the assembly power along the irradiation time. First of all a thorough comparison between WIMS-E and CASMO-3 results was carried out. It was found that the agreement between the two different programs is fully satisfactory. For instance, in Figure 7 the k-infinity versus EFPD for both programmes is shown for the AP600 and the special assembly which contains the plutonium rods having a plutonium content equal to 1.0 FU. In this latter case, WIMS-E results were obtained by weighing the reactivity values resulting from single pin cell calculations proportionally to the relative fraction of special and standard rods. In spite of this approximation and that on the thermal flux (see paragraph 5) the results are coherent and show the same overall trend. A similar agreement was also found for the reactivity coefficients, for which the different geometry conditions and assumptions may play a more significant effect.

CASMO-3 results are shown in the following figures. Figure 8 and 9 display the infinite multiplication factor versus EFPD for different special rod compositions. In these cases the presence of 208 standard rods over a total of 264 and the correct description of the assembly flux distribution render the k-infinity curves of the various assemblies closer to the reference one (see also Figure 2). Figures 10 and 11 show the evolution of the generated power (normalized to the average one) during the irradiation for the two rods having the peak factor at BOL (plutonium rod) and EOL (uranium rod). The peak factor ranges from 1.7 to 1.3 at BOL to 1.25 to 1.14 at EOL depending on fissile plutonium concentration. While the initial peak in plutonium rods might be accepted if its thermal conductivity were relatively high, this is not the case for the standard rods at EOL. This aspect appears important, because it could affect the possibility to operate at high assembly power. Fuel cycle management should not be of great help to this concern, because the present out-in fuel cycle strategy implies that the two older batches be located in the central part of the core, which is a high-flux zone. On the other hand the plutonium fuel should determine a more flatten flux both in radial and axial directions, so to decrease the maximum assembly power. This will be investigated in the near future.

The burnable poison insertion in all plutonium rods was envisaged to reduce the initial peak factor. This solution was studied only to dampen this effect and not for the long-term reactivity control as usually done. Obviously this latter possibility is still applicable in this particular context. The burnable poisons were gadolinia (Gd₂O₃) or erbia (Er₂O₃).

The study requirements were: i) the initial infinite reactivity of the three-batches core should not be lower than 1.066; ii) burnable poisons should be completely burnt within the first cycle, in order to avoid any penalty in the overall discharge burnup. Some results are shown in Figures 12 to 15 in terms of k-infinity and normalized power generation versus EFPD. The erbia clearly appears a not viable solution. Erbium is burnt very slowly along the whole fuel life, because of its relatively low absorption cross section (Figure 12). In spite of a substantial reactivity penalty at the end of the first cycle (about 400 EFPD), the initial power peak undergoes only a limited reduction (Figure 13). On the other hand gadolinia appears convenient as it is evident from Figures 14 and 15, which show that the reactivity maximum pass from 1.37 to 1.24 and the initial peak from 1.52 to 1.35 for the unpoisoned case and the poisoned one with 1.2% of gadolinia respectively. The burnable poison percentages are referred to the inert matrix weight (density around 4200 kg/m³).

As for the uranium peak at EOL an important countermeasure is the reduction of the nominal fuel power density. Therefore, our choice to assume as reference reactor the AP600, which is characterized by a lower power density than conventional PWRs, seems to be well justified.

Reactivity coefficients concerning moderator voids and temperature (and then density) are shown in Figures 16, 17 and 18. The void reactivity coefficient is not constant with the void fraction and then the coefficients for two different void percentages are given. These plots show that the plutonium rods, in general, worsen the reactivity coefficients rendering their value less negative. However, the limited percentage of plutonium rods renders
the various curves not too different from the standard uranium ones. It can be concluded that the insertion of these special rods should not change appreciably the reactor dynamic behaviour.

The isotopes evolution during the irradiation is displayed for plutonium content 1.0 FU in Figures 19 and 20 both for military-grade and civil plutonium. Peculiar characteristics of plutonium rods at BOL and EOL for all rod compositions are shown in Table III and IV.

**TABLE III: FUEL BURNUP AND EOL INVENTORY FOR MILITARY- GRADE PU (WT% OF INITIAL HM)**

<table>
<thead>
<tr>
<th></th>
<th>0.71 FU</th>
<th>1.0 FU</th>
<th>1.3 FU</th>
</tr>
</thead>
<tbody>
<tr>
<td>EPPD</td>
<td>1128</td>
<td>1209</td>
<td>1294</td>
</tr>
<tr>
<td>Pu-239</td>
<td>0.833 wt%</td>
<td>1.037 wt%</td>
<td>1.222 wt%</td>
</tr>
<tr>
<td>Pu-240</td>
<td>5.388 wt%</td>
<td>6.919 wt%</td>
<td>8.147 wt%</td>
</tr>
<tr>
<td>Pu-241</td>
<td>4.658 wt%</td>
<td>4.935 wt%</td>
<td>5.121 wt%</td>
</tr>
<tr>
<td>Pu-242</td>
<td>5.305 wt%</td>
<td>4.921 wt%</td>
<td>4.610 wt%</td>
</tr>
<tr>
<td>Burnt Pu-239</td>
<td>99.10%</td>
<td>98.88%</td>
<td>98.68%</td>
</tr>
<tr>
<td>Burnt total Pu</td>
<td>83.81%</td>
<td>82.19%</td>
<td>80.90%</td>
</tr>
</tbody>
</table>

**TABLE IV: FUEL BURNUP AND EOL INVENTORY FOR CIVIL PU (WT% OF INITIAL HM)**

<table>
<thead>
<tr>
<th></th>
<th>0.71 FU</th>
<th>1.0 FU</th>
<th>1.3 FU</th>
</tr>
</thead>
<tbody>
<tr>
<td>EPPD</td>
<td>1105</td>
<td>1175</td>
<td>1248</td>
</tr>
<tr>
<td>Pu-239</td>
<td>0.812 wt%</td>
<td>1.116 wt%</td>
<td>1.465 wt%</td>
</tr>
<tr>
<td>Pu-240</td>
<td>7.958 wt%</td>
<td>10.174 wt%</td>
<td>11.929 wt%</td>
</tr>
<tr>
<td>Pu-241</td>
<td>5.777 wt%</td>
<td>6.266 wt%</td>
<td>6.677 wt%</td>
</tr>
<tr>
<td>Pu-242</td>
<td>9.801 wt%</td>
<td>9.260 wt%</td>
<td>8.798 wt%</td>
</tr>
<tr>
<td>Burnt Pu-239</td>
<td>98.60%</td>
<td>98.08%</td>
<td>97.47%</td>
</tr>
<tr>
<td>Burnt total Pu</td>
<td>75.65%</td>
<td>73.18%</td>
<td>71.13%</td>
</tr>
</tbody>
</table>

Fuel burnup is practically identical to that of standard UO₂ (1187 EFPD) at plutonium content equal to 1.0 FU, with a slight improvement for military-grade plutonium due to the limited presence of the even plutonium isotopes. It varies almost linearly with the plutonium content, with a coefficient approximately equal to 24 and 28 EFPD/0.1 FU for civil and military grade plutonium respectively. The burnt fraction of plutonium-239 is very high in any case, around 98% for civil plutonium and 99% for military-grade plutonium. The total plutonium burnt fraction ranges from 71 to 76% for civil plutonium and from 80 to 84% for military-grade plutonium. Therefore the final composition of unloaded
plutonium, which contains from 66 to 72% of plutonium-240 and plutonium-242, for military-grade and civil plutonium respectively, is such to guarantee in any condition the lack of proliferation potential. This fact allows us to choose the plutonium content on the basis of an overall optimization study. These values might be modified at core-level, and this will be verified in the near future.

5. CONCLUDING REMARKS

The proposal of burning civil and military-grade plutonium in special non-fertile rods uniformly dispersed in PWRs assemblies in a fraction of about 20% were studied by single pin cell and assembly-level calculations. The main characteristic neutronic aspects at assembly-level are as follows.

1. The new fuel seems to have the capability of an almost complete elimination of plutonium-239. On average from 97 to 99% of it is burnt at EOL. The total burnt plutonium fraction ranges from 71 to 84% of initial loaded. The plutonium-poor spent fuel contains a major fraction of plutonium-240 and plutonium-242 (from 66 to 72%), and then it is definitely not usable for weapon purposes.

2. The slightly higher initial reactivity can be coped with by the usual long-term control solutions and/or by the insertion of burnable poisons in plutonium rods, this latter solutions having also the scope to reduce initial power peaks.

3. Reactivity coefficients, which are much less negative for single plutonium rods than the standard UO₂ values, are only slightly lower when evaluated at assembly-level, because of the prevalent fraction of uranium rods in the assembly. Then the dynamic behaviour of the reactor should not be appreciably affected.

4. The power distribution between plutonium and standard rods in the same assembly changes appreciably during the fuel life. While initial peaks in plutonium rods might be probably accepted due to their relatively high thermal conductivity, which improves fuel performance, this is not the case for standard rods at EOL. This aspect appears important also for its effects on DNB limits, but it is dampened by the present trend to reduce power density in PWR cores. Then the initial choice to assume as reference reactor the AP600, which is characterized by a 25% lower power density than conventional PWRs, is well justified. More pronounced flattening effects of plutonium fuel might decrease the maximum assembly power value. Insertion of limited amounts of gadolinia might face the initial power peak in the plutonium-type fuel.

The present choice to have a uniform dispersion of plutonium rods in the standard UO₂ assembly appears reasonable under many aspects, but a solution which envisages also a uniform dispersion of standard uranium assemblies and plutonium-type assemblies is presently under study.

These neutronic results confirm the viability of the proposed solution, and the need to examine closely the power peak problem also by core-level calculations. Then the open question is the technological assessment of the non-fertile fuel matrix. Available references reveal that this assessment is in progress at JAERI. However, other research programmes in the world are devoted to such materials both for nuclear and non-nuclear applications. As for the first ones let us remind many actinides burning proposals which envisage the adoption of these materials. As for the latter ones many activities are in progress also in our country.
Figure 1. Assembly layout

Figure 2. Single pin cell infinite multiplication factor versus EFPD
Figure 3. Single pin cell temperature coefficient versus EFPD

Figure 4. Single pin cell 50% void coefficient versus EFPD
Figure 5. Single pin cell 10% void coefficient versus EFPD

Figure 6. Single pin cell fission rate per rod unit length versus EFPD
Figure 7. WIMS-E and CASMO-3 infinite multiplication factor versus EFPD

Figure 8. Assembly-level infinite multiplication factor versus EFPD for military-grade plutonium
Figure 9. Assembly-level infinite multiplication factor versus EFPD for civil plutonium.

Figure 10. Assembly-level normalized generated power versus EFPD for military-grade plutonium.
Figure 11. Assembly-level normalized generated power versus EFPD for civil plutonium

Figure 12 Effect of erbia poisoning on the infinite multiplication factor
Figure 13. Effect of erbia poisoning on the normalized generated power

Figure 14. Effect of gadolinia poisoning on the infinite multiplication factor
Figure 15. Effect of gadolinia poisoning on the normalized generated power

Figure 16. Assembly-level 50% void coefficient versus EFPD
Figure 17. Assembly-level 10% void coefficient versus EFPD

Figure 18. Assembly-level moderator coefficient versus EFPD
Figure 19. Pu-239 and Pu-240 evolution versus EFPD for plutonium content 1.0 FU

Figure 20. Pu-241 and Pu-242 evolution versus EFPD for plutonium content 1.0 FU
ACKNOWLEDGEMENTS

The WIMS-E and CASMO-3 programs were used at the OECD-Halden Reactor Project (Norway) and at the ENEL-ATN offices (Italy), respectively. Cooperation of these organizations in making these programmes available is greatly acknowledged.

REFERENCES

A range of mixtures of plutonium and different burnable poisons (BPs) have been considered from the viewpoint of providing a possible LWR fuel without uranium. The reactor physics feasibility of such materials for enabling a considerably more effective incineration of plutonium than possible with PuO$_2$/UO$_2$ (MOX) fuel has been investigated in terms of burnup reactivity swings, temperature and void coefficients and control absorber worths. The potential for reduction of total-Pu has been shown to be ~3 times greater than with MOX, the difference in possible reduction factors being much more striking in terms of the quantity of fissile-Pu. It has been found that the specific effects of a given BP are crucial in determining the burnup and controllability characteristics of the hypothetical LWR fuel considered. As such, an improved overall performance can result from using a suitable mixture of BPs, rather than one individual type. Further, the influence of the "inert matrix" employed for the Pu-BP fuel can also be significant - on the void coefficient, in particular. The need has thus been indicated for optimizing the Pu-BP fuel composition in terms of both the BPs and the inert matrix. Finally, the scoping character of the present investigations needs to be stressed. The various results have all been obtained on the basis of unit cell and simplified fuel assembly calculations, and there has been little explicit consideration of materials aspects as such.

1 Introduction

The availability of plutonium in quantities far beyond projected needs is a relatively novel situation. With LWRs likely to continue to dominate the nuclear energy scene, it has become important to consider their potential role for reducing - and thereby helping to regulate - the large plutonium inventories world-wide. Thus, while the "self-generated" mode of plutonium recycle in LWRs, involving the use of PuO$_2$/UO$_2$ (mixed oxide, or MOX) for constituting 30 to 40% of the core, is coming into broader practice in Europe [1], it does not represent a possible solution in the above context. This is because about the same amount of new plutonium is generated as that consumed. Designs for 100%-MOX cores are being investigated, and an effective reduction of plutonium would certainly be achievable with these [2]. The buildup of new plutonium from the uranium present in the MOX, however, remains disadvantageous from the viewpoint of inventory reduction and also enhances the need for multiple recycling.

The present paper extends investigations reported recently on the physics feasibility of LWR plutonium fuels without uranium, [3, 4]. Mixtures of plutonium and different individual burnable poisons (BPs) - as well as the possible use of a combination of BPs - have been considered. Without specific regard having been given to materials aspects, the physics performance of these hypothetical fuels is assessed in terms of burnup reactivity...
swings, temperature and void coefficients and control absorber worthwhiles. The much higher potential for reducing plutonium inventory is compared with that for a 100%-MOX core, as are also the isotopic composition of the discharged plutonium and the amounts of minor actinides (Np, Am, Cm) generated. Further, the influence of the "inert matrix" has been investigated – particularly with respect to reactivity coefficients.

2 Calculational Models

The various results being presented were all obtained on the basis of unit cell and/or simplified fuel assembly calculations. In each case, the BOXER code which forms part of the LWR code system, ELCOS, was used [5].

2.1 Cell and Assembly Descriptions

Since compatibility with current-day pressurized water reactors (PWRs) is an important goal, the unit cells considered have been of PWR-type with common characteristics as given in Table 1.

<table>
<thead>
<tr>
<th>Table 1: Common Characteristics for the Unit Cells</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fuel diameter: 9.13 mm Hot full power (HFP) conditions:</td>
</tr>
<tr>
<td>Clad(material): zircaloy Fuel temp.: 600 °C</td>
</tr>
<tr>
<td>inner diam.: 9.30 mm Clad temp.: 315 °C</td>
</tr>
<tr>
<td>outer diam.: 10.75 mm Water temp.: 300 °C</td>
</tr>
<tr>
<td>Square pitch: 14.30 mm Pressure: 158.8 bar</td>
</tr>
<tr>
<td>Boron in water: none Power density: 103 MW/m³</td>
</tr>
</tbody>
</table>

Six individual BPs, as well as one particular BP-mixture, were selected for the present study, the hypothetical fuel material in each case consisting of the particular BP mixed with PuO₂ and dispersed in an "inert matrix". The plutonium density assumed was 0.8 g/cm³, i.e. close to the values which occur in LWR MOX fuels, and the inert matrix for most of the cases calculated (all, except those discussed additionally in Section 3.5) was Al₂O₃ of density 3.0 g/cm³. The BP density, in each case, was adjusted to yield a kᵢₒ of ~ 1.13 at beginning of life (BOL) with HFP (Xe-free) conditions. Table 2 gives the various poison density values used, the single BP-mixture considered being 40/60 atom% boron/holmium. The isotopic composition for the BPs corresponded to the natural composition in each case (boron, rhodium, indium, europium, holmium and hafnium).

In order to provide comparatory values for the assessment of performance indices, two reference LWR unit cells have been considered, viz.

(1) with UO₂ fuel of 4.3 w% ²³⁵U enrichment and 10.4 g/cm³ density and
(2) with MOX fuel of density 10.2 g/cm$^3$, containing 0.69 g/cm$^3$ plutonium of 71.2 w% fissile enrichment, the uranium being of 0.25 w% enrichment.

The isotopic composition assumed for the plutonium in the various Pu-BP fuels approximated that obtained from LWR UO$_2$ fuel with a burnup of 42 GWd/t and 5 y cooling time [2]. This composition is compared below with that considered for the reference MOX cell:

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Pu-BP:</th>
<th>MOX:</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{238}$Pu</td>
<td>2.7</td>
<td>1.8</td>
</tr>
<tr>
<td>$^{239}$Pu</td>
<td>54.5</td>
<td>59.0</td>
</tr>
<tr>
<td>$^{240}$Pu</td>
<td>22.8</td>
<td>23.0</td>
</tr>
<tr>
<td>$^{241}$Pu</td>
<td>11.7</td>
<td>12.2</td>
</tr>
<tr>
<td>$^{242}$Pu</td>
<td>8.3</td>
<td>4.0</td>
</tr>
</tbody>
</table>

For the relative evaluation of control rod worths, a single fuel assembly configuration has been considered. This consisted of 20 control rod positions in a 15 x 15 assembly, as indicated in Fig.1.

Table 2: Density and Isotopic Compositions Assumed for the Burnable Poisons (Pu and Al$_2$O$_3$ densities in the fuel zone are 0.8 and 3.0 g/cm$^3$, respectively, in each case)

<table>
<thead>
<tr>
<th>Cell Type (BP)</th>
<th>BP density g/cm$^3$</th>
<th>BP isotopic composition</th>
<th>atomic weights</th>
</tr>
</thead>
<tbody>
<tr>
<td>B</td>
<td>0.0188</td>
<td>20.0/80.0</td>
<td>10/11</td>
</tr>
<tr>
<td>Rh</td>
<td>0.467</td>
<td>100</td>
<td>103</td>
</tr>
<tr>
<td>In</td>
<td>0.27</td>
<td>4.3/95.7</td>
<td>113/115</td>
</tr>
<tr>
<td>Eu</td>
<td>0.0408</td>
<td>47.8/52.2</td>
<td>151/153</td>
</tr>
<tr>
<td>Ho</td>
<td>0.38</td>
<td>100</td>
<td>165</td>
</tr>
<tr>
<td>Hf</td>
<td>0.325</td>
<td>0.2/5.2/18.6/27.1/13.7/35.2</td>
<td>147/148/149/150/152/154</td>
</tr>
<tr>
<td>B+Ho</td>
<td>0.15</td>
<td>8.0/32.0/60.0</td>
<td>10/11/165</td>
</tr>
</tbody>
</table>

### 2.2 Calculational Methods and Data

The present analyses were carried out using the BOXER code which forms part of the LWR code system, ELCOS [5]. Both cell and two-dimensional transport/depletion calculations can be performed by BOXER, the former in 70 energy groups and the latter after collapsing to a reduced number of broad groups. Eight were used for the current fuel assembly calculations.

An important feature of the cell calculation is the treatment of the resonance energy range between 1.3 and 907 eV, self-shielding of the cross-sections being achieved via a two-region collision probability calculation in ~ 7500 energy points. Resonance interaction effects are thereby taken into account explicitly. Above 907 eV, the resonance cross-sections are interpolated in the more usual fashion, i.e. as functions of temperature and dilution employing appropriate tables.
The data library used for the BOXER calculations was generated from the Joint European File, JEF-1 [6]. Considerable refinement of the burnup treatment was undertaken recently in the context of actinide transmutation studies [7], and the present library contains 34 actinides, from $^{232}$Th to $^{248}$Cm, and 55 explicit (plus 2 pseudo) fission products. For the current study, data was generated for additional nuclides corresponding to the BPs considered.

Validation of BOXER calculations, using its JEF-1 library, has been carried out in the past largely on the basis of $\text{H}_2\text{O}$-moderated, UO$_2$ criticals [8]. In order to enable a more relevant assessment for the present calculations, some of the 7.5% $\text{Pu}_{\text{fiss}}$ MOX experiments in the PROTEUS-LWHCR programme [9] have also been analysed. Satisfactory agreement was obtained with experimental results, reflecting adequate accuracies for calculating individual reaction rates in the plutonium isotopes [4].

### 3 Calculational Results

The principal results discussed for the various Pu-BP fuels pertain to the reactivity variation with burnup (which is a constraint on the maximum core-residence time and hence on the effective plutonium reduction achievable), the changes in individual nuclide densities, reactivity coefficients and control absorber worths, as well as effects of the inert matrix on the physics characteristics.

#### 3.1 Reactivity Variation with Burnup

Because of the different absorption cross-sections and burnup chains involved, cell calculations for the various Pu-BP fuels yielded large differences in the reactivity variation with burnup. Fig.2 shows the results for the seven cases considered and compares these with the corresponding variations for the reference UO$_2$ and MOX cells (Section 2.1).
It is seen that relatively short core-residence times would be achievable with Rh, In, Ho or Hf as BP. On the other hand, as indicated in Section 3.3, individual reactivity coefficients for several of these Pu-BP fuels are found to be quite favourable.

With B and Eu, there is a slight increase in reactivity initially before the fall occurs, so that up to ~1500 irradiation days (corresponding to ~53 GWd/t for a current-day PWR) would be possible – from reactivity loss considerations alone. A relatively flat reactivity variation was reported earlier also with erbium [4]. In all these three cases, however,

Figure 2: $k_{\infty}$ variations with burnup for the Pu-BP unit cells (a) with Rh, In, Ho and Hf as the BP and (b) with B, Eu and a mixture of 40% B and 60% Ho. Results for the reference UO$_2$ and MOX LWR cells are shown for comparison.
there are certain difficulties with respect to the reactivity coefficients – which provides the incentive to consider mixing BPs for achieving a more favourable overall behaviour. The 40% B - 60% Ho case, depicted in Fig.2(b), provides an illustrative example in the current study.

### 3.2 Nuclide Density Changes

Apart from changes for the Pu-isotopes, the production of the minor actinides – Np, Am and Cm – has also been considered, with 1500 irradiation days assumed in each case. All nuclide densities have been normalized to the initial amount of plutonium, i.e. that at zero burnup. Since the results obtained were very similar for the various Pu-BP fuels, two representative cases (for which the 1500 irradiation days would indeed be feasible) are depicted in Tables 3 and 4, viz. Pu-Eu and Pu-B-Ho. In each case, comparative results are given for the reference MOX cell.

#### Table 3: Total-Pu and Pu-Isotopics after 1500 Days of Irradiation

<table>
<thead>
<tr>
<th>Cell Type</th>
<th>Final/Initial Pu (total)</th>
<th>Final Pu-Isotopics [w%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>MOX</td>
<td>0.697</td>
<td>2.6 39.7 29.7 17.9 10.2</td>
</tr>
<tr>
<td>Pu-Eu</td>
<td>0.315</td>
<td>5.6 7.6 32.8 19.9 34.1</td>
</tr>
<tr>
<td>Pu-B-Ho</td>
<td>0.320</td>
<td>5.5 7.3 32.7 19.7 34.7</td>
</tr>
</tbody>
</table>

#### Table 4: Nuclide Densities for the Minor Actinides after 1500 Days of Irradiation

<table>
<thead>
<tr>
<th>Cell Type</th>
<th>Total MAs Initial Pu</th>
<th>MA Composition [w%]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>$^{237}$Np $^{241}$Am $^{243}$Am $^{242}$Cm $^{244}$Cm $^{245}$Cm Others</td>
</tr>
<tr>
<td>MOX</td>
<td>0.055</td>
<td>6.0 19.5 40.2 4.3 25.6 3.2 1.2</td>
</tr>
<tr>
<td>Pu-Eu</td>
<td>0.063</td>
<td>0.0 6.7 45.9 4.3 37.6 3.9 1.4</td>
</tr>
<tr>
<td>Pu-B-Ho</td>
<td>0.067</td>
<td>0.0 7.0 46.5 4.5 36.9 3.7 1.6</td>
</tr>
</tbody>
</table>

As indicated above, the total-Pu reduction achieved is about 68% for the Pu-BP fuels, ~ 62% being fissioned and 6 to 7% being converted into MAs. This corresponds to a plutonium “incineration” rate of ~ 1.14 kg/GWd, i.e. a figure quite comparable to that being aimed at for advanced concepts in the French CAPRA program with fast reactors [10]. The result for the MOX fuel is a total-Pu reduction of 30% (~ 0.38 kg/GWd), with the conversion to MAs only slightly lower at 5.5%. If one considers the changes in Pu-isotopics in conjunction with the total-Pu reduction, one finds that the $^{239}$Pu content of the Pu-BP fuels is reduced by as much as 95%, as compared to ~ 47% in the MOX case.
As regards the minor actinides, the absence of uranium accounts for there being no $^{237}$Np produced in the Pu-BP fuels. The fact that a much lower amount of $^{241}$Am is found to occur than in the MOX case (even though its production is mainly via $^{241}$Pu decay) is due to the thermal neutron flux in the Pu-BP cells becoming several times higher towards the end of the irradiation period. $^{241}$Am is then converted to $^{242}$Am much more effectively, and the latter, in turn – due to its large fission cross-section – to fission products. The higher thermal neutron flux also results in more captures in $^{242}$Pu, whereby the fraction of $^{244}$Cm produced is larger than in the MOX case. Qualitatively speaking, the longterm radiotoxicity of the discharged fuel due to all the higher actinides – including the Pu-isotopes – would be several times lower in the Pu-BP fuels than for the MOX.

### 3.3 Reactivity Coefficients

An initial assessment of safety and controllability characteristics can be made by comparing reactivity coefficients and control absorber worths for the Pu-BP cells considered, relative to those for the reference UO$_2$ and MOX cases. The relation used for defining a reactivity coefficient of type $x$ (Doppler or void) with respect to the variation of the corresponding parameter $p$ for the cell is:

$$ C_x^i = \frac{k_i(p + \Delta p) - k_i(p)}{\overline{k}_i \cdot \Delta p} $$

with $k_i$ denoting either $k_\infty$ or $k_{\text{eff}}$, and with

$$ \overline{k}_i = \frac{k_i(p + \Delta p) + k_i(p)}{2} $$

Table 5 gives values calculated for the Doppler and void coefficients at BOL. The former has been considered in terms of the $k_\infty$ change for an increase in fuel temperature from 600°C to 1000°C. For the void coefficient, moderator voidage ranges of 0 to 10% and 0 to 100% have been considered separately. In the case of partial voidage (0 to 10%), it is adequate to consider the coefficient in terms of the change in $k_{\infty}$. For 0 to 100% void, however, $k_{\text{eff}}$ variations need to be considered since changes in migration area ($M^2$) are quite different for the cells with and without uranium. This is indicated by the comparison of $M^2$-values made in Table 6. A relative assessment of the void coefficient for 0 to 100% voidage without consideration of leakage effects would clearly be too pessimistic for the Pu-BP cells, considering that the $M^2$-changes are almost twice as large as for the UO$_2$ and MOX cases. A geometrical buckling of 3.1 m$^{-2}$ – corresponding approximately to the dimensions of a 1000 MW(e) PWR core – was assumed for deducing the $C_{\text{eff, void}}$ (0 to 100%) results of Table 5.

It is seen, as mentioned earlier (Section 3.1), that the Pu-fuels with single BPs, which have reactivity coefficients of comparable magnitude to those for the reference UO$_2$ and MOX cells, correspond to the cases with relatively large reactivity losses with burnup. The B+Ho BP-mixture considered, on the other hand, appears to combine the favourable features of a flatter reactivity variation and acceptable reactivity coefficient values quite effectively.
Table 5: Reactivity Coefficients for the Various Unit Cells at Zero Burnup

<table>
<thead>
<tr>
<th>Cell Type</th>
<th>$C_{\text{Doppler}}^\infty$ (10^{-8}^\circ\text{C}^{-1})</th>
<th>$C_{\text{void}}^\infty$ (0 to 10%) (10^{-4}/%\text{void})</th>
<th>$C_{\text{void}}^{\text{eff}}$ (0 to 100%) (10^{-4}/%\text{void})</th>
</tr>
</thead>
<tbody>
<tr>
<td>UO$_2$</td>
<td>-2.4</td>
<td>-15.2</td>
<td>-77.1</td>
</tr>
<tr>
<td>MOX</td>
<td>-3.1</td>
<td>-18.5</td>
<td>-43.3</td>
</tr>
<tr>
<td>B</td>
<td>-1.0</td>
<td>-3.9</td>
<td>-1.6</td>
</tr>
<tr>
<td>Rh</td>
<td>-1.3</td>
<td>-6.5</td>
<td>-12.3</td>
</tr>
<tr>
<td>In</td>
<td>-1.4</td>
<td>-8.6</td>
<td>-0.8</td>
</tr>
<tr>
<td>Eu</td>
<td>-1.2</td>
<td>-7.1</td>
<td>-1.6</td>
</tr>
<tr>
<td>Ho</td>
<td>-2.5</td>
<td>-20.0</td>
<td>-21.6</td>
</tr>
<tr>
<td>Hf</td>
<td>-2.1</td>
<td>-15.6</td>
<td>-3.1</td>
</tr>
<tr>
<td>B+Ho</td>
<td>-1.7</td>
<td>-12.2</td>
<td>-13.4</td>
</tr>
</tbody>
</table>

Table 6: $M^2$-values at (a) 0% and (b) 100% Moderator Voidage for Several Unit Cells

<table>
<thead>
<tr>
<th>Cell Type</th>
<th>$k_\infty$</th>
<th>Crit. Buckling</th>
<th>$M^2$ [cm$^2$]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>(a)</td>
<td>(b)</td>
<td>(a) (b)</td>
</tr>
<tr>
<td>UO$_2$</td>
<td>1.375</td>
<td>0.644</td>
<td>65.3 -6.76</td>
</tr>
<tr>
<td>MOX</td>
<td>1.205</td>
<td>0.848</td>
<td>37.9 -2.70</td>
</tr>
<tr>
<td>B</td>
<td>1.134</td>
<td>1.486</td>
<td>21.8 4.30</td>
</tr>
<tr>
<td>Ho</td>
<td>1.133</td>
<td>1.127</td>
<td>22.5 1.41</td>
</tr>
<tr>
<td>B+Ho</td>
<td>1.176</td>
<td>1.339</td>
<td>29.0 3.24</td>
</tr>
</tbody>
</table>

Fig.3 indicates how the BOL values of the void coefficients change with burnup, the Pu-BP fuels being represented by the B+Ho case. Particularly for the 0 to 100% void coefficient, the change to more strongly negative values is much more pronounced for the Pu-BP cell than for the reference uranium-containing fuels. It should be mentioned that the variation of Doppler coefficients with burnup was relatively small for all cases.

### 3.4 Control Absorber Worths

The considerable reduction of control absorber worths (soluble boron and control rods), as the fraction of MOX assemblies is increased, is a major constraint in designing a PWR core with 100% MOX fuel [11]. This results from the larger absorption cross-sections of the Pu-isotopes, relative to uranium. A similar situation would be expected for systems with Pu-BP fuels, but this has to be considered in conjunction with the corresponding control requirements.
The reactivity swing due to burnup, as indicated earlier, could be significantly lower for a suitably chosen Pu-BP fuel than for the reference UO₂ case. Table 7 compares two other types of control requirements, viz. that for equilibrium xenon and the cold-to-HFP reactivity swing. Here, too, the control needs for the Pu-BP cells are generally lower than for the reference fuels. Also shown in Table 7 are the corresponding results for (i) soluble boron worth and (ii) control-rod cluster worth (cf. Section 2.1). Both types of control absorber effects for the Pu-BP cases are seen to be very similar to those for MOX.

Figure 3: Variation with burnup of void coefficient values for voidage ranges of (a) 0 to 10% and (b) 0 to 100%
Table 7: Comparison of Typical Control Requirements and Control Absorber Worths

<table>
<thead>
<tr>
<th>Cell Type</th>
<th>Control Requirements</th>
<th>Control Absorber Worths</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Equilib. Xe (Δk∞, %)</td>
<td>Cold-to-HFP (Δk∞, %)</td>
</tr>
<tr>
<td>UO₂</td>
<td>-3.1</td>
<td>-5.7</td>
</tr>
<tr>
<td>MOX</td>
<td>-1.6</td>
<td>-7.8</td>
</tr>
<tr>
<td>B</td>
<td>-0.9</td>
<td>-0.7</td>
</tr>
<tr>
<td>Rh</td>
<td>-1.1</td>
<td>-2.9</td>
</tr>
<tr>
<td>In</td>
<td>-1.2</td>
<td>-4.1</td>
</tr>
<tr>
<td>Eu</td>
<td>-1.0</td>
<td>-1.8</td>
</tr>
<tr>
<td>Ho</td>
<td>-1.4</td>
<td>-7.9</td>
</tr>
<tr>
<td>Hf</td>
<td>-1.3</td>
<td>-6.6</td>
</tr>
<tr>
<td>B+Ho</td>
<td>-1.2</td>
<td>-4.6</td>
</tr>
</tbody>
</table>

The above considerations have been made for BOL and clearly need to be extended to account for representative average core conditions. That the control absorber worths for the Pu-BP fuels increase quite strongly with burnup is indicated in Fig.4. This is again linked to the increase in thermal neutron flux with irradiation, particularly towards end-of-life with fuel cross-sections reduced considerably. Clearly, as in the case of reactivity coefficient assessment, realistic whole-reactor calculations are needed to evaluate features related to controllability – both in terms of reactivity balance and power distributions.

3.5 Effects of the Inert Matrix

The various results presented for Pu-BP fuels have all, till now, been with Al₂O₃ considered as the inert matrix (Section 2.1). The latter’s influence on the physics characteristics has been currently investigated by calculating cases with certain possible alternatives, viz. BeO, ¹¹B₄C and ZrC. The most significant effects observed were on the reactivity coefficients, particularly the 0 to 100% void coefficient, as indicated in Table 8.

Table 8: Comparison of Reactivity Coefficient Values for Pu-BP fuels with 40% B - 60% Ho as BP and Alternative Materials as Inert Matrix

<table>
<thead>
<tr>
<th>Inert Matrix</th>
<th>Density [g/cm³]</th>
<th>C^Doppler [10⁻⁵/°C]</th>
<th>C^void (0 to 10%) [10⁻⁴/% void]</th>
<th>C^void (0 to 100%) [10⁻⁴/% void]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al₂O₃</td>
<td>3.0</td>
<td>-1.7</td>
<td>-12.2</td>
<td>-13.4</td>
</tr>
<tr>
<td>BeO</td>
<td>2.5</td>
<td>-2.0</td>
<td>-12.6</td>
<td>-12.1</td>
</tr>
<tr>
<td>¹¹B₄C</td>
<td>2.0</td>
<td>-1.7</td>
<td>-13.0</td>
<td>-13.3</td>
</tr>
<tr>
<td>ZrC</td>
<td>5.8</td>
<td>-1.6</td>
<td>-13.0</td>
<td>-15.1</td>
</tr>
</tbody>
</table>

There are two individual effects which contribute to the influence of the inert matrix on C^void (0 to 100%). The first is the moderating effect in the fully voided condition, which can shift the hard neutron spectrum to significantly lower energies resulting in
increased resonance capture in the BP and a lower $k_{\infty}$ value. The second – often partly compensating – effect is the influence on the migration area which determines the leakage characteristics. Fig. 5 and Table 9 illustrate the two individual effects.

![Graph](image)

Figure 4: Variation with burnup of (a) soluble boron worth and (b) control-rod worth

Again, realistic whole-reactor models clearly need to be considered for assessing the influence of the inert matrix on void reactivity. Which type of inert matrix is ultimately preferable will, of course, also be determined by various material properties including irradiation behaviour.
Figure 5: Neutron spectra for Pu-BP fuels with 40% B - 60% Ho as BP and Al₂O₃ and BeO as inert matrix

Table 9: $k_{\infty}$ and $M^2$-values at (a) 0% and (b) 100% Moderator Voidage for Pu-BP fuels with 40% B - 60% Ho as BP and Alternative Materials as Inert Matrix

<table>
<thead>
<tr>
<th>Cell Type</th>
<th>$k_{\infty}$ (a)</th>
<th>$k_{\infty}$ (b)</th>
<th>Crit. Buckling [m⁻²] (a)</th>
<th>Crit. Buckling [m⁻²] (b)</th>
<th>$M^2$ [cm²] (a)</th>
<th>$M^2$ [cm²] (b)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al₂O₃</td>
<td>1.176</td>
<td>1.339</td>
<td>29.0</td>
<td>3.24</td>
<td>60.8</td>
<td>1048</td>
</tr>
<tr>
<td>BeO</td>
<td>1.188</td>
<td>1.182</td>
<td>38.4</td>
<td>4.11</td>
<td>49.0</td>
<td>444</td>
</tr>
<tr>
<td>¹¹B₄C</td>
<td>1.170</td>
<td>1.186</td>
<td>31.2</td>
<td>3.25</td>
<td>54.4</td>
<td>573</td>
</tr>
<tr>
<td>ZrC</td>
<td>1.168</td>
<td>1.271</td>
<td>29.5</td>
<td>2.90</td>
<td>56.8</td>
<td>934</td>
</tr>
</tbody>
</table>

4 Conclusions

The various results presented in this paper indicate that, from reactor physics considerations alone, it should be feasible to employ LWR plutonium fuels without uranium for achieving considerably more effective Pu-incineration than possible with MOX.

A homogeneous mixture of PuO₂ and a suitably chosen BP, or combination of BPs, could yield a relatively flat reactivity-vs.-burnup curve as LWR fuel and result in a total-Pu reduction of as much as ~ 1.1 kg/GWd as compared to < 0.4 kg/GWd for a 100% MOX core. In terms of fissile-Pu, the difference in possible reduction factors is much more striking. As regards the production of the minor actinides – Np, Am, Cm – this would be significantly lower, relative to the total-Pu reduction achieved.

Qualitative consideration of controllability and safety-related characteristics for an LWR core fueled with possible Pu-BP mixtures has been made by comparing reactivity coefficients and control absorber worths with corresponding values for reference UO₂ and MOX fuels. It has been found that individual BPs such as Rh and Ho, with which acceptable
values for some of the reactivity coefficients could be obtained, are associated with relatively large reactivity losses with burnup. On the other hand, BPs such as B or Eu – while enabling long core-residence times – would not yield sufficiently negative Doppler and/or void coefficient values. Acceptable solutions may be provided by using a combination of BPs, a 40% B - 60% Ho mixture having been suggested currently.

The influence of the inert matrix employed for a given Pu-BP fuel is, in general, small, but there could be significant effects on reactivity coefficients – on void coefficient characteristics for total voidage, in particular. Thus, not only material properties, but also such physics effects could determine the ultimate choice of inert matrix.

Control absorber worths and void coefficient values for Pu-BP fuels have been found to vary considerably with burnup. Clearly, realistic whole-reactor calculations are needed to evaluate features related to controllability and safety in a more quantitative manner.

REFERENCES

PHYSICAL AND TECHNOLOGICAL ASPECTS OF CERMET FUEL. APPLICATION OF Pu BURNING IN VVER REACTORS

V.M. DEKUSAR, A.G. KALASHNIKOV, E.N. KAPRANOVA, A.D. KARPIN, I.S. KURINA, V.V. POPOV
Institute of Physics and Power Engineering, Obninsk, Russian Federation

Abstract

The VVER-500 reactor was taken as an example to consider changes in light-water reactor neutronics and fuel cycle features when transferring to plutonium (weapon-grade composition) cermet fuel in all core assemblies. Technology problems of fuel fabrication are discussed.

INTRODUCTION

Nowadays the mixed oxide fuel (MOX-fuel) is used for plutonium utilization in PWR reactors. There is only about 5% plutonium oxide in light-water reactor fuel composition, the rest is depleted uranium oxide. Because of this reason the intensive process of uranium-238 conversion into plutonium-239 takes place simultaneously with the plutonium isotope transmutation within the reactor run. It reduces plutonium burn-out efficiency and stimulates the interest in consideration of opportunity to use fuel with lower amount of uranium-238.

One such fuel may be the cermet fuel as plutonium oxide and uranium oxide mixture dispersion in zirconium matrix or plutonium oxide and magnesium oxide mixture dispersion in zirconium. In the last case the fertile material is absolutely absent, which gives an opportunity to achieve the most high plutonium burn-up.

The cermet fuel composition has the number of advantages as compared with oxide one in the problems of reactor unit safety under the normal operational conditions and in the event of of design-basic accidents with the main piping rapture.

It should be noted also that cermet fuel use allows to carry out neutronics parameters optimization without any changes of fuel rods or fuel assembly design.

1. TECHNOLOGY OF CERMET FUEL MANUFACTURE

The utilization of cermet fuel (that is sintered UO₂ particles in a Zr-based allow matrix) in VVER reactors (water moderated water cooled power reactor) enables the effective settling of several problems, making it possible:

1. To ensure the indestructibility of cermet fuel elements under designed coolant-loss accident circumstances owing to fuel element cladding temperature reduction to 500°C during in the accident (including the maximum designed accident) and owing to the absence of gas pressure under the cladding;

2. To ensure the possibility of reactor exploitation in the dynamic regime without fuel elements cladding to be damaged due to corrosional cracking under stress owing to absence of contact of agressive fission products with the cladding;

3. To keep within the fuel composition practically all fission products in case of seal or cladding failure.
Conception of the cermet fuels utilization in VVER reactors has been discussed in detail in the work [1].

The above specified advantages of cermet fuels as well as higher effectivity of plutonium burn-out in a cermet fuel element made it possible to begin in our Institute of Physics and Power Engineering, being now the State Scientific Centre, some technological work on creation of cermet fuels and cermet fuel elements. To settle the problem of cermet fuel utilization in reactors intended for plutonium burning-out, the technology

![Flowchart](image)

**Figure 1.**
Table I The neutronics and fuel cycle parameters for the VVER-500 reactor with various fuel types.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Reactor version in term of fuel</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>UO$_2$</td>
</tr>
<tr>
<td>1. Power, MWt(e)/MWt(t)</td>
<td>640/1800</td>
</tr>
<tr>
<td>2. Load factor</td>
<td>0.8</td>
</tr>
<tr>
<td>3. Refuelling factor</td>
<td>5.43</td>
</tr>
<tr>
<td>4. Material flow in unclosed cycle</td>
<td></td>
</tr>
<tr>
<td>- natural uranium, t/year</td>
<td>98.5</td>
</tr>
<tr>
<td>- weapon-grade plutonium, kg/year</td>
<td>-</td>
</tr>
<tr>
<td>5. Enrichment with fissile isotopes, %</td>
<td>3.8</td>
</tr>
<tr>
<td>6. Burn-up, MWt*day/kg</td>
<td>45.3</td>
</tr>
<tr>
<td>7. Average conversion ratio</td>
<td>0.56</td>
</tr>
<tr>
<td>8. Annual plutonium unloading, kg/%</td>
<td>113.4</td>
</tr>
<tr>
<td>9. Spent fuel radiotoxicity increase *) as compared to make-up one</td>
<td></td>
</tr>
<tr>
<td>- only of plutonium</td>
<td>-</td>
</tr>
<tr>
<td>- of plutonium, minor actinides and decay products</td>
<td>-</td>
</tr>
<tr>
<td>10. Potential biological hazard, m$^3$H$_2$O</td>
<td></td>
</tr>
<tr>
<td></td>
<td>5.1*10$^{11}$</td>
</tr>
<tr>
<td>11. Reactivity change, total/margin for the lifetime, %</td>
<td>16.2/8.0</td>
</tr>
<tr>
<td>12. Radial power peaking factor BOL/EOL</td>
<td>1.38/1.31</td>
</tr>
<tr>
<td>13. Temperature reactivity worth coefficients in the BOL, 10$^{-4}$i/K</td>
<td></td>
</tr>
<tr>
<td>- of the coolant, cold</td>
<td>0.45</td>
</tr>
<tr>
<td>MPUM</td>
<td>0.14</td>
</tr>
<tr>
<td>at-power</td>
<td>-2.66</td>
</tr>
<tr>
<td>- of the fuel, cold</td>
<td>-0.27</td>
</tr>
<tr>
<td>MPUM</td>
<td>-0.24</td>
</tr>
<tr>
<td>at-power</td>
<td>-0.18</td>
</tr>
<tr>
<td>14. Relative absorber rod efficiency (BOL)</td>
<td>1.00</td>
</tr>
<tr>
<td>15. Effective delayed neutron fraction, 10$^{-3}$, BOL/EOL</td>
<td>6.22/5.86</td>
</tr>
<tr>
<td>16. Isotopic composition of unloading plutonium, %</td>
<td></td>
</tr>
<tr>
<td>Pu-238</td>
<td>2.0</td>
</tr>
<tr>
<td>Pu-239</td>
<td>48.8</td>
</tr>
<tr>
<td>Pu-240</td>
<td>27.3</td>
</tr>
<tr>
<td>Pu-241</td>
<td>13.8</td>
</tr>
<tr>
<td>Pu-242</td>
<td>8.0</td>
</tr>
</tbody>
</table>

*) After 10 years storage.

of manufacture of the following two types of cermet fuel compositions is being now worked out:

a) a composition containing depleted or natural uranium as a fertile material and including in it plutonium dioxide and uranium dioxide (MOX) in a Zr alloy-based matrix (PuO$_2$ - UO$_2$ + Zr alloy);
b) a composition containing no fertile material and including plutonium dioxide and magnesium oxide in a zirconium-based matrix (MgO - PuO₂ + Zr alloy).

The volume ratio of zirconium alloy in the both cases is about 50 per cent, and the volume ratio of plutonium dioxide that is needed to maintain the power output of a reactor is 2.8 and 3.5 per cent, respectively.

Fuel of both kinds in powder form can be obtained by the ammonium precipitation from nitrate solution with subsequent calcination and reduction. From these powders, by means of the slip casting method, for example, spheroidal particles can be obtained thereafter subjected to sintering.

The coating of the matrix material can be deposited on spheroidal particles by the ionplasma spraying method, for example. Coated spheroidal particles are charged then into thin-wall zirconium shells and impregnated with molten Zr-base alloy.

Fuel rods obtained in this way go then to fuel element fabricating.

The flow diagram of the cermet fuel manufacturing process is presented in fig.1.

Cermet fuels obtained in this process possess high thermal conductivity (λ is higher that 10 Watt per grade per centimetre) and good dissolution in the nitric acid, that is very important in case of spent fuel element processing.

2. THE NEUTRONICS AND FUEL CYCLE FEATURES
FOR THE REACTOR WITH VARIOUS FUEL TYPES

2.1. THE REACTOR VERSION WITH THE MIXED FUEL

Table I shows the calculation results of fuel cycle parameters and neutronics ones for the VVER-500 reactor with MOX fuel and cermet one loaded in all core assemblies. This reactor version is comparing with the conventional (UO₂ fuelled) reactor. The reactor versions with MOX fuel and cermet fuel are based on the weapon-grade plutonium. The isotopic composition of weapon-grade plutonium was chosen as follows [2] :

\[
\frac{^{238}\text{Pu}}{^{239}\text{Pu}}/\frac{^{240}\text{Pu}}{^{241}\text{Pu}}/\frac{^{242}\text{Pu}}{\_} = 0.93/0.34/0.04/0.58/0.04
\]

(in weight percent). The zirconium content in cermet fuel is equal 50 volume percent. The plutonium content in MOX fuel and cermet one was chosen to preserve total power output.

The calculation results allow us to make the following conclusions.

1. The greatest amount of plutonium (260kg or 62.4% of its initial loading) is burnt-out in the reactor with cermet fuel. The same values for MOX fuel and cermet one was chosen to preserve total power output.

2. In the reactor with cermet fuel temperature reactivity worth coefficients in terms of the coolant temperature at the minimum power level under monitoring and above it are close to that in the uranium fuelled reactor. The control and safety rod system efficiency is also close to that in the uranium fuelled reactor. These circumstances simplify the reactor safety maintenance with substitution of uranium fuel with plutonium one.

2.2. REACTOR WITH THE FUEL WITHOUT ANY FERTILE MATERIALS

To evaluate the maximum feasibility of light-water reactors for plutonium burn-out consideration was given to the reactor with cermet fuel...
In form of plutonium and magnesium oxides mixture in the zirconium matrix (weapon-grade plutonium). The calculations showed that when using such fuel composition without $^{238}\text{U}$ it is really possibly to achieve the plutonium burn-up fraction more than 80%. However here serious difficulties are encountered. They are as the following.

1. The deficit of the fertile materials causes reactivity change within the lifetime to be increased approximately twice as compared to that in the traditional VVER reactor. It requires early in the run a corresponding increase of the boric acid concentration in water. This causes a positive temperature reactivity worth coefficient of water in process of the reactor heating-up and even when the reactor is at-power. This is intolerable for the reactor safety reasons. It is useful to increase refuelling factor in order to decrease margin reactivity for the lifetime.

2. It is difficult to provide the necessary power density flattening due to a high fuel burn-up.

The required reduction of boron concentration in water and power distribution flattening can be provided by the rods with burnable poisons (RBP). However then they must be installed not only into made-up fuel assemblies, but also into assemblies being in operation for several years i.e., there should be the possibility of replacing RBP during refuelling.

3. The absolute value of Doppler temperature reactivity worth coefficient decreases very significantly. For the first loading in the beginning of the run its value is equal to $-0.2\times10^{-6}$ 1/°C. To increase this coefficient use of special absorbers is required.

4. The most considerable reduction of the effective delay-neutron fraction value (this fraction is $2.5\times10^{-3}$ in the reactor with the initial loading) takes place in this reactor.

CONCLUSION

1. The use of cermet fuel instead of MOX one allows:
   - to increase plutonium consumption by 20%,
   - to decrease summary radiotoxicity of plutonium and minor actinides in discharged fuel by 30%.

2. Temperature reactivity worth coefficients in terms of the coolant temperature and the control and safety rod system efficiencies are close to those in the uranium-fuelled reactor. This simplifies the reactor safety maintenance with substitution of uranium fuel with plutonium one.

3. The use of cermet fuel without any fertile materials results in considerable neutronics parameter changes for the worse. However the reactor with such fuel deserves a careful study as one that allows to provide for maximum plutonium burn-out.

REFERENCES


[2]. GILLET T.C., DENNING R.S., RODIHALG J.L. Shielding calculation techniques for the design of plutonium proceeding facilities. Nuclear Technology. 31(2), (1976) 244.
FUEL WITH CARBON MATRIX FOR BURNING PLUTONIUM IN THERMAL REACTORS

E.A. IVANOV, G.N. KAZANTSEV, I.Y. OvCHINNIKOV,
F.P. RASKACH
SSC IPPE, Obninsk,
Russian Federation

Abstract

It is offered to use the matrix fuel for effective plutonium burning in the thermal pressurized water reactor. The fuel with carbon matrix is plutonium (or another heavy metal) dioxide into manufacturing porous graphite block. This fuel composition being applied in the thermal pressurized water reactor leads to the neutron spectrum shift and, therefore, changes the temperature reactivity coefficients and efficiency of burnable absorbers. Such technology allow us to place into the fuel rod only plutonium without uranium and thorium. The technology of reprocessing and recycling of such fuel is very simple. There are not the facility, which used in this technological chain that once can be used in the double purposes.

INTRODUCTION.

We offer for solution of the plutonium utilization problem to apply the matrix composition as fuel. The fuel with carbon matrix is plutonium dioxide which is placed into manufacturing porous graphite block. Such fuel we suggest to use into the fuel rods in the thermal pressurized water reactor as VVER-type.

The useful of carbon matrix is that one can be reprocessed very easily. Technological properties of such matrix allow us to avoid of high chemical reprocessing and isotopic separating. Any of elements in this technological chain (reactor, fuel reprocessing, production of the fuel rods etc.) can't be used in the plutonium production purpose. Thus, this technology can't have double purpose.

Fuel rods are manufactured with graphite matrix have not gap between fuel block and cladding. The thermal conductivity of the matrix more higher then of ceramic one. Hence, the operating condition of plutonium dioxide particles in such fuel is more easy then in conventional fuel.

There are some variants to produce ceramic fuel into carbon matrix. We known about two of them. The first is developed in the ASPRIIM (the author G.G.Bayburin). The second is developed in the IPPE (the authors are I.Ya.Ovchinnikov and G.N.Kazantsev etc.).

From the neutronic physics point of view this fuel has some benefits too. Graphite matrix being installed into thermal reactor fuel gives new neutronic properties of burnable absorbers. And the graphite is solid moderator, and
varying of it's concentration we can change the reactivity feedback coefficients.

1. TECHNOLOGY.

There are two technologies of production of fuel with graphite matrix, which can be used in the thermal reactors for the plutonium burning.

One of them was developed by IPPE's specialists. The initial components of such matrix fuel is manufacturing graphite and ceramic particles (uranium or plutonium dioxide etc.). The more convenient size of dioxide particles is nearly 0.6-0.8 mm, but the permissible one may be another. The specialists in the IPPE developed the technology of production of the cristallic plutonium dioxide particles through electrolization of molted heavy oxides or heavy metals.

The fuel being manufacturing by such procedure has unizotropic physical properties (thermal and electrical resistance, strength) and it is experimentally obtained that there is unizotropic shift under irradiation.

Thermal conductivity in this fuel about by six times higher then one in the ceramic fuel.

The reprocessing of such fuel have to be very simple. After removing of the cladding the reprocessing may be carried out by milling of matrix and by following separating of carbon and heavy dioxide particles into gaseous stream or into liquid with specific weight about 3-5 kg/L. In opposite to the high chemical reprocessing the main procedure reduces to simple mechanical separation.

Low strength of carbon matrix make available to except the gap between fuel block and cladding in fuel rods. Therefore the thermal conductance between hey may be increase. This fact and relatively high thermal conductivity of carbon matrix ensure more lower inner fuel temperature then one in the conventional VVERs.

The using of such technology allow us to produce fuel pins without uranium and thorium, and to place more easily into fuel absorbers and another transuranium elements. There are some problem if reprocessed thorium dioxide. The reprocessing of irradiated thorium dioxide, been used in such fuel is more easy.

There is technology, which is developed in ASPRIIM (G.G.Bayburin).

This technology is based on non powder procedure. The initial components here are porous manufactured graphite block and liquid substation. And the main process here is impregnation of graphite block by liquid.

The reprocessing of such fuel also very simple. This fuel has isotropic properties and also has high thermal conductivity.

This is high chemical technology but it can also be very preferable. Using one we can vary the heavy metal density into fuel, which should be very useful for thermal reactor with high burn-up fuel management scheme.
Both of technologies (from the IPPE and from ASPRIIM) can be used without significant restrictions for manufacturing of the fuel rods for the thermal reactors. They are similar from the neutronic physics point of view. And being applied in the thermal reactor they haven't double purpose (to burning and to accumulating of the plutonium). Both of them allow us to create fuel composition with plutonium without fertile material. Both of them allow us to vary the concentration of heavy component up to 50%.

2. NEUTRONIC PHYSICS.

Graphite being install into the pressurized water reactor's core is solid moderator and it also has influence on spectrum of slowing-down neutrons. Due to the neutron spectrum shift the properties of integral fuel burnable absorber are also changed. Thus, for uranium fuel in VVER the burning rate of integral fuel gadolinium become lower and become almost equal to one of the fissile material.

Changing ratios between the water and the fuel concentrations and installing solid moderator, we change the value of void coolant reactivity feed-buck coefficient. Everywhere it's possible to adjust such parameters of burnable absorbers so that fuel temperature reactivity feedback coefficients also are negative and the reactivity margin is very small [1].

In the every core loading variant being considered later the reactivity coefficients are negative and small on absolute values.

Some problems may occur if plutonium dioxide is used without fertile materials (thorium in our case). This problem are also solved by choosing of resonance absorber (such as rhodium, rhenium and wolfram etc.).

The numerical calculation shown that positive contributions in temperature reactivity coefficients are brought by fissile materials (U-233, U-235, Pu-239, Pu-241) and some fission products (Xe-135, Sm-149). The negative contribution - by non fissionable heavy isotopes (U-238, Th-232, Pu-240 and Pu-242), and by Rh-103 and by other resonance absorbers (W, Re etc.).

Without fissile material breeding the reactivity bias should be too big, and only burnable absorber can compensate its satisfactorily. In the binary moderated systems with high enrichment by fissile isotope the negativity of the void coolant reactivity coefficients is determined by concentration and properties of resonance absorbers.

It's a reason to use the rhodium as burnable material with or without boron.

The variants of core are considered later in this report are not optimized. We want to show that application of the carbon matrix to dilute the fuel into rod may be useful for solving some problems of the plutonium burning. Thus, examples of the reactor core characteristics demonstrates only principal possibility of the using of the fuel based upon carbon technology. And it's possible that parameters of reactor cores will be further improved.
3. REACTOR CORE CHARACTERISTICS.

We considered some variants of the nuclear reactors like VVER-type with the plutonium fuel into carbon matrix. Design of reactor and parameters of coolant don't be changed. In our investigations we modified only number of fuel assembly and number of fuel rods into assembly. The outer diameter of fuel rod is non modified, cladding material - Zircalloy. Height of the core and conception of control bodies are as they in the VVER. Maintaining the coolant characteristics (pressure, temperature, drop in pressure and difference between inner or outer coolant temperatures) without changing we define the special power and temperature distribution into fuel rods and assemblies. To save hole power we must increase the outer core radius if decreasing water part in lattice (i.e. rod-to-rod step). And if rate of plutonium burning is fixed the permissible volume part of plutonium into fuel rod must be decreasing with decreasing of rod-to-rod step. Therefore the system can be critical only up to certain relative step of lattice, which is defined by the plutonium burning rate.

The fuel after irradiation isn't been able to use in military purposes. And the uranium, which produced in this reactor contains nearly 20% of U-234 and a lot of U-232 (about 900 ppm). Thus, after even the first cycle this fuel can't be interested for terrorists.

We considered such reactors cell which can be used as a partial loading in the conventional VVER-type reactors. There are some limits on containing of fissile material fraction into fuel rod. Some limits are connected with the strength and reliability of the fuel rod, and they are well-known and haven't some difference from one for conventional reactors. And there are some limits, which typical only for plutonium transmitters and burners. We should to increase (if it possible) the specific rate of the plutonium burning, and also the part of the plutonium at the end of cycle have to be small.

In the first, we considered VVER-type core with the electric power 1000MW, which fueled only weapon-grade plutonium into the graphite matrix. The fuel free from fissile material have positive temperature reactivity coefficients. To overcome this we use rhodium into carbon matrixes. Numerical calculations gives that in all variants the reactivity margin is about 2-5% and the reactivity coefficients: upon fuel temperature is about -0.01 -0.02 pcm/K on the low level and -0.05 -.1 pcm/K on the full power; and upon coolant temperature (including changing of the density) is about -0.1 -0.15 pcm/K and -0.2 -0.3 pcm/K relatively [1]. Really, we can adjust absorbers so that reactivity feedback will be more negative, but it isn't necessary. Parameters of plutonium reactors are presented in table 1.

We considered the core operating without partial reloading. Here fuel after one year long irradiation is removed and reprocessed. Equilibrium cycle means that fuel after reprocessing is mixed with fresh weapon-grade
Table 1. The main parameters of the core with plutonium loading without fertile materials. Equilibrium cycle.

<table>
<thead>
<tr>
<th></th>
<th>1</th>
<th>2</th>
</tr>
</thead>
<tbody>
<tr>
<td>the core radius, m</td>
<td>1.556</td>
<td>1.311</td>
</tr>
<tr>
<td>the number of the fuel assemblies</td>
<td>198</td>
<td>146</td>
</tr>
<tr>
<td>the assembly dimension &quot;on flat&quot;, cm</td>
<td>24.36</td>
<td>22.89</td>
</tr>
<tr>
<td>Number of rods in the assembly</td>
<td>397</td>
<td>331</td>
</tr>
<tr>
<td>lifetime, eff. days</td>
<td>350</td>
<td>350</td>
</tr>
<tr>
<td>the electric power, MW</td>
<td>1000</td>
<td>1000</td>
</tr>
<tr>
<td>absorbers</td>
<td>Rh+Gd</td>
<td>Rh+Gd</td>
</tr>
<tr>
<td>material balance per year</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pu-239, kg</td>
<td>1096.84</td>
<td>705.06</td>
</tr>
<tr>
<td>Pu-240, kg</td>
<td>203.85</td>
<td>45.29</td>
</tr>
<tr>
<td>Pu-241, kg</td>
<td>279.87</td>
<td>4.529</td>
</tr>
<tr>
<td>Pu-242, kg</td>
<td>239.44</td>
<td>0.00</td>
</tr>
</tbody>
</table>

plutonium and loaded into the core. In calculation model in the fuel reloading into the core we assumed that all gaseous left from fuel and time between reprocessing is three years.

It's shown that instead of equilibrium compositions are different but values of the plutonium consuming in this reactor are equal. It means that choice of the core parameters should be based upon the reactor safety criteria.

Table 2. The main characteristics of the core loaded by Pu and Th mixture. Equilibrium cycle.

<table>
<thead>
<tr>
<th></th>
<th>1</th>
<th>2</th>
<th>3</th>
</tr>
</thead>
<tbody>
<tr>
<td>the core radius, m</td>
<td>1.556</td>
<td>1.311</td>
<td>1.147</td>
</tr>
<tr>
<td>the number of the fuel assemblies</td>
<td>198</td>
<td>146</td>
<td>120</td>
</tr>
<tr>
<td>the fuel assembly dimension &quot;on flat&quot;, cm</td>
<td>24.36</td>
<td>23.89</td>
<td>23.10</td>
</tr>
<tr>
<td>the number of rods</td>
<td>397</td>
<td>331</td>
<td>271</td>
</tr>
<tr>
<td>the electric power, MW</td>
<td>1000</td>
<td>1000</td>
<td>1000</td>
</tr>
<tr>
<td>the time between reloading, eff. days</td>
<td>300</td>
<td>300</td>
<td>300</td>
</tr>
<tr>
<td>type of reload</td>
<td>1/3 in-out-in</td>
<td>1/3 in-out-in</td>
<td>1/3 in-out-in</td>
</tr>
<tr>
<td>material balance per year</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Th-232, kg</td>
<td>1936</td>
<td>1845</td>
<td>1936</td>
</tr>
<tr>
<td>U-233, kg</td>
<td>60.34</td>
<td>0.00</td>
<td>59.06</td>
</tr>
<tr>
<td>U-234, kg</td>
<td>12.82</td>
<td>0.00</td>
<td>12.02</td>
</tr>
<tr>
<td>Pu-239, kg</td>
<td>379.2</td>
<td>66.9</td>
<td>312.3</td>
</tr>
<tr>
<td>Pu-240, kg</td>
<td>43.32</td>
<td>23.26</td>
<td>20.06</td>
</tr>
<tr>
<td>Pu-241, kg</td>
<td>61.44</td>
<td>59.44</td>
<td>2.006</td>
</tr>
<tr>
<td>Pu-242, kg</td>
<td>32.24</td>
<td>32.24</td>
<td>0.00</td>
</tr>
</tbody>
</table>
Later we considered the same reactor but with the mixed plutonium-thorium loading [1]. At the table 2 the main parameters of such reactors and balances of the fuels materials are demonstrated. The lifetime of such cores was choose very shot - only from methodical consideration. As mentioned above, the optimization of the technical characteristics of such cores hasn't been an our aim.

As in the case with free plutonium here isn't obtained the strong dependent of material balance from the type of reactor cells. Thus, the such core may be optimizing upon safety and any another criteria too.

We considered the problem only of the weapon-grade plutonium burning. And in the main we tried to minimize the plutonium concentration in the end of cycle. In this and in the other case we supplied that fuel is used without chemical and isotopic separation of heavy metals once from another. Thus, uranium in the variant with thorium have equilibrium concentration.

CONCLUSION

There are two variants of carbon matrix fuel technology. Both of them are useful to produce fuel rods.

The fuel based on carbon matrix have some benefits.

In this fuel it's possible to load the plutonium dioxide and combined it with any heavy dioxide (thorium in our case, and may be uranium and some minor actinides). Operational conditions for ceramic particles will more easy then in conventional fuel. Such fuel rod has more lower inner temperature then ceramic one.

The reprocessing of such fuel is very simple.

Matrix composition supply reliable containment of gaseous fission products. The temperature of fuel particles relatively low and, hence, gaseous will contain by heavy ceramic itself. If gaseous will leave from fuel particles they'll kept by pores in matrix.

There are some other benefits of the such fuel. Graphite matrix being installed into thermal reactor fuel gives new neutronic properties of burnable absorbers. In addition this, graphite is solid moderator, and varying of it's concentration we can change the reactivity feedback coefficients. In such fuel there are more of variable parameters, i.e. that parameters of this system more flexible and, hence, the optimization of the core characteristics with such fuel is simplified.

And, it's very important that the every facilities from the technology chain, which is present above can't be applied for accumulation of the weapon-grade nuclides. The all stages of process will be reliable protected against terrorism.
REFERENCE

SESSION 4: MOLTEN SALT
The incineration of minor actinides is a nonsense if the problem of plutonium, that is the most important of the actinides, is not resolved.

The concept analysed in the present paper is an extension of the ones developed for minor actinide burning. The most promising hybrid system concept considers fuel and target both as liquids.

From the results obtained, it is possible to discard solid and sodium targets. The solutions adopting composite target seem, at present stage, the most promising, but still remains the problem of the Pu production at a level not acceptable in a Pu burning system.

Due to the necessity of electrical energy production, the blanket using plutonium dissolved in molten salt permits to reach the higher thermal efficiency.

1. INTRODUCTION

A hybrid system is composed of a particles accelerator, a target, for neutron production, a blanket, for transmutation or fissile material production, and their respective interfaces (i.e. the window between accelerator and target).

These systems were studied in the past for fissile material production. In the last years these ideas were reconsidered for the minor actinide (MA) and long-lived fission product destruction as alternative to the traditional final disposal of nuclear waste.

As plutonium is the most important of the actinides (about 95% as mass), the incineration of minor actinides is a nonsense if not resolved the plutonium problem.

Plutonium is an excellent nuclear fuel, the most convenient solution should be the burning in a nuclear reactor to produce electrical energy. But in the case of very big stockpiling, it seems interesting to burn a part in less efficient, but less time consuming systems, such as thermal hybrid systems as demonstrated by the Academy of Science of the United States.

The concept analysed in the present paper is an extension of the ones developed for MA burning.

The most promising hybrid system concept considers both fuel and target as liquids.

2. PLUTONIUM

In the present work, plutonium means all the actinides present in the discharged PWR standard spent fuel except the uranium isotopes. Standard PWR spent fuel is defined as UO₂ fuel enriched at 3.3% irradiated at 33000 MWd/t
3. TARGET

The target can be solid or liquid. In the first case, it is possible to consider two accident scenarios: loss of coolant and loss of beam. In the first case if the beam is not switched off quite soon the target will melt, in the second the fast quenching can provoke a strong thermal shock.

The molecular dynamic calculations seem to demonstrate that long-range order disappears in the areas travelled through by very energetic particles; this means that in these zones the material of the target becomes glass. This result should be confirmed by the behaviour of the tungsten window at Paul Scherrer Institute (PSI) [1]; which, after a thermal shock is broken into very small parts, practically like powder.

The previous considerations seem to indicate the convenience of liquid targets.

In the case of a heavy metal flowing target, the protons do not penetrate very much and the majority of the neutrons are produced in the first 20 cm of the target. The most interesting solution to flatten the neutron flux shape is to use composite targets: a low mass number liquid, such as lithium or sodium (primary target), surrounded by a secondary target made of heavy metal (such as lead or uranium).

A set of calculation for pure materials was carried out considering uranium, lead, tin, sodium and beryllium. Uranium targets produce some amount of plutonium as spallation product; lead produces the long-lived $^{206}$Pb; low mass number targets have low neutron yield and very high fraction of neutron are in high energy rage ($E > 20$ MeV). Due to the low neutron yield, it is possible to discard sodium as spallation target.

A second set of calculation was performed for composite targets, U-Be and Pb-Be, with the central beryllium zone at different radii. Low energy neutron ($E < 20$ MeV) distribution in the target is significantly improved. The neutron yield is drastically reduced and the production of Pu isotopes remain high in the case of uranium targets. The neutron yield for U-Be target with 2 cm diameter for the beryllium zone (22.94) is quite similar to the yield corresponding to lead (23.35).

A third set of calculations was performed a target containing the fuel. That is plutonium dissolved in $^7$LiF. The same Pu concentrations of the fuel 10, 50, 100 g/l were considered. The maximum neutron yield achieved is 2.34 at 100 g/l; this means that direct spallation on fuel is not convenient.

Taking into account all the parameter considered the most convenient solution to use lead target.

4. BLANKET

For a neutron flux greater than $1.0 \times 10^{15} \text{n}/(\text{cm}^2 \text{s})$ it is impossible to use the traditional system: solid fuel and cooling water. In fact some blanket proposals consider liquid fuel: actinide dissolved or dispersed in heavy water. The most recent blanket concepts are based on plutonium or minor actinides dissolved in molten salts, particularly $^7$LiF - $^9$BeF, used in the Molten Salt Breeder Reactor Experience at the Oak Ridge National Laboratory.

A series of MCNP cell calculations were carried out considering the Pu at the above mentioned concentrations and:

1. dissolved in heavy water and moderated by heavy water;
2. dissolved in $^7$LiF and moderated by heavy water;
3. dissolved in $^7$LiF and moderated by graphite.
The highest $K_{inf}$ values correspond to the case of Pu moderated and dissolved in $D_2O$; the minimum one for Pu dissolved in $^7LiF$ and moderated by graphite.

In any case the minimum $K_{inf}$ value is 1.45550; this means that the three sets of solvent-moderator studied are equivalent.

The outlet temperature of the molten salts can reach values of about 640 °C without increasing the pressure, as comparison for $D_2O$ we have 300 °C at 10 MPa. The efficiency of the Thermal Cycle for molten salt can reach 42%, against 33% for $D_2O$.

The increase of efficiency means that the same amount of electrical energy is produced reducing the mass of fission products, particularly the long-lived ones.

In [2] is analysed another important advantage of the molten salt blanket that is the reprocessing.

Hybrid systems having lead target with an effective radius of the fissile zone of 250 cm and a height of 500 cm reach the criticality for a Pu concentration of 7 g/l and the corresponding neutron flux is $1.1 \times 10^{15}$ n/(cm² s). With that characteristics preliminary calculations demonstrate that it is possible to burn about 552 kg of Pu per year.

5. CONCLUSION

From the results obtained, it is possible to discard solid and sodium targets. The solutions adopting composite target seem, at present stage, the most promising, but still remains the problem of the Pu production at a level not acceptable in a Pu burning system.

Due to the necessity of electrical energy production, the blanket using plutonium dissolved in molten salt permits to reach the higher thermal efficiency.

The hybrid concept considered results suitable for plutonium burning.

REFERENCES


Plutonium should not be simply burned out and be used to produce a new fissile $^{233}$U, because for solving the global energy/environmental problems in the next century a huge-size Fission Industry should be established preparing huge fissile materials. It seems to be established by THORIMS-NES [Thorium Molten-Salt Nuclear Energy Synergetics] composed of simple Molten-Salt Fission Reactors [MSR: FUJI-series] and $^{233}$U producing Accelerator Molten-Salt Breeders [AMSB]. These new Th facilities will be developed by utilizing/eliminating the "excess Pu" separated from Pu weapon-heads and the spent-fuel of U-Pu reactors. A semi-real scenario for proceeding such work in the first half of the next century is presented. THORIMS-NES will have big advantages not only in safety and radiowaste issues, but also in nuclear-proliferation/terrorism and economy guaranteeing low R & D cost.

1. INTRODUCTION

In the Pu disposition issue, our targets in final stage should be put on:
(A) nearly complete elimination of the world's Pu stocks [1],
in parallel achieving the followings:
(B) economical utilization of energetic potentiality of Pu (and TRU) as an useful producer of neutron or fissile materials, not simply burning out. (C) real establishment of new/rational huge-size nuclear industry to solve the global energy/environmental problems in the next century.

The real meaning of the target (C) will be explained in detail in Chapter 2. To establish this (C), the target (B) should be proceeded in the best way.

Such an idealistic Pu disposition work might be realized by the modification of our Th-breeding synergetic system: THORIMS-NES ("Thorium Molten-Salt Nuclear Energy Synergetics [2], which is composed of [I] simple fission power stations [Molten-Salt Reactor (MSR): FUJI-series], [II] fissile-fuel producers by spallation/fission reactions of 1 GeV-proton Accelerator Molten-Salt Breeder: AMSB, and [III] Dry process plants.

Detailed explanation of this new technology modifying the original THORIMS-NES in general has been given in another paper of this meeting by Lecocq et al. [3]. A brief explanation will be given in Chapter 3 too, in which especially a little detail of AMSB and A-plan will be mentioned.

Its real application in global strategy of next century will be discussed in Chapter 4.

2. FUTURE NUCLEAR ENERGY DEMAND

The next 21st century will be a transient period from fossil fuel age to solar age through nuclear energy era, as a global and especially local climate could not accommodate the excess heat emission several times bigger than the present artificial heat generation. Therefore, the heat-emission type energy technologies (even nuclear fusion or satellite electric-generation) could not be utilized as major ones in the 22nd century.

Such advanced prediction will be understood from the illustrations in Fig.1, basically depending and extending Marchetti’s prediction on the future energy [4, 2, 5]. If we tentatively accept the same global energy growth rate of 2.3%/year as the past, the necessary fission energy will be "1,000 - 2,000" TWe-Year in the next century. This is "500 - 1,000" times larger than the past (peaceful) fission energy production of only "2" TWe-Year. (In here, we have to recognize that even such huge nuclear energy will not be enough to solve the CO₂ Greenhouse effect as shown in Fig.1(C) [4]).

Such a huge total amount and a steep increase corresponding to about 10 years of doubling-time in the nuclear energy will not be achieved by the present U-Pu solid-fuel cycle system such as LWR, HTGR and LMFBR, due to several difficulties connected with (a) breeding ability, (b) safety [including severe accidents], (c) radio-wastes [including production of trans-Uranium elements], (d) nuclear-proliferation and -terrorism [including Plutonium-elimination], and (e) public and institutional acceptances related with the technological simplicity, flexibility and economy in the global applications (cf. the left column of Table 1 in reference [3]).

Therefore, we have to prepare an essentially new technology.
3. NEW PHILOSOPHY: THORIMS-NES
---THORIUM MOLTEN-SALT NUCLEAR ENERGY SYNERGETICS---

3.1. General principles

Already, to solve the several energy problems shown in Fig. 1, a new philosophy named as THORIMS-NES has been proposed to the public. It depends on the following three principles [2, 5].

[I] Thorium utilization,
[II] Application of molten-fluoride fuel technology,
[III] Separation of fissile producing breeders (process plants—AMSB: Accelerator Molten-Salt Breeder) and power generating reactors (utility facilities—MSR: Molten-Salt Reactor).

Fig. 1 Global Future Energy Prediction.

[(A) is a further extension of Marchetti's estimate of historical trend in energy substitution; (B) the prediction of yearly growth rate in the past primary energy consumption; (C) the CO₂ yearly emission from several fossil fuels; and (D) the nuclear fission-energy production based on (A) & (B).]
These facilities followed by chemical dry-process plants will be easily coupled each other preparing a simple "Molten-Salt Fuel Cycle" with a minimum handling and transportation. In this fuel-cycle several radio-waste species including TRU and F.P. will be destroyed gradually without any separation works [6].

3.2. Modification for Pu-burn and $^{233}$U-production

3.2.1. General approach

The modification of THORIMS-NES might be established following on the three plans:

1. **D-plan**: Pu (and Trans-Uranium elements [TRU]) fluoride separation by Dry-process from the spent solid-fuels accumulating in the world. The technological basis has been examined by French, Russian, etc..

2. **F-plan**: Pu-burning and $^{233}$U-production by MSR [FUJI-Pu]. The most rational safe technology for the effective Pu-incineration will be an application of MSR fertilized by Th. The detail of neutronic examination will be reported by Mitachi et al. in this meeting [7].

3. **A-plan**: Pu-burning and $^{233}$U-production by AMSB-Pu, in parallel with F-plan. AMSB-Pu was mostly examined and is being improved by the cooperation of Chigrinov's group, Minsk [8].

3.2.2. FUJI-Pu

A simple rational design of MSR named FUJI has been proposed including a fuel-self-sustaining type FUJI-II already [9], and will be explained including Pu-burning version: FUJI-Pu in ref. [7].

The significant advantage of MSR in burning dismantled weapons fuel has been excellently examined by ORNL peoples [10]. Soon after dismantling weapon heads of Pu (or $^{235}$U) can easily convert to fluoride diluting by other fluorides. This salt mixture itself is fairly resistive to nuclear-proliferation/diversion. If necessary, it will be improved more by spiking by high gamma active fluorides (including $^{60}$Co, $^{106}$Ru, $^{144}$Ce, or $^{238}$Pu) or by denaturing $^{235}$U by $^{238}$U.

3.2.3. AMSB-Pu

The basic idea of AMSB was invented on 1980 depending on the "single fluid type Molten-Salt target/blanket concept"[11], which is significantly simple and practical in structure and operation [Fig. 2]. The target/blanket vessel is a simple pot of 4.5m in diameter and 7m in depth. Proton beam will be injected in off-center position of molten-salt vortex. Therefore, several serious technological problems related with (i) material compatibility and radiation-damage, (ii) heat removal, (iii) spallation chemistry, and (iv) target shuffling (uniform continuous reaction) are mostly able to solve by this design concept except the proton-beam injection port engineering.

Depending on the initial developmental effort of MSR basic technology, A-plan developing AMSB-Pu will be proceeded in a few years delay from FUJI-Pu. A typical composition of target/blanket salts is

$$7\text{LiF-BeF}_2\text{-ThF}_4\text{-}^{233}\text{UF}_4\text{-}^{239}\text{PuF}_3 = 64-18-[18-(X+Y)]-X-Y \text{ mol%: \[X, Y = 0\sim0.7\]}$$
Fig. 2. Schematic set-up of single-fluid-type Accelerator Molten-Salt Breeder (AMSB). [system-size]: 4.5m in diameter, 6m in depth.


The most important items for R&D of AMSB-Pu will be the followings[12, 13, 6]:

(A) 1 GeV, 100-300 mA proton Linear Accelerator: In the target/blanket salt system a little lower voltage, 1 GeV, will be convenient, although it should be optimized in final design. Proton will penetrate into salt more than 1 m, where is a most intense reaction zone, and such situation is effective for heat removal.

As a long term program, more economical non-monochromatic beam accelerator development should be encouraged as an industrial machine than Linac.

(B) Injection port engineering: It is a most serious unclear item. However, the vapor of salt might be mostly condensable on duct wall (cf. Fig. 2), applying several additive techniques such as electrostatic collection. Gaseous species in molten salt should be carried away to be separated in outside of reactor core. Such practice might be solved by the real beam test increasing its intensity stepwise.
(C) Accuracy of neutronic calculations: The target/blanket salt system of AMSB contains several kinds of nuclei including light ones. Therefore, neutronic calculation is not easy and low accuracy in reaction products yield and in heat generation rate yet. Furukawa and Kato unsuccessfully aimed to proceed the experimental analysis of large target salt block getting the help of SIN (now PSI) group in 1981, but now again planning it under the cooperation with Russian group. It will be valuable for the development of spallation theorem in general, too.

(D) Reactor chemical aspects: Several chemical issues relating with "spallation chemistry" has been successfully examined [12, 13, 6] depending on chemical basis of MSR developed by ORNL. The chemical processing procedures of salt will be more flexible in our substantially subcritical system. The transmutation of hazardous radioisotopes could be proceeded by the minimum separation work simply circulating through target/fuel salt cycle in THORIMS-NES.

(E) System engineering design optimization: The size of target/blanket system is important relating with [a] radiation damage of reflector graphite and reactor vessel wall, [b] inventory of fissile, fertile and reaction products for resulting the most important optimal reactor performance, and finally [c] total economy.

Its optimization connecting with salt composition and the operation purpose of this facility should be improved depending on the progress of several technological items.

4. A SCENARIO OPENING THORIUM-ERA

   BY APPLYING PU-BURN AND $^{233}$U-PRODUCTION

4.1. Technological basis

To establish the targets (A), (B) and (C) mentioned in Chapter 1, the three plans, D-, F- and A-plan, have been proposed (cf. sec. 3.2.1.).

If we proceed the above plans to burn Pu and produce $^{233}$U, these efforts would fully contribute to prepare the real standard THORIMS-NES finally and automatically.

The conventional methods for Pu disposition are depending on solid-fuel concepts, which are not suitable for establishing the targets (A) (C). They are not fully eliminating Pu, or need repeated processing and fuel-refabrication or in a weak neutron economy. Such situations are illustrated in Fig. 3, in which advantages of the FUJI-Pu and AMSB-Pu are also shown [5].

The conservative performances of FUJI-II, FUJI-Pu and AMSB-Pu in Pu-burning/$^{233}$U-production are presented in Table I. In future, FUJI-II and FUJI-Pu should be understood that they would include 1GWe-class superFUJI or superFUJI-Pu of higher performance than those of Table I, and AMSB-Pu would also be able to improve more.

Table I. PRELIMINARY PERFORMANCE OF FUJI [PER 1GWe]

<table>
<thead>
<tr>
<th>Pu-inv.</th>
<th>$^{233}$U-inv.</th>
<th>Pu burn/Y</th>
<th>$^{233}$U prod.</th>
<th>Electric output</th>
</tr>
</thead>
<tbody>
<tr>
<td>FUJI-Pu</td>
<td>3 t</td>
<td>0.86 t</td>
<td>0.7 t</td>
<td>1 GWe</td>
</tr>
<tr>
<td>FUJI-11</td>
<td>2 t</td>
<td>self-sust.</td>
<td>1 GWe</td>
<td></td>
</tr>
<tr>
<td>AMSB-Pu</td>
<td>5 t</td>
<td>5 t</td>
<td>1.0 t</td>
<td>1 GWe</td>
</tr>
</tbody>
</table>

174
CONCEPTUAL PRESENTATION OF FISSILE-MATERIAL CHANGE IN SEVERAL PU-INCINERATION Reactors

Fig. 3. Conceptual Presentation of Fissile-Material Change in Several Pu-Incineration Reactors.
4.2. An example of scenario to open THORIMS-NES era

Proven U-Pu cycle systems would not realize the energy production predicted in Fig. 1(D) owing to the social, technological and economical reasons. However, THORIMS-NES would be able to realize the several global-energy scenario applying the above D-, F- and A-plans \[5, 11\]. Here, one of the simple examples has been shown in Table II and Fig. 4 \[cf.14\].

Table II. REPLACEMENT OF U-PU REACTORS BY TH-U REACTORS
ELIMINATING PLUTONIUM \[cf. Fig. 4\]

<table>
<thead>
<tr>
<th>Total Energy Stations</th>
<th>U-Pu Cumulative Pu</th>
<th>D-Plan Pu supp.</th>
<th>F-Plan FUJI-Pu:FUJI</th>
<th>A-Plan AMSB-Pu</th>
</tr>
</thead>
<tbody>
<tr>
<td>(GWe)</td>
<td>(GWe)</td>
<td>(ton)</td>
<td>(t/10Y)</td>
<td>(GWe) (fac./10Y)</td>
</tr>
<tr>
<td>2000</td>
<td>300</td>
<td>300</td>
<td>1,200</td>
<td></td>
</tr>
<tr>
<td>2010</td>
<td>550</td>
<td>550</td>
<td>2,550</td>
<td>start start</td>
</tr>
<tr>
<td>2020</td>
<td>1,000</td>
<td>850</td>
<td>4,650</td>
<td>730 100; 50</td>
</tr>
<tr>
<td>2030</td>
<td>1,850</td>
<td>1,000</td>
<td>7,425</td>
<td>4,890 200; 350</td>
</tr>
<tr>
<td>2040</td>
<td>3,460</td>
<td>1,050</td>
<td>10,525</td>
<td>4,860 0; 1,810</td>
</tr>
<tr>
<td>2050</td>
<td>6,800</td>
<td>600</td>
<td>12,825</td>
<td>2,700 0; 5,700</td>
</tr>
<tr>
<td>2060</td>
<td>9,300</td>
<td>100</td>
<td>13,875</td>
<td>580 0; 9,100</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>New Systems</th>
<th>Pu</th>
<th>233(^{U})</th>
<th>233(^{U})</th>
</tr>
</thead>
<tbody>
<tr>
<td>burn</td>
<td>produc. residual</td>
<td>(GWe)</td>
<td>(t/10Y)</td>
</tr>
<tr>
<td>2000</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2010</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2020</td>
<td>150</td>
<td>430</td>
<td>350</td>
</tr>
<tr>
<td>2030</td>
<td>850</td>
<td>3,090</td>
<td>2,550</td>
</tr>
<tr>
<td>2040</td>
<td>2,460</td>
<td>3,710</td>
<td>5,450</td>
</tr>
<tr>
<td>2050</td>
<td>6,200</td>
<td>3,450</td>
<td>5,750</td>
</tr>
<tr>
<td>2060</td>
<td>9,200</td>
<td>2,580</td>
<td>4,000</td>
</tr>
</tbody>
</table>

\[/10Y : cumulative value in 10 years till the date\]

Tentatively the total electric production of U-Pu fueled power stations in future will be assumed as 4 times larger in maximum than the present. Even so low this will still produce more than \(10^4\) ton Pu (assuming 300Kg/GWe-Ynet) till 2050. In here, we can apply the present Purex process plants, too, but it might not become significant due to the high capital/operation cost. As soon as possible, D-plan should be started to prepare more proliferation-resistant mode process, because a simple storage of spent-fuels will be non real solution. Separated Pu should be burned out soon by F- and A-plan.

In the stage before realizing D- and F-plan, dismantled weapon-heads Pu (and Pu from spent-fuels, too) should be converted to fluoride in the diversion protection form even not yet being decided the final MSR fuel-salt composition \[cf. sec. 3.2.2 \[10\].

Pu (TRU) disposition could be started from 2010 by F-plan, and from 2020 by A-plan in parallel. The former activity will become 200 GWe in maximum scale about
2030, burning about 2,600 ton Pu (TRU) or more. The latter will become 650 facilities in peak about 2040, burning about 10,000 ton Pu (TRU) or more.

The duty of FUJI-Pu will be finished until 2040. However, it can be operated as proper Th-233U power stations (FUJI) till the end of reactor life without any modification. The technological development of AMSB-Pu will be significant among 2020 and 2040. The initial AMSB-Pu will be in lower grade not producing any outer electricity. Afterward its new version will produce excess electricity improving in performance by near critical condition, which would allow an excess electric production of 1 or 2 GWe/facility [5].

After the middle of 2040's decade, in which Pu would be almost eliminated, AMSB-Pu should be gradually dismantled recovering 233U fissile, which is strongly effective for initiating a lot of FUJI power stations. Therefore, the main leading role of AMSB-Pu will be in the period of 30 - 40 years, although AMSB will be continuously useful for radio-waste (including minor actinides) incineration as a minor purpose [6].

5. DISCUSSIONS

5.1. ADDITIVE TECHNOLOGICAL RATIONALITY

5.1.1. CONTAINER MATERIALS: Fortunately, an easy manufactureable, weldable and high temperature resistant alloy compatible with molten fluorides has been developed basically depending on the effort of Dr. H. Inouye, ORNL. This alloy,
Hastelloy N [Ni-(15~18)Mo-(6~8)Cr in weight %], has been modified adding about 1% Nb for the protection of Te attack on surface [15]. This was experimentally supported by Kurchatov Institute [16].

This alloy doesn’t need to be introduced in reactor core region suffering any severe irradiation and thermal shock. [MSR core is occupied by graphite and fuel-salt only.] The most thin alloy part will be about half inch diameter tube in the intermediate heat exchanger. Therefore, the reactor design would be much simple and easy.

In practice, the corrosion of Hastelloy N was surprisingly small even in the small experimental reactor MSRE due to the negligible contamination by air and moisture. Larger commercial power stations would be much safer in corrosion.

5.1.2. FUJI-Pu: If we could get a cooperation with ORNL, the construction of pilot-plant: 7MWe-miniFUJI will be established less than 7 years (except licensing time) using reserved MSRE fuel-salt. The first FUJI-Pu might be operated among 15 years from now [2, 5].

5.1.3. AMSB-Pu: Its physical basis is simple and clear. After preparing 10 mA-1 GeV proton Linac, M.S. target/blanket experiments will be started in years later. Except the development of high current Linac, a beam-inject port engineering will be most important R & D item. However, the vapor of salt might be mostly condensable on duct wall, applying several additive techniques such as electrostatic collection. Gaseous species including T should be carried away by molten salt flow to be separated at outside of reactor core. [cf. sec. 3.2.3.]

5.2. SAFETY

MSR has essentially no severe accident, and subcritical AMSB is more safe. Therefore, engineering safety of THORIMS-NES is significantly high [2]. The other safety features will be shown by Lecocq et al. [3].

5.3. Anti nuclear-proliferation and -terrorism:

In THORIMS-NES, $^{232}$U content in fissile $^{233}$U will become more than 500 ppm [4]. Therefore, the very hard gamma (2.6 MeV) activity of daughter $^{208}$Tl in explosive 10 Kg $^{233}$U will emit nearly lethal dose of 200 rem/hr at 50 cm distance, and the protection shield will be required lead more than 20 cm in thick. It is the hardest and least desirable material for weapon, and its safeguard/detection is very easy.

Alternative Approach for diversion protection:

FUJI-Pu would be located in safe-guarded sites inside countries with extensive nuclear experience. Ordinary FUJI without Pu and without processing could be outside those states (the inside/outside concept).

If $^{232}$U content in $^{233}$U is comparatively low in early stage of this fuel cycle, isolated $^{233}$UF$_4$ would be instantaneously denatured to 12% $^{233}$U by natural U. This treatment will not produce any serious difficulties, because denatured $^{233}$U-fuel of FUJI is a fluoride mixture of $^{233}$U, $^{238}$U, $^{232}$Th in the composition of 0.2, (0.2 x 7 =)1.4 and 12 mol% respectively, which produces $^{239}$Pu about 10
times smaller than $^{233}\text{U}$. Especially in the case of fuel-self-sustaining FUJI, Pu content will become negligible among a few years. (Similar denatured $^{235}\text{U}$ version of FUJI is nominated as FUJI-Y and discussed already.)

5.4. Economy

Significant economical advantage of THORIMS-NES has been concluded as a result of preliminary comparative examination between superFUJI and LWR. The main reasons are mostly depending on (1) negligible fuel cost (only an addition of Th and small amount of $^{233}\text{U}$ in full life) comparing with the same order of magnitude as the construction cost in LWR, (2) simple reactor core structure without fuel assembly and fuel handling machine (core internals are only graphite and small number of control rods), (3) very high thermal efficiency more than 46% by ultra-supercritical steam turbine, (4) simple safety system because of the higher safety of MSR and smaller reactor building, etc. and (5) higher capacity factor because of no necessity of refueling and vessel-opening works. (6) In this fuel-cycle system nuclear material transportation is very few due to the continuous fuel burning, and a very few addition of fissile/fertile materials only.

AMSB will be much expensive than 1 GeV superFUJI. However, it will not be important, because its total activity in the scenario of Table 4 will be only about 15 TWe·Y, less than 2, or 1% of total 1,000 or 2,000 TWe·Y capacity.

5.3. R & D PROGRAM

Its technological basis of F-plan has been prepared by the excellent efforts of ORNL among 1947-76[15]. Therefore, the commercialization of FUJI-Pu in smaller size (100-300 MWe) will be performed in 15 years at least. Such public nuclear power stations should be simpler in configuration/operation/maintenance, and power-size flexible not like as Fission or Spallation Breeding power stations [MSBR or AMSB].

Depending on the above initial developmental effort of MSR basic technology, A-plan developing AMSB-Pu will be proceeded in about 10 years delay from FUJI-Pu. Its optimization connecting with salt composition and the operation purpose of this facility should be proceeded depending on the progress of several technological items.

The time schedule of developmental program has been shown in Table III.

Table III. DEVELOPMENTAL PROGRAM OF THORIMS-NES FOR PU DISPOSITION

<table>
<thead>
<tr>
<th></th>
<th>2000</th>
<th>2010</th>
<th>2020</th>
<th>2030</th>
</tr>
</thead>
<tbody>
<tr>
<td>miniFUJI-Pu</td>
<td>----</td>
<td>----</td>
<td>(FUJI)</td>
<td></td>
</tr>
<tr>
<td>FUJI-Pu</td>
<td>----</td>
<td>----</td>
<td>----</td>
<td>----</td>
</tr>
<tr>
<td>superFUJI-Pu</td>
<td>----</td>
<td>----</td>
<td>----</td>
<td>(superFUJI)</td>
</tr>
<tr>
<td>AMSB proto</td>
<td>----</td>
<td>----</td>
<td>(st)</td>
<td>(hg)</td>
</tr>
<tr>
<td>AMSB-Pu</td>
<td>----</td>
<td>----</td>
<td>(sg)</td>
<td>(AMSB)</td>
</tr>
</tbody>
</table>
6. CONCLUSIONS

It has been demonstrated that a modification of THORIMS-NES accommodating Pu-burnable versions FUJI-Pu and AMSB-Pu might achieve full burn-out of Pu and opening of global Th era (pure Th-233\textsuperscript{U} fuel cycle) till the middle of next century.

This strategy will be fundamentally public acceptable and economical due to the following reasons: (1) simple, continuous and flexible operation and maintenance works for Pu-disposition; (2) safe: no severe accidents; (3) anti nuclear-proliferation and terrorism due to the strong gamma activity of accompanied 232\textsuperscript{U} in fuel/target salt of THORIMS-NES; (4) very few radio-wastes due to nearly no TRU and few maintenance/operational works; (5) very few nuclear materials transportation; (6) small R&D cost owing to few items; (7) high economical potential; and (8) real establishment of idealistic Breeding Fuel Cycle.

In conclusion, here, the Pu disposition issue should be managed promising to solve the energy/environmental problems including world poverty symbolized as "North-South" problem in the next century. This will be a reply to the final proposal of David Lilienthal [18].

We are expecting to start the worldwide debate for international collaboration aiming further improvements in this field.

ACKNOWLEDGEMENTS

The authors wish to express sincere thanks to many friends of USA, France, Russia, India, Switzerland, Canada, Turkey, Germany, Belgium, Japan and the other countries for their kind cooperation and encouragements.

REFERENCES

[3] Lecocq, A., FURUKAWA, K., "Rational Pu Transmutation for 233\textsuperscript{U}", this Meeting.
Exam. Pu-Transmu. by Small Molten-Salt Fission Power St., in this Meeting.


NEUTRONIC EXAMINATION ON PLUTONIUM TRANS_MUTATION BY A SMALL MOLREN-SALT FISSION POWER STATION

K. MITACHI, Y. YAMANA, T. SUZUKI
Toyohashi University of Technology
Toyohashi

K. FURUKAWA
Tokai University, Hiratsuka

Y. KATO
Japan Atomic Energy Research Institute,
Tokai
Japan

Abstract

Neutronic calculations were made to examine the conceptual feasibility of the small molten-salt reactor, FUJI-Pu, for effective plutonium diminution and effective $^{233}\text{U}$ production to use thorium as energy resource. Though the reactor in this report has almost the same features of the one reported previously, the authors intended to consume more Pu and to produce more $^{233}\text{U}$ by changing the reactor operation mode, i.e. yearly isolation of uranium isotopes by applying fluorination and evaporation method. Burn-up characteristics were calculated for 900 days operation at rated power by using ORIGEN-2. The results show that the reactor can transmute 980kg of plutonium isotopes and produce 489kg of $^{233}\text{U}$ annually for 1GW$_e$ of the electricity generation. Hence, the reactor could be an efficient Pu burner and an efficient $^{233}\text{U}$ producer, comparing with LWRs operated with MOX fuel.

1. Introduction

For the effective disposition of weapon-head Pu and reactor grade Pu, the application of Th-$^{233}\text{U}$ recycle system might be the best. Its practical and economical realization depend on fluid-fuel concepts due to the high gamma activity of $^{233}\text{U}$ fuel, which is highly efficient for the prohibition of nuclear proliferation and terrorism.

Among several fluid-fuel concepts, the molten fluorides fuel was verified to be the only usable liquid fuel by the long effort of Oak Ridge National Laboratory in 1947-1976. They aimed to establish a fission breeding power station MSBR$^{[1]}$. However, it has several serious difficulties in design and global application in future. Therefore, our group is proposing more rational approach, named as THORIMS-NES$^{[2]}$, which will be reported by another papers presented in this meeting$^{[3]},{[4]}$. This symbiotic system uses simple size-flexible molten-salt fission power stations. Its significant characteristics are not only high safety and high simplicity in configura-
tion (no core-graphite exchange and no continuous chemical processing), but also few works in operation, maintenance and transportation due to the idealistic near-breeder character even smaller 150MW_e molten-salt reactor, named FUJI-II [5],[6].

Such small molten-salt reactors could be applied for the purpose of Pu disposition. In recent two years, we have been examining this issue in several modes to evaluate and improve its performance[7]. One of them will be reported here, FUJI-Pu, in which the initial fuel salt is composed of fully Pu fissile and Th fertile components. The reactor will be operated with successive addition of Pu and Th, and almost yearly isolation of \( ^{233}\text{U} \) by applying simple fluorination and evaporation method. This operation mode increases neutron economy and minimizes \( ^{233}\text{U} \) burning, resulting higher addition and consumption of Pu and higher production of \( ^{233}\text{U} \). The reactor will be able to realize by few R&D work and cost within 20 years and open a new Th era without Pu.

2. Initial core characteristics

2.1 Calculation method

A schematic diagram of the reactor in the present study is depicted in Fig.1. The primary feature of the reactor core is almost the same as FUJI-II. It is a graphite moderated reactor in which the single fuel-salt containing fissile and fertile materials serves as both the fuel fluid and the blanket fluid. The reactor core is divided into three zones: Core,
Blanket-r and Blanket-z. The reactor can be operated without continuous chemical processing and without graphite replacement for the period of 30 years, i.e. the plant life.

Ten group cross section data (6 for the fast neutron flux and 4 for the thermal neutron flux) are first calculated for all zones of the reactor by using the SRAC code system[8] and the nuclear data file, JENDL-3, developed at Japan Atomic Energy Research Institute. Then, the effective neutron multiplication factor, neutron flux distribution and fission reaction distribution in the reactor were estimated by using two dimensional diffusion code, CITATION.

2.2 Semi-optimized reactor core

It is desirable for the reactor to have higher fuel conversion ratio, smaller plutonium inventory and lower neutron radiation damage. A series of survey calculations were conducted to obtain the overview of the nuclear characteristics of the initial cores, widely changing the parameters such as the concentration of plutonium, the fuel-salt volume fraction in the core, the reactor geometry, etc. As a result, a semi-optimized reactor core such as the plutonium burner, FUJI-Pu, was designed for the condition of acceptable exposure on graphite, $6.3 \times 10^{13} \text{ n/cm}^2/\text{s}$ for the fast neutron flux (>50keV). The exposure on Modified Hastelloy N was limited to $2.3 \times 10^{13} \text{ n/cm}^2/\text{s}$ for the fast neutron flux (>0.8MeV) and $1.2 \times 10^{13} \text{ n/cm}^2/\text{s}$ for the thermal neutron flux (<0.18eV). These limitations on the irradiation cited from the reference[1] can ensure the reactor operation for about 15 equivalent full power years. This is equivalent to about 30 years of reactor run at the load factor of 0.5.

The fuel-salt is a molten-salt mixture of the fluorides of lithium, beryllium, thorium, uranium and plutonium. The composition in the initial core is listed in Table I. The isotopic composition of plutonium supplied to the reactor is shown in Table II for a typical fuel burned in a LWR for about 33GWd/ton of burn-up and 1 year cooling. Several important parameters of FUJI-Pu are listed in Table III. The reactor vessel is 5.3m in diameter and 4.7m in height. The reactor can generate 250MW$_{th}$ of heat. The temperature of fuel-salt at the outlet of the reactor becomes 935K, capable of generating the super heated steam with 25MPa pressure and the temperature of 810K. The output of electricity becomes 100MW$_e$ by installing the steam power generating system with the net thermal efficiency of 40%.

<table>
<thead>
<tr>
<th>Composition</th>
<th>LiF</th>
<th>BeF$_2$</th>
<th>ThF$_4$</th>
<th>PuF$_3$</th>
</tr>
</thead>
<tbody>
<tr>
<td>mol%</td>
<td>71.75</td>
<td>16.0</td>
<td>12.0</td>
<td>0.25</td>
</tr>
</tbody>
</table>

Table I

COMPOSITION OF FUEL-SALT ASSUMED IN THE CALCULATIONS
Table II

ISOTOPIC COMPOSITION OF PLUTONIUM ASSUMED IN THE CALCULATIONS

<table>
<thead>
<tr>
<th>Isotopes</th>
<th>238Pu weight %</th>
<th>239Pu weight %</th>
<th>240Pu weight %</th>
<th>241Pu weight %</th>
<th>242Pu weight %</th>
</tr>
</thead>
<tbody>
<tr>
<td>238Pu</td>
<td>1.5</td>
<td>55.0</td>
<td>25.3</td>
<td>13.2</td>
<td>5.0</td>
</tr>
</tbody>
</table>

The composition is typical for PWR fuel burned-up to 33GWD/ton and left for 1 year before reprocessing.

Table III

PRINCIPAL DESIGN PARAMETERS OF FUJI-Pu

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermal capacity</td>
<td>250 MW&lt;sub&gt;th&lt;/sub&gt;</td>
</tr>
<tr>
<td>Net electric generation</td>
<td>100 MW&lt;sub&gt;e&lt;/sub&gt;</td>
</tr>
<tr>
<td>Thermal efficiency</td>
<td>40 %</td>
</tr>
<tr>
<td>Power density</td>
<td>6.8 MW&lt;sub&gt;th&lt;/sub&gt;/m&lt;sup&gt;3&lt;/sup&gt;</td>
</tr>
<tr>
<td>Reactor vessel Diameter/Height</td>
<td>5.3/4.7 m</td>
</tr>
<tr>
<td>Core zone Maximum radius</td>
<td>1.57 m</td>
</tr>
<tr>
<td>Core zone Graphite fraction</td>
<td>83 vol%</td>
</tr>
<tr>
<td>Blanket-r zone Thickness</td>
<td>0.33 m</td>
</tr>
<tr>
<td>Blanket-r zone Graphite fraction</td>
<td>80 vol%</td>
</tr>
<tr>
<td>Blanket-z zone Thickness</td>
<td>0.40 m</td>
</tr>
<tr>
<td>Blanket-z zone Graphite Fraction</td>
<td>78 vol%</td>
</tr>
<tr>
<td>Maximum neutron flux</td>
<td>5.7x10&lt;sup&gt;13&lt;/sup&gt; n/cm&lt;sup&gt;2&lt;/sup&gt;s</td>
</tr>
<tr>
<td>(Graphite (&gt;50keV))</td>
<td></td>
</tr>
<tr>
<td>(Metal (&gt;0.8MeV))</td>
<td>2.3x10&lt;sup&gt;11&lt;/sup&gt; n/cm&lt;sup&gt;2&lt;/sup&gt;s</td>
</tr>
<tr>
<td>238Pu concentration</td>
<td>0.25 mol%</td>
</tr>
<tr>
<td>ThF&lt;sub&gt;4&lt;/sub&gt; concentration</td>
<td>12.0 mol%</td>
</tr>
<tr>
<td>Volume in reactor</td>
<td>7.5 m&lt;sup&gt;3&lt;/sup&gt;</td>
</tr>
<tr>
<td>Total volume</td>
<td>12.0 m&lt;sup&gt;3&lt;/sup&gt;</td>
</tr>
<tr>
<td>Flow rate</td>
<td>0.55 m&lt;sup&gt;3&lt;/sup&gt;/s</td>
</tr>
<tr>
<td>Temperature In/out</td>
<td>835/935 K</td>
</tr>
<tr>
<td>Fuel conversion ratio</td>
<td>0.58-0.64</td>
</tr>
<tr>
<td>Inventory</td>
<td></td>
</tr>
<tr>
<td>238Pu+241Pu</td>
<td>275 kg</td>
</tr>
<tr>
<td>Th</td>
<td>17.3 ton</td>
</tr>
<tr>
<td>Graphite</td>
<td>167 ton</td>
</tr>
</tbody>
</table>

2.3 Distribution of neutron flux

The distributions of the fast neutron flux in the initial core are shown in Fig.2 and those of the thermal neutron flux in Fig.3. Comparing to FUJI-II, the core volume of FUJI-Pu was made larger to lower the neutron flux and to increase the fuel conversion ratio of the reactor. The core was divided into three zones, and the fuel-salt volume fraction was adjusted in each zone not only to lower the peak value in the distribution of the neutron flux, but also to increase the fissile production in the blanket. 10cm thick graphite plate containing 0.1mol% of boron was placed outside of the graphite reflector to lower the neutron radiation damage on the reactor vessel. The boron consumed by the irradiation of neutron flux during the plant life is about 20% of that contained in the initial graphite plate. As shown in the figures, the neutron flux is made rather uniform in the core. The peak values are 5.7x10<sup>13</sup>n/cm<sup>2</sup>/s (>50keV) in the
Figure 3: Distributions of the thermal neutron flux. The neutron energy is lower than 0.86 eV. The abscissa is the radial distance from the reactor core. The parameter \( r \) denotes the axial distance from the mid-plane of the reactor core.

Figure 4: Distributions of the fast neutron flux. The neutron energy is higher than 0.86 eV. The abscissa is the radial distance from the reactor core.
core, $2.3 \times 10^{11} \text{n/cm}^2/\text{s} (>0.8\text{MeV})$ and $0.3 \times 10^{12} \text{n/cm}^2/\text{s} (<0.18\text{eV})$ at the inner surface of reactor vessel. These values are within the exposure limit above mentioned, so that replacement of the graphite moderator and the reactor vessel will be unnecessary during the plant life.

Figure 4 Procedure to calculate 1 group cross section data from JENDL-3.

3. Burn-up characteristics

3.1 Calculation method

Burn-up characteristics of the reactor were calculated by the procedure illustrated in Figs. 4 and 5. Using RESENDD program\[^3\], 1900 group cross section data of 136 nuclei were first reconstructed from JENDL-3 and the cross sections were composed into 1 group cross section data with 1900 group neutron spectrum obtained by means of cell calculation adapting the collision probability routine in the SRAC code system, as reported in reference [10]. Then, the fission and the capture cross sections of actinides were replaced by those calculated by the SRAC code system to count the self shield effect by resonant nuclei. With these 1 group cross section data, the burn-up calculations were carried out by using ORIGEN-2\[^11\].

In the molten-salt reactor, the fuel-salt was kept homogeneous by its circulation through the core, pumps and heat exchangers. But the composition and the concentration of fissile materials and fission products always changed in the fuel-salt, because of the nuclear reaction in the core and the additional fuel supply from outside of the plant. In order to simulate the continuous change of the composition and the concentration of the fuel-salt, the burn-up time was divided into a series of short intervals "day", and the fission reaction rates was calculated by CITATION for every zones of the reactor. Then, the irradiation and decay calculation were carried out by using ORIGEN-2 for every time intervals $d_i$, defined as follow.
Figure 5 Procedure to calculate burn-up characteristics by using ORIGEN-2.

\[
d_{\text{core}} = \text{day} \times \frac{W_{\text{core}}}{W_{\text{total}}}
\]

with cross sections of Core zone,

\[
d_{\text{bl-r}} = \text{day} \times \frac{W_{\text{bl-r}}}{W_{\text{total}}}
\]

with cross sections of Blanket-r zone,

\[
\text{day} = d_{\text{core}} + d_{\text{bl-r}} + \ldots.
\]

\[
W_{\text{total}} = W_{\text{core}} + W_{\text{bl-r}} + \ldots.
\]

where \(W_{\text{core}}\) is the thermal output of Core and \(W_{\text{bl-r}}\) is the thermal output of Blanket-r. The irradiation and decay calculation were performed with total mass of the fuel-salt under total thermal output, \(W_{\text{total}}\). The calculations were carried out for time interval \(d_{\text{core}}\) with cross sections of Core, for \(d_{\text{bl-r}}\) with cross sections of Blanket-r, etc. The calculation procedure for the time interval "day" was repeated for 900 days run time of the reactor.

Burn-up calculations were carried out for FUJI-Pu running in two operation modes, namely, Case-1 and Case-2. Case-1 simulates the continuous reactor operation at rated power for 900 days without any chemical
processing. Case-2 simulates the reactor operation with uranium removal at every 300 days in batched way by applying simple fluorination and evaporation method after 65 days fuel-salt cooling.

3.2 Feeding rate of Plutonium and Thorium

With the plant operation, the fissionable plutonium isotopes and thorium decrease, and the fission products accumulate in the core, causing lower neutron multiplication factor. Hence, the fissile and fertile materials should be fed to keep the steady reactor operation. The feeding rates of Pu and Th required for this purpose are depicted in Fig. 6. The broken curves show the feeding rate in Case-1 and the solid curves the ones in Case-2. In Case-1, the feeding rate of Pu is about 2600g/day at the beginning and decreases to about 400-500g/day after around 50 days. In Case-2, much amount of Pu should be supplied in every 300 days in order to compensate fissile diminution in the core. Th needs to be fed at about 120g/day after around 300 days in both cases.

The burn-up of the fuel salt causes a change in the neutron flux distribution. The profiles in Case-2 are depicted with two dotted chain curves in Figs. 2 and 3. The profiles of the fast neutron flux after 900 days operation become lower than those of the initial core by 17% in the central zone of the core, while the profiles of the thermal neutron flux are lower than those of the initial core by about 40% throughout the core.
3.3 Inventory of heavy metals

The changes of the fissile inventories in the reactor are shown in Fig.7. The fissile materials in the core are $^{239}$Pu and $^{241}$Pu at the beginning of the reactor operation. These isotopes are consumed with the reactor run, while $^{233}$U is produced from protactinium in the reactor so that the amount of $^{233}$U in Case-1 occupy 23% of total fissile in the core at around 900 days as shown with broken curve in the figure. But in Case-2, fissionable Pu is always dominant in the core, as illustrated with solid curve.

Heavy metal inventories in the reactor system are listed in Table IV. In Case-1, FUJI-Pu can transmute 226kg of Pu and produce 112kg of fissile U and Pa and 15kg of minor actinides in 900 days operation at 100MW$_{e}$ of the rated power. Similarly, the reactor can transmute 242kg of Pu and produce 121kg of fissile U and Pa and 17kg of minor actinides in the Case-2.

![Figure 7](image)

Figure 7 Time-changes of the fissile inventories. Broken curves indicate the time-changes in Case-1, and solid curves the ones in Case-2. Total means the total mass of fissiles ($=^{233}U+^{235}U+^{239}Pu+^{241}Pu$).

3.4 Fuel conversion ratio

The fuel conversion ratio, CR, is defined as the ratio of the fissile nuclei generated in the reactor system to those consumed by the fission and capture reactions in a unit time interval. CR in Case-1 shown with broken
Table IV
FEEDS AND INVENTORIES OF HEAVY METALS

<table>
<thead>
<tr>
<th></th>
<th>Initial</th>
<th>Case-1 operation Feed 900 days</th>
<th>Case-2 operation Feed 900 days</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{233}$Pa</td>
<td>0.</td>
<td>0.</td>
<td>-10.8 kg</td>
</tr>
<tr>
<td>$^{233}$U</td>
<td>0.</td>
<td>0.</td>
<td>106.4 kg</td>
</tr>
<tr>
<td>$^{235}$U</td>
<td>0.</td>
<td>0.</td>
<td>0.3 kg</td>
</tr>
<tr>
<td>total U</td>
<td>0.</td>
<td>0.</td>
<td>115.2 kg</td>
</tr>
<tr>
<td>$^{239}$Pu</td>
<td>205 kg</td>
<td>282 kg</td>
<td>246 kg</td>
</tr>
<tr>
<td>$^{241}$Pu</td>
<td>49 kg</td>
<td>68 kg</td>
<td>114 kg</td>
</tr>
<tr>
<td>total Pu</td>
<td>373 kg</td>
<td>513 kg</td>
<td>660 kg</td>
</tr>
<tr>
<td>Th</td>
<td>17.4 ton</td>
<td>133 kg</td>
<td>17.4 ton</td>
</tr>
<tr>
<td>Am+Cm</td>
<td>0.</td>
<td>0.</td>
<td>15 kg</td>
</tr>
</tbody>
</table>

curve in Fig. 8 is about 0 at the beginning of the reactor run, and increases to about 0.6-0.64 after 200 days, because the production of $^{233}$U in the reactor is delayed owing to the decay time of protactinium, Pa. CR in Case-2 indicated with solid curve decreases sharply at every 300 days, because CR does not count the fissile uranium isolated out of the reactor system.

Figure 8 Time-changes of the fuel conversion ratio, CR, and the plutonium conversion ratio, $\xi$. Broken curves indicate the time-changes in Case-1, and solid curves the ones in Case-2.
In the same way, we define plutonium conversion ratio, $\xi$, as the ratio of integrated amount of $(^{233}U + ^{235}U + Pa)$ produced to integrated fissionable Pu consumed during the reactor operation, so that $\xi$ counts the fissile uranium removed out of the reactor system. The changes of $\xi$ are depicted in Fig.8. Though the $(^{233}U + ^{235}U + Pa)$ production is enhanced in Case-2, the fissile Pu consumption is increased at the same time, resulting almost the same values of 0.46-0.5 in the two cases.

3.5 Production rates of uranium and plutonium

The production rates of $^{233}U$, fissile $U$ and $Pa$, $^{239}Pu$ and total $Pu$ isotopes in Case-1 are shown by the broken curves in Fig.9, and the production rates of those in Case-2 by the solid curves. The reactor operation with yearly isolation of $U$ isotopes increases neutron economy and minimizes $^{233}U$ burning, resulting higher addition and consumption of $Pu$ and higher production of fissile $U$ and $Pa$, as shown in the figure.

The molten salt reactor was compared with LWRs in view of the plutonium consumption. As one can see from Table V, FUJI-Pu can transmute 919kg of $Pu$ in Case-1 and 980kg in Case-2 annually for $1GW_e$ of the electricity generation. These amounts are about 50% larger than the ones consumed by a LWR, verifying higher performance of FUJI-Pu as the plutonium burner.

Using uranium oxide fuel discharged at 45GWd/ton of the burn-up, the boiling water reactor BWR(U) can produce 230kg of plutonium annually for $1GW_e$ generation. If charged with MOX fuel in place of uranium oxide fuel, the reactor BWR(MOX) is able to burn 649kg of plutonium. But, LWRs charged...
Table V
PRODUCTION RATES OF PLUTONIUM, URANIUM AND MINOR ACTINIDES

<table>
<thead>
<tr>
<th></th>
<th>BWR(U)</th>
<th>BWR(MOX)</th>
<th>FUJI-Pu Case-1</th>
<th>FUJI-Pu Case-2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermal output</td>
<td>3000 MW\text{th}</td>
<td>3000 MW\text{th}</td>
<td>250 MW\text{th}</td>
<td>250 MW\text{th}</td>
</tr>
<tr>
<td>Electrical output</td>
<td>1000 MW\text{e}</td>
<td>1000 MW\text{e}</td>
<td>100 MW\text{e}</td>
<td>100 MW\text{e}</td>
</tr>
<tr>
<td>Net generation per 1GW\text{e}·y</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>total Pu</td>
<td>230 kg</td>
<td>-649 kg</td>
<td>-919 kg</td>
<td>-980 kg</td>
</tr>
<tr>
<td>Am+Cm</td>
<td>25 kg</td>
<td>133 kg</td>
<td>62 kg</td>
<td>71 kg</td>
</tr>
<tr>
<td>$^{239}$Pu,$^{241}$Pu</td>
<td>142 kg</td>
<td>-617 kg</td>
<td>-990 kg</td>
<td>-1062 kg</td>
</tr>
<tr>
<td>$^{233}$U,$^{235}$U+Pa</td>
<td>0.</td>
<td>0.</td>
<td>453 kg</td>
<td>489 kg</td>
</tr>
<tr>
<td>total fissile</td>
<td>-659 kg</td>
<td>-639 kg</td>
<td>-537 kg</td>
<td>-573 kg</td>
</tr>
</tbody>
</table>

BWR(U) denotes the boiling-water reactor operated with uranium-oxide fuel and BWR(MOX) denotes the reactor operated with mixed-oxide fuel. The generation rates of plutonium isotopes were calculated from the results of reference [12]. The discharge burn-up of fuel elements of BWR(U) and BWR(MOX) is 45GWd/ton.

with MOX fuel seem to be ineffective for plutonium disposition, because about 70% of Pu remains unused in the discharged fuels, and multiple recycling is necessary. Moreover, Pu causes harder neutron spectrum and may spoil neutron economy. Therefore, the diminution of plutonium by LWRs might be expensive and wasteful of the energy resource.

FUJI-Pu can consume 990kg of fissile Pu and produce 453kg of fissile U in Case-1, and consume 1062kg of fissile Pu and produce 489kg of fissile U in Case-2, so that the net rate of the fissile consumption is about 537kg/GW\text{e}·y in Case-1 and 573kg/GW\text{e}·y in Case-2, respectively. The net rate of the fissile consumption of LWRs were 659kg/GW\text{e}·y for BWR(U), and 639kg/GW\text{e}·y for BWR(MOX). It can be said that FUJI-Pu can transmute large amount of plutonium and consumes little fissile comparing with LWRs run with the MOX fuel. FUJI-Pu run in two operation modes produce 62kg and 71kg of minor actinides annually for 1GW\text{e} generation. This may cause a problem in the incineration of the radioactive wastes, although more amount of minor actinides are produced by BWR(MOX).

4. Conclusions

A theoretical consideration has been made of the nuclear characteristics of the small molten-salt power reactor, FUJI-Pu, in which plutonium recovered from spent-fuels of the LWRs was supposed to be used. The reactor adopted yearly isolation of uranium isotopes to increase Pu consumption and $^{233}$U production. And the following conclusions were made.

1) The reactor transmutes 980kg of Pu annually for 1GW\text{e} generation,
which is larger than that by a LWR operated with MOX fuel.

2) The net fissile uranium produced by the reactor is 489kg annually for 1GW_e generation.

3) The fuel conversion ratio of the reactor is about 0.6, and the plutonium conversion ratio is about 0.46.

4) The reactor can be operated without graphite replacement for 30 years of the reactor life.

The authors wish to express sincere thanks to Dr. Tsuyoshi Misawa and Mr. Masahiko Osaka of Nagoya university on their help and advise in calculating the nuclear cross section data by using the code "RESENDD".

REFERENCES


[3] LECOCQ,A.,


STUDY OF THE POSSIBILITY OF USING MOLTEN SALTS FOR PLUTONIUM UTILIZATION AND ACTINIDE TRANSMUTATION

V.S. NAUMOV, A.V. BYCHKOV,
O.V. SCIBA, P.T. PORODNOV
Federal Scientific Centre Research Institute
of Atomic Reactors, Dimitrovgrad,
Russian Federation

Abstract

Proposed is the concept of using molten salts for plutonium utilization and actinide transmutation in the electronuclear power complexes. Described is the arrangement of fuel cycle with account to fuel composition, content of plutonium and total volume of fuel salt. The technological flowsheet provides for electrochemical reprocessing methods with electrolytic actinide release into metallic alloy and with their further return to the fuel salt.

Traditionally RIAR carries out research and development work on plutonium utilization in closed fuel cycle of FBR.

For 25 years the following milestones have been:
- development of pyroelectrochemical technology for FBR MOX fuel production; to date about 3 tons of MOX fuel have been produced;
- development of vibropacked fuel pins and its technology; currently they are being irradiated in BOR-60, BN-350 and BN-600 reactors;
- development and construction of remotely controlled facility for granulated fuel, vibropacked fuel pins and fuel assemblies production;
- full-scale demonstration of Pu fuel utilization in FBR;
the BOR-60 reactor has been successfully operated on MOX fuel for about 12 years;
- 26% burn-up was reached with MOX fuel in the BOR-60;
- one experiment was carried out on the pyrochemical reprocessing of spent FBR MOX fuel.

Now, study of Pu utilization in FBR, PWR and Pu-burner reactors is one of main experimental trends in the RIAR activity.

Plutonium can be used in modern type reactors as oxide fuel but plutonium recycle can be effectively realized only on the basis of pyroelectrochemical reprocessing and vibropacking technology.

For future nuclear systems it would be possible to use new principles related to Pu and other fissile materials recycle. One of proposed ways of implementing this could be to use molten salt fuel.

The concept of using liquid fuel based on halide molten salts in nuclear reactors has been known since the 50s.

The efficiency of using molten salts is determined by some of their specific properties and features:
- ability to dissolve many inorganic compounds both in gaseous and in solid state;
- enough inertness to the structural materials;
- intensity of the processes taking place in those environments due to the high values of the mass
Among molten salts the fluorides and chlorides of metals are characterized by the high chemical and thermal stability due to the greatest heat formation.

In spite of the fact that individual fluorides have high melting temperatures their mixtures with a particular component composition are characterized by lower melting temperatures. For example, LiF-BeF₂ mixtures has an eutetic point at 350°C and LiF content of 47 mol%.

Hypothetic compositions of molten fluoride solvents for fuel corresponding to eutetic compositions are presented in Table 1.1 [1.2].

The use of these salts as components of fuel composition is conditioned by the following advantages:

- high radiation resistance permitting work with fuel at burnup of 100 MWd/kg;
- high thermal resistance permitting reprocessing fuel with specific heat release exceeding 1 MWd/kg;
TABLE 1.2. PHYSICAL PROPERTIES OF FLUORINE SALT

<table>
<thead>
<tr>
<th>Solar composition</th>
<th>Melting temperature, °C</th>
<th>Density at 700°C, g/cm³</th>
<th>Volume expansion coefficient at 100°C, ppm/°C</th>
<th>Viscosity at 700°C, cP</th>
<th>Thermal conductivity, cal</th>
<th>Specific thermal conductivity, cal/g</th>
</tr>
</thead>
<tbody>
<tr>
<td>53.5NaF-40ZrF₄-6.5UF₄</td>
<td>540</td>
<td>3.27</td>
<td>3.36</td>
<td>5.7</td>
<td>0.005</td>
<td>0.24</td>
</tr>
<tr>
<td>71LiF-16BeF₂-12ThF₄-1.0UF₄</td>
<td>500</td>
<td>3.25</td>
<td>2.52</td>
<td>7.1</td>
<td>0.00287</td>
<td>0.32</td>
</tr>
<tr>
<td>73.7LiF-16BeF₂-10ThF₄-0.3UF₄</td>
<td>500</td>
<td>3.25</td>
<td>2.01</td>
<td>2.29</td>
<td>-</td>
<td>0.34</td>
</tr>
<tr>
<td>67LiF-30.5BeF₂-2.5UF₄</td>
<td>464</td>
<td>2.10</td>
<td>1.90</td>
<td>5.5</td>
<td>-</td>
<td>0.57</td>
</tr>
<tr>
<td>52LiF-48BeF₂</td>
<td>350</td>
<td>2.16</td>
<td>4.98</td>
<td>-</td>
<td>-</td>
<td>0.67</td>
</tr>
<tr>
<td>76LiF-14BeF₂-10UF₄</td>
<td>500</td>
<td>3.32</td>
<td>5.93</td>
<td>-</td>
<td>-</td>
<td>0.39</td>
</tr>
<tr>
<td>Water (at 20°C)</td>
<td>0</td>
<td>1.0</td>
<td>2.0</td>
<td>1.0</td>
<td>0.00143</td>
<td>1.0</td>
</tr>
</tbody>
</table>
- possibility to create high concentration of transuranium elements in melts because salts which are
used as working environments are not neutron moderators.

The physical properties of fluoride salts mixtures as liquid nuclear fuel are given in Table 1.2 [3].
These properties of fluoride salts suggest that one of the ways for utilization of plutonium excess and
long-lived actinides can be electronuclear power facilities using accelerator and blanket with circulating
fuel or molten salt reactors (MSR).

The classic projects of molten salt reactors (MSBR, MSRE) developed at Oak-Ridge National
Laboratory demonstrated a high solubility of uranium and thorium compounds in fuel salt.

---

**FIG. 1.1. Solubility of PuF$_3$ in LiF-BeF$_2$ melts.**

---

**FIG. 1.2. Solubility of PuF$_3$ in LiF-ThF$_4$ melts.**
Most data on composition and usage of fuel salts are based on the data of phase diagrams of such systems which provide an estimated solubility value for these salts.

Figs.1.1-1.2 present data on solubility of PuF\textsubscript{3} in LiF-BeF\textsubscript{2} and LiF-ThF\textsubscript{4} melts [2,4-6]. Therefore, as evident from the data presented for the MSCR and MSBR reactors the Pu content in the molten salt at 0.5 mol.% is quite possible. The same concentration of Pu in fuel salt is expected to be at the ABC/ATW facility (see Fig.1.3.)

At the present time the main scientific prerequisites for problem of plutonium utilization in molten salt fuel has been formulated.

1. The investigations were performed to study the behaviour of uranium and plutonium compounds as well as some properties of minor-actinides and the main fission products in these environments.

2. A great amount of information has been generated sufficient for substantiation of pyroelectrochemical and pyrochemical fuel reprocessing and for development of different models of this cycle depending on the initial and final fuel state.

3. The fundamental possibility for handling and reprocessing of molten salt fuel has been demonstrated.

4. The foundations of closed fuel cycle using molten salts have been developed.

Studying the processes of plutonium utilization and actinide transmutation at MSR and also at the ABC/ATW facilities it is necessary to consider and evaluate the alternatives related to the arrangement of their fuel cycle due to the fact that the use of plutonium and minor-actinides propose that the effective burning is possible with its multiple recycling for irradiation.

The well-known flowsheet of the MSBR fuel cycle was developed at ORNL.
The consideration of fuel cycle options for such facility was performed with account to fuel composition, content of plutonium and total volume of fuel salt.

In contrast to fuel cycles of MSBR and MSRE projects which involved the flowsheets with complete fuel composition reprocessing and a great number of technological operations, the flowsheet of the actinide transmutation facility can be rather simplified (Fig.1.4.). The gas removal system, for example, may be similar to that developed in the MSBR project. Moreover, the fuel salt flux should pass through the "noble" FP release circuit. In this case the complete reprocessing is possible to be carried out according to the flowsheet allowing for the pyroelectrochemical reprocessing.

These methods suggest that on the first stage the electrolytic deposition of all actinides into the metallic liquid alloy is performed. After that, the salt from the first apparatus (electrolyzer) is delivered to the second apparatus intended for rare earth elements removal. On the second stage the salt comes back to the first apparatus where the electrochemical extraction of actinides into fuel salt takes place. In this case the more electropositive elements (for this system such elements as Zr, Mo,Fe etc.) will be accumulated in the alloy.

After reprocessing the fuel salt again returns into the actinide burning facility circuit. The metallic alloy is subject to further reprocessing in molten salt to have the fission products in compact form as oxide deposits or in the form of metallic alloy with further disposal.

Such technology will make it possible to maintain a particular content of fission products and rare earths that will depend on the molten fuel volume subjected to reprocessing.

The range of the expected separation factor of Pu from rare earths is 20-50 and that of the salt from impurities is 100-500.

From the technical standpoint the flowsheet of this fuel cycle option has the most similar efficiency with the one developed on the basis of the ORNL data. Its application can lead to sufficient fuel purification from the impurities with the simple equipment used.

Therefore, study of the possibility of using molten salts for plutonium utilization and actinide transmutation shows that the fuel cycle based on the use of molten salts in fuel reprocessing can organically be integrated into the structure of nuclear power complex and will allow tackling the problem of high toxic radioactive wastes in a new way. Nevertheless, to have these problems solved, further work in the field of fundamental research and development of separate methods for fuel handling are necessary.

On our opinion the practical implementation of such programs and projects will greatly depend on the efficiency of the international collaboration on this problem.

This work was performed by supported of the U.S. LANL under contract 5058L0013-9Y.

REFERENCES


SESSION 5: FAST REACTORS
STUDY ON Pu BURNER FAST REACTOR CORES WITHOUT URANIUM

M. ISHIKAWA, A. SHONO, T. WAKABAYASHI
O-arai Engineering Center, Power Reactor and Nuclear Fuel Development Corporation,
Ibaraki, Japan

Abstract

In Japan, extensive study of fast reactor cores, which can efficiently burn plutonium and/or minor actinides, has been continued in order to demonstrate the flexibility of the actinide utilization in fast reactors. Recently, concerns over the excessive plutonium have been significantly increased from the viewpoint of nuclear proliferation. Fast reactor cores without uranium are considered as one of the most effective solutions to reduce the plutonium stock. The present paper summarizes nuclear characteristics of the fast reactor cores fueled with PuO₂/ceramics, instead of PuO₂/UO₂, including their burnup performance of plutonium, reactivity coefficients and some problems from the safety viewpoint. As a result of these surveys, such unconventional cores without uranium are found to consume the plutonium at very large burnup rate close to the theoretically maximum value of 110-120 kg/TWh. Further, sodium void reactivity in the non-uranium cores decreases extremely, which is preferable to safety consideration. On the other hand, some disadvantages to substantiate a reactor design of the non-uranium core would be large burnup reactivity loss and small Doppler effect.

1. Introduction

Recent global relaxation of the demand for primary energy sources leads to the serious delay of fast breeder reactor development and construction. We now have to expect a large amount of overbalance and excessive stock of plutonium from light water reactor (LWR) operation. In Japan, there are firm policies\(^1\) for the atomic energy usage: (a) to restrict Japanese nuclear research, development, and utilization exclusively for the peaceful purpose, (b) to recycle nuclear fuel, that is, to reprocess the spent fuel, recycle the recovered plutonium and uranium, and use them as nuclear fuel repeatedly, (c) to accept the IAEA's safeguards agreement under the Non-Proliferation Treaty (NPT), and, as a consequence of above-mentioned fundamental lines, (d) not to reprocess and stockpile more plutonium than is needed to feed reactors as fuel, since we understand that there are some concerns over the proliferation of nuclear weapons with the dispersion of reactor grade plutonium.

The emphasis of Japanese policy for the plutonium utilization is to treat plutonium as a valuable energy resource, not as useless nuclear waste. Due to their good neutronics characteristics, fast reactors intrinsically have good flexibility of plutonium consumption as well as breeding. In the course of these backgrounds, we are now investigating an efficient way to control the plutonium stock using fast reactor system. As an international cooperation related to the efficient plutonium
burning, the CAPRA project\(^{(2)}\) is also ongoing to assess the feasibility and to develop the reality.

The present paper describes recent results of our plutonium burning study in fast reactor cores.

2. Approach to Efficient Burning of Plutonium

There are general approaches to suppress the generation of Pu and to burn Pu efficiently in conventional MOX-type fast reactor cores, as shown in Fig.1. First of all, it is natural that the blanket region whose objective is Pu breeding from depleted uranium, must be eliminated from the core. Next, in the core fuel region, the Pu enrichment of the core fuel should be increased as much as possible, in order to make the amount of U238 minimum so that the conversion ratio in the core region decreases.

The concrete methods to increase the Pu enrichment may be: (a) to obtain small volume ratio of fuel by employing innovative fuel design such as wide gap between fuel pellet and clad, large annular pellet hole, spacious fuel-pin pitch, scattered empty pin, etc., (b) to enhance neutron leakage from the core region by the adoption of shorter core height, smaller core size, etc., and (c) to introduce neutron absorber material such as B4C to the fuel.

It is a straightforward consideration if we extensively evolve above-mentioned methods for the conventional MOX burner cores, that the ideal solution to burn Pu
most efficiently is to completely exclude the generating source of Pu, i.e., uranium, from the core. In this “non-uranium” case, we can expect the Pu burning rate close to the theoretically limited consumption of 110-120 kg/TWhe, because almost all thermal energy generated in the core arises only from the fission reaction of Pu.

As actual fuel material of non-uranium idea, however, we cannot apply pure PuO2 fuel, since the core size would be too small and/or the fuel pin configuration would be irrational from the viewpoint of power generating core. We adopted here a dilution method where PuO2 is dispersed with some non-fuel material such as ceramics to construct reasonable configuration of the core.

The concerns of this unconventional approach to burn Pu would be: (a) performance of the core as power generation plant, especially, length of continuous operating period, (b) controllability and safety of the core with unfamiliar characteristics such as small Doppler effect, (c) soundness of fuel during irradiation, and (d) suitability to fuel manufacturing, reprocessing in the fuel cycle. Thus, there are many physical and industrial aspects to be investigated in the non-uranium core option. As the first step of feasibility study, we here concentrate on the neutronics characteristics of the core.

3. Characteristics of Reference “Non-Uranium” Core

In this chapter, nuclear characteristics of reference cores without uranium are described with the design parameters and the calculational method.

3.1 Reference Core

We take a power generation core of 800 MW electric (2080 MW thermal) as the reference case. The main design parameters of the core are specified in Table 1, and the configuration and dimensions are shown in Fig.2.

Table 1 Design Parameters of Reference "Non-Uranium" Core

<table>
<thead>
<tr>
<th>Item</th>
<th>Parameters</th>
</tr>
</thead>
<tbody>
<tr>
<td>Reactor Power</td>
<td>800 MWe (2,080 MWT)</td>
</tr>
<tr>
<td>Operation Period</td>
<td>182.5 days/cycle</td>
</tr>
<tr>
<td>Number of Refueling</td>
<td>6 batches</td>
</tr>
<tr>
<td>Pu Isotopic Composition</td>
<td>Pu238/Pu239/Pu240/Pu241/Pu242/Am241 = 1.8 / 58.2 / 22.3 / 11.1 / 5.5 / 1.1</td>
</tr>
<tr>
<td>Core Concept</td>
<td>Two-Region Homogeneous Core</td>
</tr>
<tr>
<td>Core Height</td>
<td>60 cm</td>
</tr>
<tr>
<td>Core Diameter (Outer / Inner)</td>
<td>523 / 368 cm</td>
</tr>
<tr>
<td>Volume Ratio of Core Region</td>
<td>Fuel / Coolant / Structure / Void = 23.4* / 57.3 / 14.6 / 4.7</td>
</tr>
</tbody>
</table>

* includes Ceramics Diluent
The reference core is a two-region homogeneous type and 60 cm high, which is a kind of pancake shape to enhance neutron leakage from the core. On the other hand, the core diameter is quite large, 5.2 m, to keep half-a-year continuous operation period which would be needed from the economy viewpoint of a power generation plant. The number of refueling batches is six, hence each fuel subassembly is irradiated in the core for three years. The isotopic ratio of the Pu fuel, 69% of Pu fissile contents, is assumed as spent fuel of some typical LWR. Some kinds of ceramics such as Al2O3, BeO, MgO or CeO2 are selected as diluent of PuO2. The fuel volume ratio is 23%, which is quite low compared with conventional MOX-type breeder cores. Note that we increased the coolant volume ratio corresponding to the decrease of fuel volume ratio in the reference case, however, there would be some alternatives such as increase of the structure material or diluent.

3.2 Analytical Method

The nuclear data used here is an ABBN-type 70-group constant set for fast reactors based on Japanese Evaluated Nuclear Data Library (JENDL) version 2(3). Concerning with nuclear data dependency of the analysis, we also applied the latest version of nuclear data, JENDL-3.2(4), and found no visible difference in the results. The burnup composition change of fuel and the burnup reactivity loss are obtained by two-dimensional diffusion calculation with collapsed seven energy groups. The reactivities of sodium void and Doppler are evaluated by 70-group calculation to the condition with zero sodium density or isothermally increased...
temperature in the core region, respectively. The components of these reactivities are estimated by the diffusion perturbation theory.

### 3.3 Characteristics of Reference Core

The neutron spectrum of a reference non-uranium core with Al$_2$O$_3$ ceramics is compared with both a conventional fast breeder core and a MOX-type Pu burner core in Fig.3. The spectrum of the non-uranium core is greatly softened from these conventional MOX cores because of enhanced slowing down by lighter elements existing in non-fuel materials. The extreme change of the spectrum is expected to affect reactivity characteristics.

Some important core characteristics of the non-uranium cores are summarized in Table 2, which also includes those of a MOX-type Pu burner core with 30% of Pu enrichment for comparison.

1. The Pu consumption rates of the non-uranium cores are almost double of the MOX-Type Pu burner and the value of 110 kg/TWhe is close to theoretical maximum as expected.

2. Due to lack of Pu conversion, the burnup reactivity loss is extremely increased to 1.1-1.2 %/dk/kk'/month. If a longer operating period than half-a-year is required, we may have to consider some remedies for the control rod number or other design parameters.

![Fig. 3 Comparison of Neutron Spectrum](image)
Table 2  Characteristics of Reference "Non-Uranium" Cores

<table>
<thead>
<tr>
<th>Fuel Type</th>
<th>MOX burner</th>
<th>PuO₂/BeO</th>
<th>PuO₂/Al₂O₃</th>
<th>PuO₂/MgO</th>
<th>PuO₂/CeO₂</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu Content in Fuel (in/out, (v/o))</td>
<td>28.5/31.5</td>
<td>25.9/25.6</td>
<td>26.9/27.3</td>
<td>26.8/27.1</td>
<td>26.9/27.6</td>
</tr>
<tr>
<td>Pu Consumption (kg/TWhe)</td>
<td>55</td>
<td>110</td>
<td>111</td>
<td>110</td>
<td>110</td>
</tr>
<tr>
<td>Pu Consumption (kg/TWhe)</td>
<td>58</td>
<td>118</td>
<td>116</td>
<td>116</td>
<td>114</td>
</tr>
<tr>
<td>Conversion Ratio*</td>
<td>0.51</td>
<td>0.15</td>
<td>0.12</td>
<td>0.12</td>
<td>0.11</td>
</tr>
<tr>
<td>B.U Reactivity Loss (%Δk/kk'/month)</td>
<td>0.49</td>
<td>1.20</td>
<td>1.13</td>
<td>1.13</td>
<td>1.10</td>
</tr>
<tr>
<td>Na Void Reactivity* (%Δk/kk')</td>
<td>2.14</td>
<td>-0.66</td>
<td>-0.95</td>
<td>-0.98</td>
<td>-1.03</td>
</tr>
<tr>
<td>K_Doppler* (X10⁻³Td/k/dT)</td>
<td>-6.6</td>
<td>-4.7</td>
<td>-2.8</td>
<td>-2.9</td>
<td>-2.4</td>
</tr>
</tbody>
</table>

Reactor Power 2080MWh, Reactor Size=0.6mH×5.2mD
Pu Vector (Pu239/Pu239/Pu239/Pu241/Pu242/Am241 , (w/o))= (180/5820/2230/1110/550/110)
Volume Fraction (Fuel / Na / Structure)=23.4 / 57.3 / 14.6
* at Beginning of Equilibrium Cycle

(3) The sodium void reactivity shows marked decrease from the conventional MOX core and reaches even negative values. This is quite favorable from the safety point of view. The mechanism of the great decrease can be understood from the component analysis shown in Fig.4. The dominant reduction of sodium void reactivity was achieved by the decrease of scattering term, which came from the spectral softening as mentioned above. We also found the effect of neutron leakage rather insignificant for the sodium void reactivity change.

![Fig. 4 Components of Sodium Void Reactivity](image-url)
(4) One of the most concerns for the non-uranium cores would be the large decrease of Doppler reactivity absolute values, since the missing U238 is the dominant source of the Doppler contribution in conventional MOX cores. However, the Doppler coefficients of the non-uranium cores still keep negative. The contribution of individual nuclides is clarified in Fig.5, by which we understand the negative Doppler reactivity originates mainly from two nuclides, that is, Pu240 and iron. We need to be very careful for the results. Pu240 is one of the fuel isotopes, but the initial amount depends on the kind of LWR and/or the burnup degree of the spent fuel from which the Pu originates, and the isotopic composition also largely changes in time during the reactor operation. On the other hand, iron is included in structure material, therefore, the iron contribution to the Doppler reactivity in actual reactor operation will be quite smaller than the isothermal condition due to both smaller temperature change and thermal response delay. We are now studying the effect of the small Doppler reactivity to the safety characteristics by transient analysis.

(5) We should point out some other concerns for the non-uranium cores. One is the small value of effective delayed-neutron fraction, approximately two-thirds of conventional MOX cores, which comes from the difference of delayed-neutron yield between plutonium and uranium. Although we could manage this in the plant control system design phase, the verification by dynamic calculation would be needed. The other concerns are related to the power distribution swing in the core region and the power mismatch among different burnup-experienced subassemblies by many refueling batches. In the conventional MOX cores,
Table 3  Effect of B$_4$C Introduction in Core Region
(Fuel Type: PuO$_2$/Al$_2$O$_3$)

<table>
<thead>
<tr>
<th>B$_4$C Volume Ratio (%)$^*$</th>
<th>0</th>
<th>10</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu Content in Fuel (in/out, (v/o))</td>
<td>18.4/19.0</td>
<td>28.2/30.4</td>
</tr>
<tr>
<td>Pu Consumption (kg/TWhe)</td>
<td>108</td>
<td>111</td>
</tr>
<tr>
<td>PuC Consumption (kg/TWhe)</td>
<td>114</td>
<td>111</td>
</tr>
<tr>
<td>Conversion Ratio**</td>
<td>0.14</td>
<td>0.09</td>
</tr>
<tr>
<td>B.U. Reactivity Loss (%$\delta$k/k'$/month)</td>
<td>0.86</td>
<td>0.44</td>
</tr>
<tr>
<td>Na Void Reactivity** (%$\delta$k/k')</td>
<td>-0.76</td>
<td>1.48</td>
</tr>
<tr>
<td>$K_{Doppler}^{**}$ ($\times 10^{-3}$Tdk/dT)</td>
<td>-3.4</td>
<td>-0.17</td>
</tr>
</tbody>
</table>

Volume Fraction (Fuel/Na/Structure) = 41.6/37.5/20.9
Core configuration is slightly different from the reference core.
$^*$ to Fuel Volume
** at End of Equilibrium Cycle

these power changes in time are alleviated by the conversion of U238 to Pu. We expect, however, this power drift could be handled in the optimizing effort of the design, because the Pu burner cores have low power density in nature so as to reduce the burnup reactivity loss and to keep a sufficient operating period.

Table 4  Effect of UO$_2$ Introduction Homogeneously in Core Region

<table>
<thead>
<tr>
<th>Fuel Type</th>
<th>PuO$_2$/BeO</th>
<th>PuO$_2$/CeO$_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu Consumption (kg/TWhe)</td>
<td>17% Decrease</td>
<td>12% Decrease</td>
</tr>
<tr>
<td>PuC Consumption (kg/TWhe)</td>
<td>14% Decrease</td>
<td>11% Decrease</td>
</tr>
<tr>
<td>Conversion Ratio*</td>
<td>$0.15 \Rightarrow 0.24$</td>
<td>$0.11 \Rightarrow 0.19$</td>
</tr>
<tr>
<td>B.U. Reactivity Loss (%$\delta$k/k'$/month)</td>
<td>9% Decrease</td>
<td>9% Decrease</td>
</tr>
<tr>
<td>Na Void Reactivity* (%$\delta$k/k')</td>
<td>$0.6 \sim 0.7$ Increase</td>
<td>$0.8 \sim 0.9$ Increase</td>
</tr>
<tr>
<td>$K_{Doppler}^*$ ($\times 10^{-3}$Tdk/dT)</td>
<td>$22 \sim 25%$ Increase</td>
<td>$11 \sim 30%$ Increase</td>
</tr>
</tbody>
</table>

UO$_2$ amount : 10 v/o of Fuel Volume, U/Pu$\approx 0.3$
Volume Fraction (Fuel/Na/Structure) = 23.4/57.3/14.6
* at Beginning of Equilibrium Cycle
4. Improvement of Reactivity Coefficients

Although the non-uranium cores possess ideal performance of Pu burning rate, we have found some disadvantages if we take them as power generation sources. Some options to improve the core characteristics are surveyed here.

4.1 Introduction of Burnable Poison

In order to reduce the large burnup reactivity loss, 10% fuel volume of B$_4$C was introduced homogeneously in the core region as a burnable poison. The effects on core characteristics are summarized in Table 3. The burnup reactivity loss was decreased largely, almost a half of the reference case. However, the sodium void reactivity was increased by approximately 2%$\Delta k$/k', and the Doppler reactivity came close to zero. These changes are caused by the spectral hardening due to neutron absorption by boron. It seems reasonable we discard this option from the safety aspect.

4.2 Re-introduction of UO$_2$ Homogeneously in Core Region

Apparently, the disadvantages of the reference cores arise from the lack of uranium itself. There might be a possibility that a small amount of UO$_2$ recovers the unfavorable characteristics of the non-uranium core without serious influence on Pu burning performance. This is a kind of trade-off problem. We first selected the introduction of UO$_2$ homogeneously in the core region. Table 4 shows the effect for the two diluent cases, where the amount of UO$_2$ added is 10% of fuel volume. As negative changes, the Pu consumption rate decreased by 12-17%, and sodium void reactivity increased 0.6-0.9%$\Delta k$/k'. On the other hand, the burnup reactivity loss improved by 9%, and the Doppler 11-30%. We judge the merit of this option is not so attractive compared with the sacrifice of burner performance.

4.3 Adoption of UO$_2$ Internal Blanket Axially in Core Region

In the conventional MOX-type FBR design, axially heterogeneous core is considered as one of the most promising concept to enhance the core performance. We considered the similar idea could be adopted in the Pu burner case, because the internal blanket can be expected to affect the reactivity characteristics efficiently by the placement in the highest importance region, while Pu burning is the characteristics of the whole core summation.

Table 5 compares the core characteristics of the internal blanket case with those of a reference non-uranium core and a MOX-type Pu burner core. Although the Pu consumption rate of the internal blanket case is decreased by 15% from the reference non-uranium core, the improvement of burnup reactivity is 24%, which is far larger than the sacrifice of burning performance. In addition, increase of the sodium void reactivity is only 0.5%$\Delta k$/k', which is much smaller than the homogeneous UO$_2$ case.

A marked improvement of this option appears in the isothermal Doppler reactivity, which recovered by 87% and even close to that of the conventional MOX-type FBR cores. Although we have to consider the degradation of the Doppler
Table 5  Effect of UO$_2$ Internal Blanket Axially in Core Region

<table>
<thead>
<tr>
<th>Fuel Type</th>
<th>MOX Burner*</th>
<th>PuO$_2$/Al$_2$O$_3$**</th>
<th>Axial Internal Blanket*** in PuO$_2$/Al$_2$O$_3$***</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu Content in Fuel (in/out, (v/o))</td>
<td>28.5/31.5</td>
<td>15.7/15.6</td>
<td>18.5/17.5</td>
</tr>
<tr>
<td>Pu Consumption (kg/Thw)</td>
<td>55</td>
<td>111</td>
<td>94 (-15%)#</td>
</tr>
<tr>
<td>Pu Consumption (kg/Thw)</td>
<td>58</td>
<td>119</td>
<td>102</td>
</tr>
<tr>
<td>Conversion Ratio****</td>
<td>0.51</td>
<td>0.13</td>
<td>0.23</td>
</tr>
<tr>
<td>B.U. Reactivity Loss (% $\delta$ k/kk'/month)</td>
<td>0.49</td>
<td>1.13</td>
<td>0.86 (-24%)#</td>
</tr>
<tr>
<td>Na Void Reactivity**** (% $\delta$ k/kk')</td>
<td>2.14</td>
<td>-0.09</td>
<td>0.44 (+0.53% $\delta$ k/kk')#</td>
</tr>
<tr>
<td>$K_{Doppler}$**** ($\times 10^{-3}$Td/k/dT)</td>
<td>-6.6</td>
<td>-3.8</td>
<td>-7.1 (+87%)#</td>
</tr>
</tbody>
</table>

Volume Fraction (Fuel/Na/Structure) = 23.4 / 57.3 / 14.6, ** 41.6 / 37.5 / 20.9

** Thickness of Axial Internal Blanket (Inner / Outer Core) = 7.4 / 3.8 cm

# Difference from Reference No-Uranium Core

reactivity in the actual reactor operation because of temperature distribution, the adoption of internal blanket in the Pu burner cores seems worth pursuing furthermore.

Other advantage of this concept may appear as the alleviation of the power swing in the core region, because we could optimize the thickness of the internal blankets between inner and outer core regions independently.

Finally, we surveyed the validity of this axially heterogeneous concept for various reactor sizes, core height, and fuel volume ratio. Figure 6 shows the results related to sodium void reactivity and Doppler reactivity. The internal blanket was found to play an important role for the improvement of reactivity characteristics in various types of non-uranium fueled core.

5. Concluding Remarks

The present study of Pu burning in unconventional fast reactor cores without uranium can be summarized as follows:
- The Pu consumption rates of the non-uranium cores are approximately 110 kg/TWhe, which is close to theoretical maximum.
- The other advantage of the non-uranium cores is extremely low sodium void reactivity.
- There are some concerns with the large burnup reactivity loss and the small Doppler effect. Although, these disadvantages seem to be within the range settled in the design-work elaboration, it is favorable to improve the reactivity characteristics by some creative idea.
- One possibility to improve the core characteristics would be the adoption of the
internal blanket axially in the non-uranium core. The advantage for the reactivity coefficients may exceed the degradation of Pu burning performance.

Finally, general R&D themes to substantiate the efficient Pu burner concept would be considered as:
- Improvement and optimization of the core concept related to reactivity coefficients, power swing, reactor core size and operation length per cycle,
- Verification of safety characteristics including transient condition,
- Accumulation of material property, especially, of fuel with diluent,
- Compatibility with fuel cycle such as fuel fabrication and reprocessing,
- and, investigation of reactor physics by critical experiment and analysis.

REFERENCES

Pu BURNING IN FAST REACTOR CORES USING UNCONVENTIONAL FUEL WITHOUT U-238

G.G. BAIBURIN
ARSRIIM, Moscow

A.P. IVANOV, I.Yu. KRIVITSKI,
V.I. MATVEEV, E.V. MATVEEVA
SSC IPPE, Obninsk

Russian Federation

Abstract

Use of fast reactors for actinide burning could serve as rather effective option of common problem solution of nuclear power long-lived waste activity reduction. Analysis of fast reactor possibilities for such objectives shows the expediency of use of specialized cores (or specialized fast reactors), fuel of which contains actinides without U replaced by inert matrix. Concept of such a core conformably to the BN-800 reactor type is considered in the paper.

Results of calculations of basic physics parameters of two reactor core options are given: for plutonium burning and for minor actinides burning. Means to improve power distribution flattening are shown. Assessment of reactivity effects are given. Issues of safety provision for cores under consideration are discussed.

1. INTRODUCTION

Fast reactors can be used efficiently for burning long-lived actinides contained in the spent fuel of thermal reactors which are working within the open fuel cycle. Plutonium is one of the most dangerous elements in the spent fuel actinide group from ecological point of view. Utilization of plutonium, which has been accumulated and is still being accumulated as a result of NPP (nuclear power plant) operation is one of the main tasks within the frame of general problem of activity decrease of the long-lived wastes of the nuclear power. Along with the NPP plutonium, the use of military plutonium can be considered, although it has of course some specific character. Another side of this problem is the necessity of burning minor actinides: americium, neptunium, curium, which will determine nuclear power wastes activity long time after the plutonium has been extracted.

Actinide burning is possible in the cores of existing LMFBR's (Liquid Metal Fast Breeder Reactors), although it would be advisable to make some changes in the core design (e.g., fertile blanket elimination, fuel enrichment increase, etc.) in order to improve the efficiency of the process. However, actinide burning can be carried out more perfectly in the fast reactor core using new fuel type in which U$^{238}$ is replaced by inert matrix. The expediency of U$^{238}$ elimination is obvious
because it is actinide supplier under irradiation. The main results of such core physics study made conformably to the BN-800 reactor design are presented below.

2. FUEL CORE CONCEPT AND BASIC DATA FOR CALCULATIONS.

At present various fuel compositions without U\(^{238}\) on the basis of MgO, Al\(_2\)O\(_3\), AlN and others are considered. The production of fuel pellets - fuel pins - from this materials is based on technology of powder metallurgy. Here we briefly consider another approach to the obtaining of such compositions, based on utilization of porous materials, saturated by fissile isotopes. We can consider ZrC, C, Ti and others as such materials. At present the most studied material is composed material on the basis of zirconium carbide with porosity from 50 to 80 %, saturated by plutonium oxides. This material was developed by All-Russia Scientific Research Institute of Inorganic Materials named after Academician A.A.Bochvar (ARSRIIM).

It is supposed that technology of introduction of other actinide oxides (Np, Am, Cm) will not bring principal difficulties.

It can be expected, that porous ZrC framework weakly interacting with neutrons, provides fuel column integrity under high burn-up of fissile isotopes. The technology of actinide oxides introduction is based on utilization of the following compositions:
- colloid solution of the plutonium dioxide;
- metal-organic composition.

The methods of obtaining of colloide oxide solution are sufficiently developed and used in Zol-Gel processes of granulated MOX (Mixed Oxide) fuel production.

The technology of production of metal-organic composition, containing plutonium was developed and standed the test under real manufacture conditions.

Re-generation of irradiated materials on the basis of PuO\(_2\)-ZrC can be carried out on the traditional mode of nuclear fuel reprocessing which is based on extraction technology of radiochemical industry without changes of existing scheme.

The main characteristics of fuel material are presented in Table I.

\(U^{238}\) replacement by inert matrix in fast reactor fuel will result in certain after-effects, the main of which are as follows.

1. An increase of reactivity change rate with the fuel burn-up, giving rise to the problem of maintenance of required refueling intervals.
2. Considerable increase of the total nonuniformity of the core power profile as a result of power rate difference between fresh and irradiated subassemblies. This also gives appreciable dilution of the coolant outlet temperature.
3. An increase of neutron leakage portion in the general neutron balance, leading to the decrease of sodium void reactivity effect (SVRE) and to the corresponding reactor safety improvement.

4. Considerable reduction of Doppler connected negative component of temperature and power reactivity effects which would apparently require some special measures in order to intensify negative reactivity feedback.

Table I  THE MAIN CHARACTERISTICS OF FUEL MATERIAL

<table>
<thead>
<tr>
<th>Characteristics</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Porosity, %</td>
<td>80</td>
</tr>
<tr>
<td>Mass of inert matrix in fuel element, kg</td>
<td>0.0353</td>
</tr>
<tr>
<td>Effective density of inert matrix in fuel element, kg/m³</td>
<td>1340</td>
</tr>
<tr>
<td>Coefficient of linear expansion of inert matrix, 10⁻⁶ 1/°C</td>
<td>6.73</td>
</tr>
<tr>
<td>Thermal conductivity, W/m °C</td>
<td>20.5</td>
</tr>
<tr>
<td>Melting point, °C</td>
<td>3500</td>
</tr>
</tbody>
</table>

Using the fuel not containing U²³⁸ to burn minor actinides makes it possible in general to provide criticality with any isotope of minor actinide group or with their mixture. However, this core design is hardly feasible because of too high positive SVRE and low effective delayed-neutron fraction values.

The above mentioned unfavorable consequences of utilization of the fuel containing inert matrix instead of U²³⁸ should be neutralized or alleviated at the core design development stage.

Two problems have been considered and solved in the course of studies, which results are presented below. One of these problems is determined by the high nonuniformity of power rate in different subassemblies, the other one - by the necessity of taking into consideration physical safety criteria when choosing fuel composition for the minor actinide burning.

The first problem can be solved by means of transposition of fuel subassemblies from the periphery to the center of the core at the refuelling. This provides fissile isotopes concentration distribution along the core radius that makes power rate profile more flat. This flattening extent depends on set fuel burn-up, and it can correspond more or less to the core power profile needed. Anyhow, this method of flattening is rather efficient and it provides acceptable average core power rate value. Along with the assembly transposition the necessary coolant flow rate distribution adjustment is provided in accordance with the subassembly power rate (in definite range). It should be noted that the subassembly transposition was carried out during normal refueling of the BN-600 reactor core.
Table II  THE MAIN CHARACTERISTICS OF THE REACTOR AND CORE UNDER CONSIDERATION

<table>
<thead>
<tr>
<th>Characteristics</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 Reactor thermal power, MW</td>
<td>2100</td>
</tr>
<tr>
<td>2 Efficiency, %</td>
<td>38</td>
</tr>
<tr>
<td>3 Maximum linear power of fuel element, kW/m</td>
<td>500</td>
</tr>
<tr>
<td>4 Average core specific power rate, MW/m³</td>
<td>400</td>
</tr>
<tr>
<td>5 Maximum fuel burn-up, % h.a.</td>
<td>50</td>
</tr>
<tr>
<td>6 Core subassemblies life-time, eff. days</td>
<td>300-400</td>
</tr>
<tr>
<td>7 Refuelling interval, eff.days</td>
<td>60-80</td>
</tr>
<tr>
<td>8 Breeding ratio</td>
<td>0</td>
</tr>
<tr>
<td>9 Sodium void reactivity effect, % Δk/k</td>
<td>0</td>
</tr>
</tbody>
</table>

The second problem is connected to the necessity of meeting physical safety criteria when providing efficient minor actinide burning. Utilization of fuel composition based on plutonium and inert matrix results in considerable SVRE reduction (down to approximately - 2 % Δk/k) This allows some amount of minor actinides to be added into the fuel composition providing for example zero SVRE in the core. The best results can be obtained using fuel composition made on the base of U^{235} and minor actinides. This provides acceptable range of effective delayed-neutron fraction. On this base an acceptable fraction of minor actinides in the fuel can be determined.

Calculations were made for the cores containing usual fuel elements, subassemblies and control rods according to the BN-800 reactor design[1]. Table II shows the main characteristics of the reactor and core under consideration.

Preliminary calculation results have shown that satisfactory flattening of the core power profile can be achieved at the core refueling ratio of 5. In accordance with this, 5 annular sub-zones are provided each containing equal number of subassemblies, which are placed in succession in all these sub-zones during refueling operations or subassembly transpositions from the periphery to the center of the core.

In order to improve power rate distribution flattening two-zone core design is considered with different plutonium content in the subassemblies of both peripheral and central sub-zones (this core is similar to that with two fuel enrichment zones made to achieve required power rate distribution).
Fuel subassembly transpositions similar to those mentioned above are carried out during refueling in each sub-zone. The main characteristics of core types under consideration are presented in Table III.

Figs. 1 and 2 show 2D calculational models for two core types.

3. CALCULATION RESULTS

3.1. Codes

Codes used for calculations are as follows:

- RBR-80 [2]: 2D diffusion code with burn-up calculation module - for isotope concentration, power profile, fuel balance and SVRE calculations;
- MMCFK[3]: 3D code, using Monte-Carlo method for Boltzman kinetics equation solution, for more accurate calculation of SVRE and control rod worth.

ABBN-78[4] Russian constant system together with ARAMACO-C1[5] nuclear data preparation system were used in the codes.

<table>
<thead>
<tr>
<th>Characteristics</th>
<th>One zone core (type 1)</th>
<th>Two zone core (type 2)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. Fuel subassemblies number in the core including</td>
<td>565</td>
<td>565</td>
</tr>
<tr>
<td>- central</td>
<td>565</td>
<td>175</td>
</tr>
<tr>
<td>- peripheral</td>
<td>390</td>
<td></td>
</tr>
<tr>
<td>2. Subassemblies number in the radial blanket including:</td>
<td>384</td>
<td>384</td>
</tr>
<tr>
<td>- steel subassemblies</td>
<td>186</td>
<td>186</td>
</tr>
<tr>
<td>- boron shield subassemblies</td>
<td>198</td>
<td>198</td>
</tr>
<tr>
<td>3. Control and safety rods, including:</td>
<td>30</td>
<td>30</td>
</tr>
<tr>
<td>- shim rods (SHR)</td>
<td>16</td>
<td>16</td>
</tr>
<tr>
<td>- control rods (CR)</td>
<td>2</td>
<td>2</td>
</tr>
<tr>
<td>- safety rods (SR)</td>
<td>9</td>
<td>9</td>
</tr>
<tr>
<td>- passive safety rods (PSR)</td>
<td>3</td>
<td>3</td>
</tr>
<tr>
<td>4. Core height, m</td>
<td>1.05</td>
<td>1.05</td>
</tr>
<tr>
<td>5. Axial steel blanket height, m</td>
<td>2*0.3</td>
<td>2*0.3</td>
</tr>
</tbody>
</table>
3.2. Neutron Spectrum

Estimated index values, characterizing neutron spectrum in the core with zirconium carbide base inert matrix and without $^{238}\text{U}$ are represented in Table IV compared to mixed fuel core. Average microscopic cross-sections of various isotopes have been chosen as these indexes.

Represented data show some neutron spectrum features of the core where $^{238}\text{U}$ is replaced by Zirconium carbide base inert matrix, i.e. considerable spectrum softening at the lower part of energy range and some its hardening at the upper part - for the fuel containing minor actinides.

3.3. Critical Parameters

Specific content (density) of fissile material in the fuel elements can be considered as critical parameter for the core having no $^{238}\text{U}$. This parameter

<table>
<thead>
<tr>
<th>Table IV</th>
<th>AVERAGE MICROSCOPIC CROSS-SECTIONS OF VARIOUS ISOTOPES (BARN)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Index</td>
<td>$\sigma_{f}^{\text{U}^{238}}$</td>
</tr>
<tr>
<td>Fuel having no $^{238}\text{U}$</td>
<td>0.042$^{**}$</td>
</tr>
<tr>
<td>Fuel with inert matrix</td>
<td>0.047$^{**}$</td>
</tr>
<tr>
<td>Traditional mixed fuel</td>
<td>0.046</td>
</tr>
</tbody>
</table>

$^{**}$ $\text{PuO}_2$, $^{**}$ $^{235}\text{O}_2 + 35\%$ minor actinide oxides

224
calculation results are represented in Table V for the following three types of considered core composition:
- plutonium oxide;
- plutonium oxide with 10% minor actinides addition;
- U\textsuperscript{235} oxide with 35% minor actinides addition.

NPP grade plutonium composition Pu\textsuperscript{239}/Pu\textsuperscript{240}/Pu\textsuperscript{241}/Pu\textsuperscript{242} - 60/25/10.9/4.1% has been used in analysis. For minor actinide mixture the following composition has been adopted: Np\textsuperscript{237}/Am\textsuperscript{241}/Am\textsuperscript{242m}/Am\textsuperscript{243}/Cm\textsuperscript{242}/Cm\textsuperscript{244} - 59.45/27.59/0.021/9.64/0.018/3.28%.

Minor actinide weight contents of 10% and 35%, indicated above, correspond to the core SVRE zero value. Calculations were made for the stable refueling procedures.

Table V

| SPESIFIC CONTENT OF FISSLIE MATERIAL IN THE FUEL ELEMENT |
|-----------------------------------------------|------------------|-----------------|------------------|
| Core fuel | Composition material on the base zirconium carbide impregnated with actinide oxide | PuO\textsubscript{2} | PuO\textsubscript{2} + 10% minor actinides | U\textsuperscript{235}O\textsubscript{2} + 35% minor actinides |
| Fissile material density (kg/m\textsuperscript{3})\textsuperscript{(*)} | 1280 | 1490 | 2060 |

\textsuperscript{(*)} Fuel element material mass material (heavy atoms) to the fuel element volume ratio
3.4. Reactivity Effects

3.4.1. Sodium Void Reactivity Effect

Sodium void reactivity effect was calculated for "before refueling" condition which is the most unfavorable for this parameter. Calculations were made in both diffusion and transport approximations. SVRE value for plutonium oxide version is -2.22 %Δk/k when calculated using RBR-80 diffusion program and -1.73+0.1%Δk/k when calculated by Monte-Carlo method using MMCFK. This significant SVRE decrease as compared to that of traditional fuel core, can obviously result from the neutron leakage fraction increase in the core having no U\textsuperscript{238}. If the fuel having minor actinide addition is used, the SVRE value is equal to zero, and this was chosen, as it has been indicated above, when minor actinide permissible fraction was determined.

3.4.2. Temperature Reactivity Effects, Reactivity Change Rate as a Function of Fuel Burn-up

Doppler effect value in the core having no U\textsuperscript{238} is sufficiently lower (almost 10 times) as compared to that of traditional mixed fuel core. One could suppose that effects related to the core size and shape changes would increase considerably in this core because of large neutron leakage rate. However, according to the estimates which have been made, change of K\textsubscript{eff} sensitivity in relation to the core size change has proved to be insignificant. Besides fuel material made on the base of porous zirconium carbide matrix has sufficiently lower thermal expansion coefficient in comparison with that of oxide fuel. For the same reason (U\textsuperscript{238} absence) reactivity change rate as function of the fuel burn-up is appreciably increased. Estimated values of the main reactivity effects for the plutonium oxide core are represented in Table VI.

<table>
<thead>
<tr>
<th></th>
<th>Axial expansion sensitivity coefficient Δk/k/m</th>
<th>Fuel axial expansion effect Δk/k /°C</th>
<th>Doppler constant T d(Δk/k)/dT</th>
<th>Reactivity change as function of fuel burn up during one month % Δk/k</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fuel without U\textsuperscript{238} with PuO\textsubscript{2}</td>
<td>1.57*10^-1</td>
<td>0.17*10^-2</td>
<td>0.0009</td>
<td>3.3</td>
</tr>
<tr>
<td>Traditional mixed fuel</td>
<td>1.35*10^-1</td>
<td>0.26*10^-2</td>
<td>0.007</td>
<td>0.72</td>
</tr>
</tbody>
</table>
Similar characteristics for the fuel with minor actinides addition are values of the same order of magnitude, although some additional Doppler effect decrease is possible in this case.

The reactivity change rate as a function of the fuel burn-up is also decreased considerably for this core type, making it possible to increase the refueling intervals in comparison to those for Pu oxide core (in our case - up to 80 eff. days). Thus, reactivity coefficients providing negative temperature and power feedback are sufficiently decreased in the core with no U\(^{238}\) content. Perhaps, in order to increase negative feedback special measures would be required, e.g. resonance absorbers insertion into the fuel.

Increasing reactivity change rate as function of the fuel burn-up rate results, at the given shim rods number in appreciable reactor refueling interval decrease. For the core design under consideration this interval has been decreased down 60 days when using Pu oxide, and - down to 80 days when using U\(^{235}\) oxide with 35% minor actinides addition.

It should be noted that the reactor operation under decrease refueling interval conditions can be realized, although this operation mode is not perfect from the point of view of power production.

There are some ways within the frame of the design to increase fuel burn-up compensation system efficiency by increasing boron carbide enrichment value (from 60% up to 80% or 90%) and by means of functions redistribution between separate rod groups. In general, acceptable operation interval provision for the nuclear power reactor using fuel without U\(^{238}\), is a problem, which has to be solved in the further design studies.

3.4.3. Core Power Profile Uniformity

The problem of keeping core power profile non-uniformity within the permissible limits can be solved by means of fuel subassemblies transposition from the periphery of the core to the center of the core during refueling. In the cases under consideration maximum fuel element linear power values are in the range of 440-500 kW/m. When moving subassemblies the necessary coolant flow redistribution should be also provided in accordance with the power rate.

3.4.4. Efficiency of Actinide Burning

Actinide burning can be carried out in the most efficient way if the fuel without U\(^{238}\) is used in the core. Analysis has shown that using such core in the BN-800 type reactor, up to 600 kg of NPP grade plutonium can be burnt during 1 year with Pu oxide fuel and - up to 250 kg of minor actinides (Np, Am, Cm) with U\(^{235}\) and minor actinides mixture fuel.
CONCLUSION

Studies made for the BN-800 type reactor have shown principal feasibility of the core with the fuel containing inert matrix instead of U$^{238}$, although some parameters of this core are different from those adopted in the reactor design.

Detailed analysis is needed to provide reactor safety because of sufficient weakening of the temperature and power feedbacks, and this will obviously lead to special measures development aiming at the reactor safety increase. Reactor refueling interval reduction is also the disadvantage of this core, although some margin exists in the real BN-800 design for its increase (absorber rod boron carbide enrichment increase, functions redistribution between control and safety rod groups).

The possibility of the most effective burning of highly active long-lived actinides, accumulated as a result of nuclear power engineering activity, is the simulating factor for the further studies and development of the core without U$^{238}$ content.

REFERENCES


MONONITRIDE MIXED FUEL FOR FAST REACTORS

B.D. ROGOZKIN, N.M. STEPENNOVA, Yu.E. FEDOROV, M.G. SHISHKOV, O.N. DUBROVIN, L.V. ARSEENKOV
All-Russia Scientific and Research Institute of Inorganic Materials, Moscow, Russian Federation

Abstract

As regards the property combination, methods of fabrication, reprocessing and irradiation behaviour mononitride fuel is considered to be most promising (like monocarbide and carbonitride) for inherently safe fast reactors.

One of the most important properties of mononitride fuel is its creep. The studies carried out in Russia established the dependence of creep on composition, stresses, temperature and fission number.

Methods of mononitride fuel fabrication use as source materials oxides and metals, weapon's grade plutonium included. The method of fuel preparation from metal is most simple in its instrumentation and technology and results in more pure (as far as impurities are concerned) mononitrid.

In-pile BR-10, SM-2, MIR and BOR-60 reactors tests of mononitride fuel revealed its adequate compatibility with structural materials, lower gas releases compared to oxide fuel, no caesium or iodine induced corrosion and feasibility of burn-up more than 9% h.a. at the heat generation rate of 400-800 W/cm.

Studies of hydrometallurgical (Purex-process) and electrochemical molten salt methods of regeneration using unirradiated mononitride fuel indicated their promising character and feasibility.

1. INTRODUCTION

The mononitride fuel (as well as carbonitride one) possess unique set of valuable for reactor fuel properties—high concentration of fission elements, thermoconductivity (that increases with temperature rising), high melting point, good compatibility with structural materials and coolants (sodium, sodium-potassium alloy, lead, lead-visitmut alloy etc.), the possibility of different ways of reprocessing etc. That is why this fuel is considered as most advanced one for fast reactors and space reactors /1,2/. In Russia the problem of development and use of mononitride fuels is solved on the broad and carefully planned scale. The investigation programme includes the following main items:

- investigation of physical and chemical properties of UN, UPuN, UN-UC, PuC, UPwC, UPuCN in pre- and in-reactor conditions;
- the development of fuel synthesis methods from initial metals and oxides and fabrication of fuel that possess specified density, porosity, impurity content, form, sizes and compositions;
- the investigation of compatibility of mononitride fuel with steels and coolants in pre- and in-reactor conditions.
- the development of the express methods for elements containment in fuel composition checking;
- fuel pins and subassemblies design;
• irradiation of MeC, MeN fuel pins and subassemblies in BR-10, CM-2, MIR, BOR-60 and BN-350 reactors;
• the electrochemical and hydrometallurgical reprocessing methods investigation.

Putting into the practice of the mononitride fuel is aimed to the building of the commercially profitable closed fuel cycle for the inherently safe fast reactor. It should be noted that the mononitride fuel investigations are performed simultaneously with the investigations of carbide and carbide fuels. Some deficiency of the mononitride fuel connected with the catching of neutrons in reaction $^{14}\text{N} (n,p) ^{14}\text{C}$ can be eliminated by using of $^{15}\text{N}$ isotope.

The investigations of mononitride fuel UPuN was preceded by UN fuel investigation. The main results are presented below.

2. CREEP

The creep ($\varepsilon$) investigation was conducted in complete range of uranium carbide-nitride compositions for temperature and stresses changing in range 1200 — 1873 K and 10 — 80 MPa, correspondently. There were found two extreme points - minimum creep rate for composition UCN and maximum creep rate for composition UC$_0.7$N$_0.3$ The values of whose rates are 2·10$^{-1}$ and 5·10$^{-1}$ correspondently for temperature 1580 K and $\sigma$=20 MPa /4/.

Steady state creep rate of uranium mononitride for stresses higher than 40 MPa does not depend on grain size and may be described by equation

$$\varepsilon = \sigma^3 \exp (-Q/RT);$$

where $\sigma$ — stress, [MPa];

$Q = 326570$ J.

The uranium mononitride U$_{0.8}$Pu$_{0.2}$N creep rate depends on oxidic and carbide content:

a) For oxidic and carbide content in the range of 0.2—0.3%:

$$\varepsilon = 35 \sigma^{1.27} \exp (-Q/RT);$$

b) For oxidic and carbide content less than 0.15%:

$$\varepsilon = 308\sigma^{1.35} \exp (-Q/RT);$$

where $\sigma$ = 10 — 60 MPa; $Q = 40000$ kal.

A facility for irradiation creep investigation in IRT-2000 and BR-10 reactors have been created.

The results of irradiation creep investigation have shown that the creep rate was ten times higher than that for preirradiating testing for temperature 1000-1800 K. Conventional temperature boundary, where thermal creep is observed, depends remarkably on fission density in the specimen. So, as the fission density changes from $1.0\cdot10^{13}$ cm$^{-3}$ to $3.6\cdot10^{13}$ cm$^{-3}$ the boundary lowers from 1500K to 1250K.
3. THE URANIUM AND MIXED URANIUM-PLUTONIUM MONONITRIDE FABRICATION PROCESS

3.1. Equipment

In Russia there was built a number of devices and facilities for investigation and fabrication of small amount of uranium carbide-nitride fuel and mixed uranium-plutonium carbido-nitride fuel. Some facilities make possible to fabricate uranium mononitride fuel for the whole core of fast BR-10 reactor loading or for full-scale experimental subassembly of BN-350 reactor. Those facilities are a number of connected in one line glove boxes with implemented experimental and technological devices. The cells are filled with dry inert gas. The fabrication process of mononitride fuel depends in great extent on the method that have been choosen for irradiated fuel reprocessing and on the type of final product for repeated use in the closed fuel cycle. It is nessesary to take into account the existing supply of oxide raw materials and accumulated metallic plutonium too.

Therefore facilities for mononitrides fabrication from initial oxides and metallic uranium and plutonium were created. For uranium mononitride fabrication from initial uranium the facility of continiuos operation type was built. This facility have productivity about 1.5 kg per hour and is a prototype one for fabrication of mixed nitrides from initial metals. There are designed and put into operation facilities for fabrication of microspherical uranium mononitride fuel by plasma melting of powder and by centrifugal-arc method.

It is nessesary to note that when the new data concerning the physical and mechanical properties of mononitride fuels or modified design of fuel pins and subassemblies appear we can determine new requirements to quality, dessity and size of mononitride columns /2-8/.

3.2. Fabrication of nitrides from oxides

It was stated that for carbothermical method of the uranium and plutonium mononitrides fabrication one can use initial oxides itself as well as jointly precipitated uranium and plutonium oxides. The amount of carbon added in mixture depends on oxigen coefficient and on experimentally choosen value of technological surplus that is usually in range of 0.5—2%. Initial powder mixed in mills or mixers of different type is subjected to pressing with pressure value equal to 100-300 MPa and are slowly heated up to 1400 °C in vacuum or argon and then in chemically pure nitrogen atmosphere. The duration of treatment with temperature 1650 - 1850 °C can be from 2 to 10 hours depending on activity of initial substances. The cooling of mixture is conducted in the inverse order. For this process accomplishing the furnaces of permanent and periodical operation are designed (for UN). Those furnaces can operate with suspeded granulated mixture. The fabricated mononitrid is analysed and its phase composition is defined.

The finally obtained porous mass is grilled and pounded until the powder size not exexeds 40 μm.

The mononitride powder is mixed with binding substances (ethylene glycol, paraffin, polyvinil acetate, zinc or sodium stéarate etc), and pressurised in fuel stack form with pressure value of 100-300 MPa. This stacks are sintered in nitrogen at 1550-1750 °C. The temperature and duration of the sintering depends on required density.
The described scheme of UN and UPuN fabrication ensures the oxygen and carbon content less than 0.25%. In Russia there achieved the level of about 500 kg per year production of uranium mononitride fuel from oxides.

For production capacity examination of developed method there was manufactured mixed mononitride fuel with plutonium mononitride content equal to 30 and 35%. It was stated that the temperature level can be lowered by 50 — 60 °C. Those results are well correlated with that obtained by Indian specialists.

There was performed technological research on synthesis of solid solutions UN-ZrN and fabrication of fuel columns with different density, shape and size. The carbothermical reduction makes possible to product the solid solutions of full variety of composition. The syntheses temperature of solid solutions is equal to 1800 — 2100 °C.

There is developed the method of synthesis from oxides of solid solutions Pu0.55 Zr0.45 N.

3.3. The fabrication of mononitride fuel from initial metals

There was conducted in Russia the complex research and development work for synthesis of uranium and plutonium mononitrides from initial metals and following fabrication of fuel columns with different values of density.

This method makes possible to organize the low temperature process of synthesis at the temperature 200—650°C. As initial substances one can use initial uranium or plutonium as well as their alloy. Initial metals or alloys are hydrogenated by hydrogen at temperature 100—220°C and resulting powders are nitrided by nitrogen at the temperature 200—650°C. The process is stable and lead to uranium nitride and mixed uranium and plutonium nitrides formation.

The uranium nitride fabrication can be made on the permanent operation type facility.

The uranium and plutonium nitrides are powders with grain size 4—100 μm. The grain size depends on fabrication duration and temperature.

It was stated that nitride powders are suitable for uranium and plutonium mononitride fuel columns of different density fabrication in spite of rather high nitrogen content. For fuel column fabrication cold pressing with pressure 100-500 MPa and following sintering at 1550—1750 °C in argon or in argon-nitrogen mixture are used. A number of different sintering types are developed. Resulting monophase mononitrides fuel columns have a specified grain size and density. In any case the even distribution of plutonium through the fuel volume is provided (the deviation is less than 1%). The impurity (C, O) content in mononitride depends on initial substances purity and does not exceed 0.15%. By means of this method there were manufactured experimental bunches of fuel columns for irradiation in BR-10, SM-2, MIR, BOR-60 and BN-350 reactors.

3.4. The analysis facilities

For the fuel analysis ensuring there was developed for all stages of fabrication the set of analysis methods such as chemical, neutron, rentgen, р-radiographical, metallographical and microrentgen analysis. There have been built the foundation of a neutron method for oxygen and nitrogen content determination in fabricated mononitride fuel pins. For gas phase analysis the chromatographs are used.
4. IRRADIATION TESTS OF MONONITRIDE FUEL IN RUSSIA

Irradiation tests of mononitride fuel have been began from the late sixties in SM-2, MIR, BR-10 and BOR-60 reactors. The aim of irradiation tests was as following:

- the irradiation properties of mononitride fuel (swelling, gas release, fuel-cladding and fuel-coolant compatibility etc) investigation;
- the high burn-up fuel pin design and fuel core density optimisation;
- the investigation of fuel fabrication technology (impurity content) influence on the irradiation properties of fuel.

The main irradiation tests results:

1. All uranium and mixed mononitride fuel pins that achieved the burn-up level of 4.0—9.0% with the linear power 1045—600 W/cm was intact. Fast neutron fluence \( (E > 0.1) \) was \( 4.38 \times 10^{22} - 4.54 \times 10^{22} \text{ cm}^{-2} \). The irradiation time was ranged from 0.5 to 4—5 years. Neither iodine nor caesium corrosion was detected. The maximum fuel pin cladding deformation did not exceed 1.4%. The fuel-sodium and fuel-lead interaction was not found.

2. The mononitride fuel swelling depends on oxygen and carbon content, core center temperature and fuel density. For gas bonded fuel pins with fuel density equal to \( \approx 92\% \) of theoretical value, cladding diameter 5.9 mm and center temperature 1440—1470 °C the oxygen and carbon content increasing from 0.15 to 0.4% leads to swelling rate growth from 1.2—1.5% per 1% burn-up to 1.9—2.5% per 1% burn-up. For the fuel with density 88—90% TD and with oxygen and carbon content less than 0.15% the core center temperature growth up to 1800—2200 °C caused the swelling rate increases to 1.5—1.7% per 1% burn-up. The tendency to swelling rate decrease of UN and U Pu N fuels was noted when the fuel density decreased from 94% to 85—89% TD. The swelling of mononitride fuel with density 92% TD in sodium-bonded fuel pins did not exceed 1.0—1.1% per 1% burn-up for center temperature 1000—1100 °C.

3. Gas fission product (GFP) release depends on oxygen and carbon content, fuel density and core center temperature. The gas release from MeN fuel in the He-bonded pins was 2—4 times lower than that from oxide fuel and was equal to 25—50%. The GFP release from mononitride fuel increases 1.5—2 times when the oxygen and carbon content increases from 0.15% wt. to 0.3—0.4% wt. for the same irradiation conditions.

GFP release from mononitride fuel increases from 23—25% to 40% when the core center temperature increases from 1300—1450 °C to 1850—2200 °C.

4. The mononitride fuel keeps the structural and dimensional stability better than monocarbide one.

\( \alpha \)-radiography and microrentgen analysis results show that there is no visible plutonium redistribution along the axial and radial directions in the fuel columns. The amount of released cesium not exceeds 5%.

The fuel-cladding interaction zone did not exceed 110 m after 2—4 years of irradiation in BR-10 and BOR-60 reactors and has the local character. Mainly it was a zone of increased etching. In that zone there was noted the increasing of carbon content. Besides when the mononitride fuel contained less than 0.15% wt. of oxygen.
and carbon, the carbon concentration in interaction zone was 2—3 times less than for fuel with carbon content 0.3—0.4%.

5. The irradiation tests make possible to conclude that it is worth while to use fuel with density value equal to 85—89% TD and maximal open porosity for the He-bonded pins, and with density 92—95% TD for liquid metal bonded pins.

For the good performance of mononitride fuel pins with gelium gap it is nessessary to decrease oxigen and carbon content to 0.15% wt. and never exceed the value 800—850 W/cm for the linear power rate.

5. THE MONONITRIDE FUEL REPROCESSING

There are two ways of the mononitride fuel reprocessing that are developing in Russia:

a) Hydrometallurgical reprocessing (PUREX-process);

b) Electrochemical reprocessing in molten halides.

The resulting product of hydrometallurgical reprocessing are oxides and of electrochemical reprocessing - metals.

5.1. Hydrometallurgical reprocessing of mononitride fuel

There was finished the development of first stage of hydrometallurgical reprocessing of nonirradiated uranium and mixed mononitride fuel with density 85—95% TD.

After a pin cladding removing the mononitride fuel is dissolved in nitric acid. It was found that the dissolving rate for mononitriedes is essentially higher than the for oxides and the dissolution process is more stable. The dissolution rate of uranium mononitride can be approximated in the following way:

$$V=0.4885+0.2\cdot10^{-5}\cdot T^2 [\text{HNO}_3]+0.19\cdot10^{-2} [\text{HNO}_3]-2.03[\text{HNO}_3]^2,$$

where $V$ - the dissolution rate, g/min.-cm$^2$; $T$ - the temperature, K; $[\text{HNO}_3]$ - nitric acid concentration, mole/l.

This dependency can be used in the following range of nitric acid concentration and temperature:

- $4.0—12.4$ mole/l for $T=365 — 363$ K;
- $6.0—12.4$ mole/l for $T=333 — 313$ K.

The further operations can be performed in the same way as for oxide fuel reprocessing.

5.2. Electrochemical reprocessing of mononitride fuel in molten halides

The development of electrochemical reprocessing of nonirradited mononitride fuel in molten halides was initiated in Bochvar Institute /10/. This method make possible to keep in the produced fuel the minor actinides for their following burning in reactors. There was investigated the following items: electrolyte composition (40—45% KCl, 40—45% LiCl, 10—16% UC13), the method of uranium chloride intrusion in the electrolyte, anode dissolution of uranium mononitride.
Electrolyse was conducted in an electrolyser built in an air tight box containing argon. It was shown that a circonium, molibden, rutenium remain in the sludge of anodic part of electrolyser.

A number of experiments for electrochemical dissolution of uranium mononitride was conducted. In those experiments there were obtained about 500 g of uranium in the form of dendritic shaped crystals. The cathode deposit (70% of uranium and 30% of electrolyte) was melted, electrolyte was send to repeated electrolysis and the uranium to refining melting and further fabrication of mononitride fuel.

The oxygen and carbon content in fabricated and initial fuel was practically the same.

The investigation in Bochvar Institute have showed that in principle the electrochemical reprocessing of mononitride fuel and following building of closed fuel cycle for fast reactors on the basis of mononitride fuel fabrication and electrochemical reprocessing is feasible.

5.3. The feasibility of $^{15}$N utilisation

Both the hydrometallurgical and electrochemical reprocessing permit the $^{15}$N utilisation. The catching of $^{15}$N during the hydrometallurgical reprocessing can be performed on the stage of preliminary oxidation of the mononitride.

During the electrochemical reprocessing the nitrogen is emitted as a gas on the stage of anodical dissolution. This gas can be easily bind into uranium or another element nitrides that desintegrate after heating and release the gaseous nitrogen. That nitrogen can be send to repeated nitriding.

Fig. 1 presents principle diagram of fuel cycle using hydrometallurgic and electrochemical reprocessing.

**CONCLUSION**

1. UC-UN, UPuN have been investigated at 1200—1873K and stresses 10—80 MPa.

2. The methods of UC, UN, UCN, UPuC, UPuN, UZrC, PuZrC fabrication have been developed from the oxides, metals, including weapons-grade plutonium.

3. Radiation tests of UN, UPuN, have been conducted in the reactors SM-2, MIR, BOR-60, BR-10 up to burn up ~9% h.a. at power rating 400—1000 W/cm, which demonstrated high operational reliability with MeN-containing fuel elements.
4. It has been found that hydrometallurgical (PUREX) process of non-irradiated UC, UN, UPuC, UPuN reprocessing can be carried out at the operating apparatus-technological line of the PO "Mayak" RT-plant.

5. The experiments held at the VNIINM demonstrated a potentiability of non-irradiated MeN reprocessing by electrochemical technique in molten salts.

6. A flow sheet of closed fuel cycle has been offered.

REFERENCES


PLUTONIUM DISPOSAL AND BURNING IN LEAD COOLED FAST REACTOR

V.V. ORLOV
Research and Development Institute of Power Engineering, Moscow, Russian Federation

Abstract

Utilization of accumulated plutonium will take the decades and this problem should be considered in connection with the long-term prospects of nuclear power. The paper addresses the requirements to the nuclear technology which can for the large-scale development, reasons and results of the conceptual design of lead cooled Fast Reactor.

1. INTRODUCTION

Irradiated fuel from NPPs contains about 1,000 t of the fission Pu; this amount will be increasing by 50-100 t/y, mostly in the spent fuel pools. In France, UK and Russia large radiochemical reprocessing plants are in operation which at full capacity can separate about 20 t of R-Pu per year from the NPP fuel.

Reduction of nuclear weapons will add about 200 t of W-Pu which should be considered together with R-plutonium (for example, it can be mixed with R-Pu). Apart from Pu, one should bear in mind about 2,000 t of HEU from the nuclear weapon which can be diluted to reach X<20% enrichment.

This is a very valuable fuel close in heat value to 10 bln. t of high-quality coal. Accumulation of high-toxic Pu, increasing the risk of proliferation, brings about the desire of many people to get rid of it as fast as possible.

However, this cannot be done fast enough, because preparation and implementation of any option of Pu disposal or utilization will take decades, incurring considerable expenses. First of all, it is necessary to construct facilities for long-term safe Pu storage. The construction costs will constitute the major part of the storage discount costs, hence reducing the incentives for fast emptying of the storage facilities.

Finally, utilization of Pu and HEU will not be a cost-effective option in the near decades, with sufficient resources of U, excess capacities for its production and enrichment and low U market prices. According to some estimates, depletion of rich ores and higher U prices may be expected by the second quarter of the next century, and only then serious economic incentives will appear for using Pu and HEU and closing the fuel cycle. (Large NPP fuel reprocessing plants were constructed in some countries in the 1970-1980s awaiting rapid growth of the world's nuclear power and U prices, construction of fast reactors. However, due to man causes, all these fell short of the expectations.)

Hence, there are sound arguments for considering utilization of Pu (and, probably, HEU) within the framework of long-term development of nuclear power. Loss of part of Pu (\(^{241}\)Pu) in long-term storage and accumulation of \(^{241}\)Am would hardly be treated as a decisive argument in favor of rapid utilization.
Today, the predominant opinion deals with long-term nuclear power evolution on a moderate scale using conventional reactors, largely, LWR. Report "Energy in the Tomorrow's World" presented at the XYWEC Congress (Madrid, 1992) forecasts doubling of nuclear capacities by 2020 which will still account for 6% of the primary energy produced in the world. In this case, no extra fuel will be needed soon.

However, the situation may develop in another way. The above-mentioned report indicates inadequate power supply for the major and rapidly increasing part of the Earth population which is also confronted with the problem of environment at degradation. Even now, the nonuniform distribution of conventional fuels is one of the contributors to the international tension, and by the end of the considered period, depletion of oil and gas resources will also manifest itself. These problems will become more serious with more and more countries getting involved into industrial development.

Seeing no real radical changes in the structure of the world's energy production, the authors of the report urge the government to support the development of new energy technologies right now, not waiting for 20 or 30 years, to make them viable commercially and technologically.

The fission nuclear power undoubtedly has the right potential for radical solution of the problems posed, and is the most prepared, among the new energy technologies, for a large-scale evolution.

This evolution may become the content of the next stage — "a second nuclear era", after A. Weinberg /1/. It depends on the solution of a number of problems, including fission fuel resources. The accumulated amounts of Pu and HEU can ensure the start of the next stage and shall be considered from this viewpoint.

2. LARGE-SCALE NUCLEAR POWER

Nuclear power can stabilize consumption of the conventional fuels, if by doubling its capacities every ~ 20 years it rises by an order by the mid of the next century, increasing its share in the world's primary energy production to at least about 1/3 as compared to the current 5% level. However, such a rise could hardly come as a result of technical evolution of conventional nuclear technologies. If we remain within the framework of these technologies, we cannot even convincingly justify such prospects.

LWRs and other thermal reactors consume too much uranium while fast reactors capable of solving the problem tuned out to be too expensive. But the most important thing is that the nuclear technology of the first stage matured 40 years ago on the basis of the experience and perceptions existed at that time has neither experimental nor theoretical grounds for such a long-term forecast of its safety.

The nuclear power experience constitutes $6 \cdot 10^3$ reactor-years, while the large-scale nuclear power would reach about $10^6$ reactor/years in the next century.

The proof for elimination of NPP nuclear catastrophes would require consideration of events with the probabilities of $10^{-6}$ and lower, which we usually do not know, the same as possible correlations between the events, unique events, especially associated with human activities, which are difficult to be estimated in terms of probabilities.
Recognition of nuclear power as a conventional technology for large-scale application in both developing and developed countries will need nuclear technologies satisfying new requirements. A new technology takes much time, but we can speak about its development and demonstration in 15-20 years, since we are aware of its basic components in the existing civil and military nuclear engineering and in recent developments. The discussion below refers essentially to a new evolutionary technology culminating the benefits of the first stage. Hence, the new stage in nuclear power may be launched as early as in the first decades of the next century.

3. REQUIREMENTS TO NEW NUCLEAR TECHNOLOGY

Above all, this concerns a convincingly proved prevention of NPP nuclear catastrophes. Certainly, we are not speaking in terms of an absolute safety. We can consider accident as prevented if its probability has been reliably estimated as much lower than $10^{-6}$ (for example, collision with an asteroid). Large-scale nuclear power is hardly possible without political measures adequate safeguarding against nuclear attacks on NPPs. However, one cannot ignore potential non-nuclear actions (acts of terrorism, missile attacks). These actions may be estimated at the maximum level and considered as initiating events for maximum accident calling for NPP design to prevent a catastrophic radioactive release.

As a limiting value, $10^4$ Ci in J equivalent can be adopted which is 1,000 times lower than the Chernobyl release and requires no extraordinary measures to protect population and the areas. If one is successful in proving that the release will remain within the above limits in case of an external effect resulting in maximum damage to NPP structures and systems damage, it would still be more valid for the accidents caused by internal effects.

With the catastrophes prevented, the problem of NPP safety becomes just a more ecological-economical problem. The probabilities of severe accidents of $10^{-3}$—$10^{-5}$ become permissible, as defined by the insurance against loss of NPP with limited radiation consequences. Such estimates may be considered as sufficiently reliable.

In a most general form, the requirements to the reactor consist the following: in case of a maximum accident, the fuel, coolant and reactor vault shall retain radioactivity releases within the specified limits /2/.

The deterministic safety requirements to radwaste management technology lie in observance of natural radiation equilibrium at their disposal which can be attained by transmutation and storage.

The problem of nonproliferation, in view of possible U enrichment, has no deterministic solution within the nuclear technology and demands political measures. The new nuclear technology shall contribute to implementation of such measures, making it more difficult to produce and recover Pu from the fuel cycle for further weapon application and facilitating the inspection.

To keep within known relatively cheap resources of uranium and current capacities of its production, its specific consumption should be reduced by an order as compared to current LWRs which corresponds to BR close to 1 or higher. Modern perceptions of power development do not require short Pu doubling time, which simplifies requirements to breeding, fuel power density, external fuel cycle and its duration.
Besides, generation of nuclear power shall remain economically profitable both for developed and developing countries. For reference, we may adopt the current LWR generation cost, though its reduction due to simplified design technology, requirements and licensing procedures would be of advantage.

The requirements seem to be contradicting and difficult to be fulfilled as a whole. However, safety and economy can be harmonized, if the inherent safety principles are applied as early as at the conceptual design stage.

The nuclear philosophy, the key words of which "inherent safety" have been introduced in wide use by A. Veinberg, has already influenced modern NPP designs. However, the potential hazards of fast runaway, loss of coolant, fires and explosions occurred at TMI and Chernobyl are inherent in modern reactors and cannot be eliminated completely. Engineered safety measures reducing the probability of such accidents have become the major cause of nuclear plant sophistication and higher costs.

To provide maximum safety together with design simplification and lower cost of NPPs the concept of the new nuclear technology shall be chosen based on this philosophy.
4. LEAD COOLED FAST REACTOR

That were the reasons why we started developing LCFR after the Chernobyl. The conceptual stage of the reactor is now close to completion. Besides calculational and preliminarily design efforts for 300, 600 and 1000 MW\(e\) reactors, the critical assembly experiments, steel corrosion tests on Pb-loops and some other tests were carried out /3,4,5/. Due to the characteristics inherent in LMFR (reactivity margin \(K \leq \beta_{\text{eff.}}\), feedback), lead coolant (chemical passivity, high density and boiling temperature, natural circulation), UN - PuN fuel (density, thermal conductivity, thermal-radiation stability) catastrophic course of severe accidents can be avoided.

Fig 1, 2 presents a design scheme of BREST-300 reactor in metallic vessel, Fig. 3 presents a design scheme of BREST-300 reactor in reinforced concrete pool. Fig. 4 shows the reactor core structure. Table 1 shows parameters of performance of lead-cooled fast reactor with loading R-Pu. W-Pu loading in BREST-300 reactor needs to rise reactor core height to 1400 mm, W-Pu loading in BREST-600 need to rise the density of mononitride mixed fuel until 0.95 TD with equal geometry of core. Table 2 shows reactivity effects and coefficients for BREST-300 reactor (R-Pu), for W-Pu loading this effects are like.

![Diagram of BREST-300 Reactor](image)

**FIG. 2. Cross-section over header branches.**
FIG. 3. Reactor BREST-300 pond type.

FIG. 4. Cross-section of BREST core.
### Table 1  Performances of the lead-cooled reactors

<table>
<thead>
<tr>
<th>Performance</th>
<th>BREST-600</th>
<th>BREST-300</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermal output, MWt</td>
<td>1400</td>
<td>700</td>
</tr>
<tr>
<td>Electric output, MWt</td>
<td>600</td>
<td>300</td>
</tr>
<tr>
<td>FA number in core</td>
<td>357</td>
<td>185</td>
</tr>
<tr>
<td>Reactor core diameter, mm</td>
<td>3190</td>
<td>2300</td>
</tr>
<tr>
<td>Reactor core height, mm</td>
<td>1100</td>
<td>1100</td>
</tr>
<tr>
<td>Fuel element diameter, mm</td>
<td>9.0; 9.6; 10.4</td>
<td>9.0; 9.6; 10.4</td>
</tr>
<tr>
<td>Fuel element pitch, mm</td>
<td>13.6</td>
<td>13.6</td>
</tr>
<tr>
<td>Reactor core fuel</td>
<td>UN+PuN</td>
<td>UN+PuN</td>
</tr>
<tr>
<td>Fuel loading (U+Pu)N, t</td>
<td>28</td>
<td>16</td>
</tr>
<tr>
<td>Loading Pu/(^{239})Pu+(^{241})Pu, t</td>
<td>3.73/2.72</td>
<td>2.2/1.6</td>
</tr>
<tr>
<td>Fuel lifetime, years</td>
<td>5—6</td>
<td>5</td>
</tr>
<tr>
<td>Time between loadings, years</td>
<td>1—2</td>
<td>~1</td>
</tr>
<tr>
<td>Core breeding ratio</td>
<td>~1</td>
<td>~1</td>
</tr>
<tr>
<td>Lead inlet/outlet temperature, °C</td>
<td>420/540</td>
<td>420/540</td>
</tr>
<tr>
<td>Peak cladding temperature, °C</td>
<td>650</td>
<td>650</td>
</tr>
<tr>
<td>Maximum lead velocity, m/s</td>
<td>1.8</td>
<td>1.8</td>
</tr>
<tr>
<td>Power effect, % ΔK/K</td>
<td>0.17</td>
<td>0.16</td>
</tr>
<tr>
<td>Total effect, % ΔK/K</td>
<td>0.33</td>
<td>0.32</td>
</tr>
<tr>
<td>Delayed neutrons share, β_eH, % ΔK/K</td>
<td>0.35</td>
<td>0.35</td>
</tr>
<tr>
<td>SG outlet temperature, °C</td>
<td>520</td>
<td>520</td>
</tr>
<tr>
<td>SG outlet steam pressure, MPa</td>
<td>24.5</td>
<td>24.5</td>
</tr>
<tr>
<td>Net efficiency, %</td>
<td>~44</td>
<td>~44</td>
</tr>
</tbody>
</table>

In accidents with damage of the reactor vessel and building, reactivity insertion, trip of pumps, loss of cooling from by the secondary circuit, failure of steam generator, ingress of vapor bubbles into the core, release of gaseous fission products, there is no runaway at prompt neutrons, excess temperature rise, fuel failure and catastrophic radioactive releases /6/. The integral void coefficient is deeply negative and the dangerous local positive effect is not manifested. The parameters and design features of the primary and secondary circuits prevent Pb freezing in emergency and blocking of the circulation lines. To assess safety margins, besides realistic, hypothetical accidents are also considered. In case of clad failure no fast fuel collapse will take place, with formation of a secondary critical mass and fast runaway. Insertion of reactivity worth tenths of $/s does not lead to steam explosion with large release of mechanical energy.
Table 2
Reactivity effects and coefficients

<table>
<thead>
<tr>
<th>Name</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Effects, $\Delta K/K$:</strong></td>
<td></td>
</tr>
<tr>
<td>Temperature</td>
<td>$-0.2 \times 10^{-3}$</td>
</tr>
<tr>
<td>Neptunium</td>
<td>$-1.0 \times 10^{-3}$</td>
</tr>
<tr>
<td>Power</td>
<td>$-1.6 \times 10^{-3}$</td>
</tr>
<tr>
<td>Fuel burnup</td>
<td>$-0.5 \times 10^{-3}$</td>
</tr>
<tr>
<td>Cladding corrosion</td>
<td>$0.5 \times 10^{-3}$</td>
</tr>
<tr>
<td>Margin for control</td>
<td>$0.4 \times 10^{-3}$</td>
</tr>
<tr>
<td>Summary margin</td>
<td>$3.2 \times 10^{-3}$</td>
</tr>
<tr>
<td>Lead solidification</td>
<td>$5.0 \times 10^{-3}$</td>
</tr>
<tr>
<td><strong>Coefficients, $(\Delta K/K)/\text{degrees}$:</strong></td>
<td></td>
</tr>
<tr>
<td>Lead density change</td>
<td>$1.9 \times 10^{-6}$</td>
</tr>
<tr>
<td>Radial core expansion</td>
<td>$-6.7 \times 10^{-6}$</td>
</tr>
<tr>
<td>Axial core expansion</td>
<td>$-1.1 \times 10^{-6}$</td>
</tr>
<tr>
<td>Doppler</td>
<td>$-5.6 \times 10^{-6}$</td>
</tr>
<tr>
<td>Doppler constant $(\Delta K/T)-T$</td>
<td>$-5.2 \times 10^{-3}$</td>
</tr>
<tr>
<td>Delayed neutrons fraction, $\beta_{eff}$</td>
<td>$3.6 \times 10^{-3}$</td>
</tr>
<tr>
<td>Neutron lifetime, s</td>
<td>$5.3 \times 10^{-7}$</td>
</tr>
</tbody>
</table>

The inherent safety enables simplifying the reactor design, control system and constructions, eliminating intermediate circuits in the main and emergency cooling systems, safeguard vessel (a pool-type reactor without metal vessel is also considered), a spent assembly washing system. Economical estimations allow one to expect the NPP cost to be equal or lower to that of NPP with LWR.

The NPP closed fuel cycle, electromechanical fuel reprocessing /7/ and its recycling together with other actinides in the reactor, transmutation of I and Tc, utilization of Sr and Cs, and long-term storage of the remaining fission products are envisaged, enabling radwaste disposal with their radiation hazard equivalent to that of mined uranium with its decay products.

Closing of the fuel cycle at NPP, combined recycle of all actinoids, elimination of uranium blanket and low reactivity margin contribute to non-proliferation and control measures.

The concept is based on a comprehensive experience with the LMFR and PbBi cooled naval reactors and we do not expect any principle technical problems in developing and demonstrating it by the beginning of the next century.

One of the possible ways to develop the concept of natural safety is a subcritical fast reactor with an external neutron source based on a proton accelerator. Such an approach makes an obvious safety case for the reactor in all the possible and conceivable reactivity accidents.

An urgent task both for Russia and for a number of other countries is to utilize within the next 2-3 decades the plutonium accumulated in storages which comes from NPPs, extracted from their fuel, and from dismantles nuclear weapons, converted into spent fuel. Pu utilization in reactors which will meet the high safety and economy requirements discussed above and will operate in an open fuel cycle at the first prolonged stage, allows reducing the time for clearing the storages of Pu and meeting the Pu management recommendations made by CISAC of the US National Academy of Sciences. For example, amount W-Pu BREST-300 reactor core is fixed 2240 kg, annual amount R-Pu formed in spent fuel may be about 757 kg.

Such an approach to Pu utilization can be especially attractive for the next stage of nuclear power industry advance, since it dispenses with the necessity for the
economically inefficient effort to create special "burners" or to burn plutonium in the operating reactors.

5. CONCLUSION

The considered example (concept) demonstrates that a very safe and cost-effective nuclear technology can be developed within the framework of the current experience and tradition, which can become the base of a large-scale nuclear power. It is a natural way of utilizing the accumulated Pu, requiring no special "burners".

REFERENCES


Some physical characteristics of BN-800 type reactor in uranium-plutonium and thorium-plutonium fuel cycle are considered in the report. It is shown that due to use of thorium in LMR some safety parameters are improved, such as the void effect, burn-up reactivity swing. Besides, BN-800 type reactor using plutonium as fissionable material can provide effective burning of more than 500 kg Pu a year, about 660 kg U-233 being built-up therewith for light water reactors. At the same time, according to the calculations, reactor loaded with thorium has somewhat lower Doppler reactivity coefficient.

1. INTRODUCTION

In assessment of evolution routes for nuclear power engineering (NPE) at present time, solution of traditional problems should be envisaged, namely, fuel supplies for NPE and saving fuel resources, meeting the modern requirements for safety, etc., as well as the problems of increasing actuality - utilization of weapon-grade and civil plutonium, burning or minimization of build-up of minor actinides.

It is well-known that minor actinides (Np, Am, Cm) are built-up in considerable amounts (~ 30 kg) in light water reactors loaded with enriched uranium fuel, and in even larger amounts - in reactors with MOX-fuel, due to the sequence of reactions of n-, γ - and α - decays in uranium and subsequent isotopes. This makes treatment of radioactive wastes in long terms more complicated.

In this connection, use of U-233 and thorium as fuel materials could serve as an alternative to the uranium-plutonium fuel cycle. In [1,2] the scheme of fuel cycle is proposed, as follows: thorium is loaded as the raw material to LMR, and U-233 built up therewith can be used in thermal reactors. In this case, both weapon-grade and civil plutonium can be used as fissionable material.

As a result of such organization of fuel cycle, the following objectives can be achieved:

1) Effective usage of fissionable materials: plutonium is the most efficient in reactors with hard neutron spectrum, i.e., in LMR uranium-233 is efficient fuel for thermal-neutron reactor. It is well-known that coefficient of conversion for the thermal-neutrons reactors with U-233 can approach unity. Thus, saving of fuel resources is provided.

2) In LMR with thorium, no secondary plutonium is bred, and, evidently, such reactor can serve as an efficient Pu burner.
3) When thorium is used in thermal and LMR the long-lived minor actinides are in fact not bred.

4) Assessments show that due to the use of uranium-233 and thorium, in principle, safety parameters can be improved in both LMR and thermal reactors.

Some results of calculations of physical parameters for BN-800 type LMR - plutonium burner, U-233 breeder are presented in this work.

2. CALCULATION SCHEME.

The calculations were carried out in two-dimensional geometry. Diffusion approximation was considered using 26 energy groups. The calculation scheme with dimensions of zones is presented in Fig.1. It is based on the layout of BN-800 type reactor [3] with uranium-plutonium oxide fuel. The following variants were considered: with oxide fuel in the core and breeding zones with replacement of raw material by Th-232, as well as two variants with metallic thorium in the blanket and fissionable material as oxide, respectively, PuO$_2$ and UO$_2$ dissolved in metallic thorium matrix.

It was not our task to optimize the reactor core, and, accordingly, the fractions of the material by volume were not changed in all the variants. The results of calculations as compared to the breeder variant of BN-800 type reactor are given in Table 1.
3. RESULTS OF CALCULATIONS. CONCLUSIONS.

Changes in safety characteristics and those of breeding in BN-800 type reactor were investigated in different compositions of fuel of the reactor core and breeding zones. The following conclusions can be made:

3.1. Safety of the reactor.

1) Replacement of U-238 raw material by Th-232 results in decrease of sodium void reactivity coefficient, in the case of oxide fuel - by the value $-\beta_{\text{eff}}$. In the variant with PuO$_2$ fuel in thorium matrix and blanket made from metallic Th, because of harder neutron spectrum, decrease of sodium void reactivity effect (SVRE) is less considerable. For analogous variant with fissionable material U-233 the void effect becomes negative, both in the reactor core.

2) In the variants with plutonium and thorium the value of burn-up reactivity swing is decreased considerably, which is explained mainly by the fact that effectiveness of newly bred uranium-233 exceeds that of plutonium being burned. As an explanation, let us consider changes of the value:

$$\omega_i^c = \left( \frac{\Sigma_i^f - \Sigma_{cf}^f}{\Sigma_i^f - \Sigma_{cj}^f} \right) P_{\text{eff}} - g$$

for three main fissionable isotopes - U-233, U-235, Pu-239. Evidently, for plutonium -239 the value of $\omega_i^c$ will be equal to 1. As this takes place, analogous values of $\omega_i^c$ for uranium-235 and uranium-233 will be

Table 1. Physical characteristics of BN-800 type reactor with different fuel composition

<table>
<thead>
<tr>
<th>Fuel composition type</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
</tr>
</thead>
<tbody>
<tr>
<td>core</td>
<td>UO$_2$-PuO$_2$</td>
<td>ThO$_2$-PuO$_2$</td>
<td>Th-PuO$_2$</td>
<td>Th-ThO$_2$</td>
</tr>
<tr>
<td>blanket</td>
<td>UO$_2$</td>
<td>ThO$_2$</td>
<td>Th</td>
<td>Th</td>
</tr>
<tr>
<td>$\Delta K$, $%$</td>
<td>2.7</td>
<td>1.7</td>
<td>1.8</td>
<td>3.3</td>
</tr>
<tr>
<td>SVR core, $%$</td>
<td>2.9</td>
<td>2.4</td>
<td>2.9</td>
<td>-0.3</td>
</tr>
<tr>
<td>$K_{\text{eff}}$, $\text{TdE}/\text{d}T$</td>
<td>42.8-03</td>
<td>30.8-03</td>
<td>1.76-03</td>
<td>2.16-03</td>
</tr>
<tr>
<td>$\rho_{\text{eff}}$</td>
<td>0.095</td>
<td>0.075</td>
<td>0.71</td>
<td>0.04</td>
</tr>
<tr>
<td>BR</td>
<td>0.69</td>
<td>1.15</td>
<td>1.15</td>
<td>1.12</td>
</tr>
<tr>
<td>G fission, kg</td>
<td>2030</td>
<td>2240</td>
<td>2230</td>
<td>2220</td>
</tr>
<tr>
<td>G ha, kg</td>
<td>13300</td>
<td>13300</td>
<td>15000</td>
<td>15000</td>
</tr>
<tr>
<td>$R_{\text{pu}}$, kg/year</td>
<td>156</td>
<td>-520</td>
<td>-515</td>
<td>-</td>
</tr>
<tr>
<td>$R_{\text{u}}$, kg/year</td>
<td>-</td>
<td>520</td>
<td>807</td>
<td>880</td>
</tr>
<tr>
<td>$\mu_{\text{core}}$, (NN/OUT) ppm</td>
<td>-</td>
<td>900/700</td>
<td>1230/1060</td>
<td>330/170</td>
</tr>
<tr>
<td>$\phi_{\text{rh}}, ppm$</td>
<td>-</td>
<td>55</td>
<td>40</td>
<td></td>
</tr>
<tr>
<td>$\phi_{\text{ab}}, ppm$</td>
<td>-</td>
<td>80</td>
<td>110</td>
<td>90</td>
</tr>
</tbody>
</table>

Notes:
- $R_{\text{pu}}$ - amount of Pu built up per year
- $R_{\text{u}}$ - amount of U-233 built up per year
- $\Delta K$ - burn-up reactivity swing (147 days)
- $\mu_{\text{core}}$, (NN/OUT) ppm - content of (Pa232+U232) in uranium unloaded
0.68 and 1.19 respectively. Therefore, requirements to the reactivity control system become less strict. For the variant with fissionable material U-233 and thorium, the value of burn-up reactivity swing is somewhat increased, which must be taken into consideration in choosing reactivity control system for the reactor.

3) Doppler constant $K_D$ is decreased in all variants, especially in the case of metallic thorium, which is caused by harder spectrum of neutrons.

4) In the variants with thorium-plutonium fuel the value $\phi_{\text{eff}}$ is approximately 15% less, compared to the referenced reactor, which, evidently, is explained by a smaller fission cross-section of thorium, as compared to uranium-238. However, in the variant with uranium-233 and thorium $\phi_{\text{eff}}$ is increased, due to greater yield of delayed neutrons in U-233 as compared to Pu.

3.2. Fuel build-up.

1) BN-800 type reactor with uranium-plutonium oxide fuel breeds about 200 kg Pu a year. In case the raw material is replaced by Th-232, such reactor is able of effective breeding of U-233 (~ 600-660 kg/year) burning more than 500 kg Pu. Therefore, on the one hand, the problem of plutonium utilization is solved, and on the other, effective fissionable material is bred for light water reactors.

2) U-233, "purest" by U-232 is bred in the axial and radial blanket (~ 50 ppm). Content of U-232 in the reactor core with plutonium amounts to ~ 1000 ppm. In the reactor with fissionable material U-233, content of U-232 in uranium being bred is three times less.

REFERENCES


SESSION 6: WEAPONS GRADE PLUTONIUM
A process has been proposed for the transmutation of plutonium. New fuels of multi-phase materials are fabricated by making use of conventional MOX fuel facilities. They are irradiated in LWR to generate electricity, and nearly 98% of Pu-239 can be transmuted. The spent fuels, that are assemblage of mineral-like phases of geologically stable, would become HLW without further processing. The process has a high priority in proliferation resistance, environmental safety and economy.

1. INTRODUCTION

On the use of plutonium, Japan Atomic Energy Research Institute (JAERI) has been promoting a basic research on LWR that almost completely burns up plutonium, making it possible to dispose of it directly [1]. The process consists of production of chemically stable (rock-like) fuels in conventional fuel facilities, burning the fuels in LWR and disposal of chemically stable spent fuels without further processing. For the research and development of the process, a series of works has been undertaken that consist of studies on the fuels and their burning. It also includes evaluation and analysis studies of proliferation resistance, environmental safety, technology and economy of the process [2-4]. Recently, the options of plutonium disposition have been discussed in the world [5-12].

The process based on the rock-like fuels would appear to be applicable for the disposition of the plutonium. Figure 1 shows a schematic diagram of the plutonium burning process (PROFIT process). The fuels are fabricated in conventional MOX fuel facilities by almost ordinary procedures. The fuels are then irradiated in LWR to generate electricity, and the plutonium is almost completely burned. The spent fuels thus obtained will be disposable as high-level radioactive wastes (HLW) after 30-50 years cooling without further processing. Accordingly, this process is expected to meet the objectives of denaturing the plutonium, forming stable HLW and generating electricity [2-4].

In the present paper, the outline of R&D program of the process in JAERI will be described together with the consideration of the chemically stable fuels and their burnups in LWR.

![Figure 1. Schematic Diagram of the PROFIT Process](image-url)
2. CONSIDERATION

2.1 Rock-like fuels

In the PROFIT process, it is desirable that the chemically stable fuels should be designed to satisfy the requirements in TABLE I.

(1) Proliferation resistance

It is favorable that the fuels are chemically so stable that they cannot easily dissolve into nitric acid solution. Accordingly, they are expected to be inherently proliferation resistant. From chemical properties of ceramic materials, some oxides appear to meet the above requirements[13].

(2) Economy

Use of conventional technologies and facilities is economical and reliable. The additives to dilute plutonium should be abundant and/or readily obtained. In the fields of ceramics and nuclear fuels, much technological backgrounds have been accumulated in the fabrication and processing of oxide materials.

(3) Environmental safety

In the process proposed, the spent fuels are to be the high level radioactive waste(HLW) without further processing, and they are expected to be geologically stable. It is well-known that most of the minerals survived in nature are oxide compounds.

TABLE I. Requirements and Candidate Systems of the Rock-like Fuels

<table>
<thead>
<tr>
<th>REQUIREMENTS</th>
<th>MEANS</th>
<th>POSSIBLE CHEMICAL FORMS</th>
<th>CANDIDATE SYSTEMS</th>
</tr>
</thead>
<tbody>
<tr>
<td>(1) Proliferation Resistance</td>
<td>Chemical stability</td>
<td>Oxide</td>
<td>Stable ceramics, Oxide</td>
</tr>
<tr>
<td>(2) Economy</td>
<td>Technological background of fuel production</td>
<td>Oxide, Carbide, Nitride, Alloys</td>
<td>Stable ceramics, Oxide</td>
</tr>
<tr>
<td>(3) Environmental Safety</td>
<td>Chemical stability</td>
<td>Oxide</td>
<td>Stable minerals, Oxide</td>
</tr>
</tbody>
</table>

From the comparison of chemical properties and crystal structures of ceramic materials and minerals in TABLE II, two oxide systems have been chosen for the stable fuels that will have rock-like structures and compositions: \( \text{PuO}_2-\text{ThO}_2-\text{Al}_2\text{O}_3-\text{MgO} \) and \( \text{PuO}_2-\text{stabilized ZrO}_2-\text{Al}_2\text{O}_3-\text{MgO} \) systems. They will consist of fluorite, corundum and spinel type phases. The spent fuels obtained will become an assemblage of compounds such as fluorite, corundum, spinel, hibonite and alloys. The fluorite, ThO\(_2\) and stabilized ZrO\(_2\) will become the host phase of the actinide and the lanthanide in fission products. With the presence of excess corundum(Al\(_2\)O\(_3\)), the alkali- and alkaline earth elements are to be solidified by formation of hibonite type phase(SrO\(\cdot\)6Al\(_2\)O\(_3\)). The spinel MgO\(\cdot\)Al\(_2\)O\(_3\) will also be a host phase of them. The noble metals, Ru, Rh and Pd will make alloys with Mo and Tc under low oxygen potentials. These phases are analogous to stable minerals. Estimated distributions of FPs among them are summarized in TABLE III.

The rock-like fuels proposed in the present work are multi-component oxides that is chemically stable in acid solutions. They are burned in conventional LWR such as PWR,
BWR and VVER-1000, as to be described in section 2.2. The spent fuels become an assemblage of multi-phase compounds with crystal structures similar to those of stable minerals, that is disposable as HLW. They would be stable under weathering over remarkably long times. Accordingly, the fuel system appears to constitute an inherent chemical barrier, that would contribute non-proliferation of plutonium.

<table>
<thead>
<tr>
<th>STABLE CERAMICS,</th>
<th>GEOLOGICALLY STABLE MINERALS</th>
<th>CANDIDATE FUEL SYSTEMS (Constituent Minerals)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(1) Alumina(Al₂O₃)</td>
<td>(1) Corundum(Al₂O₃), Sapphire(Al₂O₃)</td>
<td>(A) PuO₂-stabilized ZrO₂-Al₂O₃-MgO, (Zirkelite-Corundum-Spinel)</td>
</tr>
<tr>
<td>(2) Magnesia(MgO)</td>
<td>(2) Spinel(MgO-Al₂O₃)</td>
<td></td>
</tr>
<tr>
<td>(3) Zirconia(ZrO₂)</td>
<td>(3) Baddeleyite(ZrO₂), Zircon(SiZrO₄), Zirkelite (Ca,Fe,Th)₂(Zr,Ti,Nb)₂O₇</td>
<td>(B) PuO₂-ThO₂-Al₂O₃-MgO, (Thorianite-Corundum-Spinel)</td>
</tr>
<tr>
<td>(4) Thoria(ThO₂)</td>
<td>(4) Thorianite(ThO₂)</td>
<td></td>
</tr>
<tr>
<td>(5) Titania(TiO₂)</td>
<td>(5) Rutile(TiO₂), Perovskite(SrTiO₃)</td>
<td></td>
</tr>
</tbody>
</table>

2.2 Plutonium burning

In reactor burnup study, survey calculations have been performed on some simplified cell models of LWR, fast reactor(FR) and high temperature gas-cooled reactor(HTGR)[2-4]. The

![Figure 2. Comparison of the burning plutonium amounts estimated by cell calculation in the PuO₂-ThO₂-Al₂O₃ fueled PWR, HTGR and FR](image)

Figure 2. Comparison of the burning plutonium amounts estimated by cell calculation in the PuO₂-ThO₂-Al₂O₃ fueled PWR, HTGR and FR
results are summarized in Figure 2. The conventional LWRs appears to be promising for the plutonium burning. Such LWRs can transmute more than 98% of Pu-239 and 85% of total plutonium, and the plutonium quality becomes completely poor in the spent fuels. In FRs, about 50% of total plutonium is transmuted but the quality of plutonium in the discharged fuel was still high. This shows that FRs are indeed suitable for plutonium recycling. The initial plutonium inventory in HTGR is relatively small, and this leads to the smaller plutonium transmutation rate.

The void reactivity and the effective delayed neutron fraction $\beta_{\text{eff}}$ have been estimated for thoria (PuO$_2$-ThO$_2$-Al$_2$O$_3$) and zirconia (PuO$_2$-stabilized ZrO$_2$-Al$_2$O$_3$) fuels in LWRs, as shown in TABLE IV. The void reactivity was calculated to be close to that of the conventional UO$_2$-PWR in the thoria fuel, but close to zero in the zirconia fuel. For both fuels, $\beta_{\text{eff}}$ was small. It is therefore necessary to estimate the reactivity coefficients accurately by performing core burnup calculation and to carry out the safety analysis study to observe the reactor kinetics behavior.

TABLE III. Estimated Distribution of Pu and FPs in Fuels and Spent Fuels

<table>
<thead>
<tr>
<th>PHASE</th>
<th>Thorianite (cub.)</th>
<th>Spinel (MgO•Al$_2$O$_3$ (cub.)</th>
<th>Corundum (Al$_2$O$_3$) (hex.)</th>
<th>Hibonite (SrO•6Al$_2$O$_3$) (hex.)</th>
<th>Alloys (Ru-Alloys) (hex.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fuels</td>
<td>Pu</td>
<td>O</td>
<td>not formed</td>
<td>not formed</td>
<td></td>
</tr>
<tr>
<td>Spent fuels</td>
<td>Pu,Am,Cm</td>
<td>O</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Ce,Nd</td>
<td>O</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Cs,Rb</td>
<td>O</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Sr,Ba</td>
<td>O</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Ru,Rh,Pd,Mo,Tc</td>
<td>O</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

© : main distribution, O : secondary distribution

TABLE IV. Void Reactivity and Effective Delayed Neutron Fraction ($\beta_{\text{eff}}$) of PuO$_2$-ThO$_2$-Al$_2$O$_3$ and PuO$_2$-Stabilized ZrO$_2$-Al$_2$O$_3$ Fueled PWRs Evaluated by Cell Calculations

<table>
<thead>
<tr>
<th></th>
<th>PuO$_2$-ThO$_2$-Al$_2$O$_3$</th>
<th>PuO$_2$-ZrO$_2$(Y,Gd)-Al$_2$O$_3$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Void reactivity</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0%-95% void</td>
<td>-0.69</td>
<td>-0.16</td>
</tr>
<tr>
<td>$\beta_{\text{eff}}$ (%)</td>
<td>0.30</td>
<td>0.29</td>
</tr>
</tbody>
</table>

256
3. SCHEDULE OF R&D

3.1 Long term schedule

The schedule of R&D consists of three phases, phase I of fundamental study, phase II of engineering study and phase III of commercial operation, as shown in TABLE V.

In Phase I, fundamentals of the fuels are being studied to substantiate formation of fuel materials. They include phase relations, crystal structures, chemical and physical properties of the fuel materials. The irradiation tests of fuel materials are also to be carried out using JRR-3M and post irradiation test facilities. In the study of reactor core design, neutronics characteristics and safety analysis are examined. The fuel behavior under reactivity-initiated accident conditions is studied by the Nuclear Safety Research Reactor (NSRR). Economic assessment will also be carried out with technological evaluation.

In Phase II, a fuel production process due to the fundamental data obtained is to be examined in conventional MOX facilities to confirm and advance the engineering parameters. Advancement of fuel compositions is also to be examined through irradiation tests of fuel assemblages. In the study of nuclear core design, the most favorable core performance is designed and the reactor safety is investigated in the areas of accidental reactivity and thermal hydraulics.

In Phase III, commercial operations will be started using MOX fuel production facilities and LWR. The spent fuels obtained are cooled for 30-50 years, then sealed in canisters and disposed in repositories.

<table>
<thead>
<tr>
<th>Phase</th>
<th>Phase I (3 years)</th>
<th>Phase II (5 years)</th>
<th>Phase III</th>
</tr>
</thead>
<tbody>
<tr>
<td>R&amp;D Items</td>
<td>Fundamental R&amp;D •Formation of fuels and spent fuels, •Reactor burnup calculation, •Safety analysis</td>
<td>Engineering R&amp;D •Irradiation exp. •Engineering test, •Optimization, •Demonstration, •Advancement</td>
<td>Operation •Disposition of Pu (site specific)</td>
</tr>
</tbody>
</table>

3.2 Present status

(1) Fuel study

In the preliminary examination of the rock-like fuels, the phase relations of fuels and spent fuels have been studied by the reaction between inert additives, PuO₂ and simulated FPs. The phases produced were identified by X-ray diffraction analysis (XRD). The distribution of FPs is determined by scanning electron micro-analysis (SEM) and electron probe micro-analysis (EPMA). In the phase relation study of PuO₂-ThO₂-Al₂O₃ and PuO₂-ZrO₂(Gd)-Al₂O₃, it has been shown that the fluorite phase, the solid solutions of PuO₂-ThO₂ and PuO₂-ZrO₂(Gd), are in equilibrium with corundum(Al₂O₃). In the sample prepared from the inert additives and simulated FPs, formation of hibonite type phase has been identified, that is in equilibrium with the fluorite and corundum[2-4].

In the phase relation study of PuO₂-ThO₂-Al₂O₃-MgO and PuO₂-ZrO₂(Gd,Y)-Al₂O₃-MgO systems, it has been shown that the fuel compound is in three-phase equilibrium of fluorite...
Figure 3. Results of X-ray diffraction of a simulated fuel in ThO$_2$-Al$_2$O$_3$-MgO system prepared at 1,400°C under low oxygen potentials.

(A): A fuel compound of 5%PuO$_2$-25%ThO$_2$-60%Al$_2$O$_3$-10%MgO consisting of fluorite(F)+corundum(C)+spinel(S)

(B): A simulated spent fuel of 5%FPs-25%ThO$_2$-60%Al$_2$O$_3$-10%MgO consisting of fluorite(F)+corundum(C)+spinel(S)+hibonite(H)+alloys(A)

+ spinel(MgO•Al$_2$O$_3$) + corundum, and the spent fuel in five-phase equilibrium of fluorite + spinel + corundum + hibonite(SrO•6Al$_2$O$_3$) + alloys, respectively, as shown in Figure 3.

The XRD and EPMA results obtained show that the actinide and lanthanide are mainly dissolved in such fluorite type phases as thorianite ThO$_2$ and zirkelite (Zr,An)O$_2$-$x$, and the noble metals and molybdenum in Ru-base alloys, respectively. The alkaline-earth elements forms hibonite type phase SrO•6Al$_2$O$_3$ with corundum Al$_2$O$_3$. Tentative phase relations obtained are shown in Figure 4. Region (1) in ThO$_2$-Al$_2$O$_3$-MgO system in Figure 4-(A) shows a three-phase equilibrium of ThO$_2$+MgO•Al$_2$O$_3$+Al$_2$O$_3$ and region (2) a two-phase equilibrium of ThO$_2$+Al$_2$O$_3$, respectively. Plutonium oxide PuO$_2$ is dissolved in ThO$_2$ by formation of solid solutions. Thus, the composition regions of (1) and (2) would be acceptable for the fuel use.

From the formation of hibonite SrO•6Al$_2$O$_3$ in the simulated spent fuels, the quaternary system of SrO-ThO$_2$-Al$_2$O$_3$-MgO system in Figure 4-(B) is to be valid, that contains a four-phase region of ThO$_2$+MgO•Al$_2$O$_3$+SrO•6Al$_2$O$_3$+Al$_2$O$_3$ and a three-phase region of ThO$_2$+SrO•6Al$_2$O$_3$+Al$_2$O$_3$.

Measurements and evaluations of physico-chemical properties have been started. Irradiation studies of the fuel compounds are going to start using JRR-3M with a thermal neutron flux of 2x10$^{14}$/n/sec-cm$^2$, hot laboratories and fuel examination facility.
TABLE VI. Void and Doppler Reactivities and the Effective Delayed Neutron Fraction ($\beta_{\text{eff}}$) Estimated for PuO$_2$-ZrO$_2$(Y,Gd)-Al$_2$O$_3$ Fueled PWR Based on 2-Dimensional Core Burnup Calculation

<table>
<thead>
<tr>
<th>Void reactivity (%$\Delta k/k$)</th>
<th>BOEC</th>
<th>EOEC</th>
</tr>
</thead>
<tbody>
<tr>
<td>moderator density 100%</td>
<td>0.0</td>
<td>0.0</td>
</tr>
<tr>
<td>60%</td>
<td>-0.322</td>
<td>-9.74</td>
</tr>
<tr>
<td>30%</td>
<td>-3.42</td>
<td>-28.6</td>
</tr>
<tr>
<td>5%</td>
<td>-17.7</td>
<td>-92.6</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Doppler reactivity (%$\Delta k/k$)</th>
<th>BOEC</th>
<th>EOEC</th>
</tr>
</thead>
<tbody>
<tr>
<td>fuel temperature 1200K</td>
<td>-0.0982</td>
<td>-0.201</td>
</tr>
<tr>
<td>900K</td>
<td>0.0</td>
<td>0.0</td>
</tr>
<tr>
<td>600K</td>
<td>0.107</td>
<td>0.209</td>
</tr>
<tr>
<td>300K</td>
<td>0.253</td>
<td>0.467</td>
</tr>
</tbody>
</table>

| $\beta_{\text{eff}}$ | 2.87$\times$10$^{-3}$ | 3.50$\times$10$^{-3}$ |

BOEC : Beginning of the Equilibrium Cycle, EOEC : End of the Equilibrium Cycle

Figure 4. Tentative phase diagrams of fuel systems

(A) ThO$_2$-Al$_2$O$_3$-MgO system at around 1400°C
(1): three-phase region of ThO$_2$+MgO$\cdot$Al$_2$O$_3$+Al$_2$O$_3$
(2): two-phase region of ThO$_2$+Al$_2$O$_3$
PuO$_2$ is dissolved in ThO$_2$ by formation of solid solution.

(B) SrO-ThO$_2$-Al$_2$O$_3$-MgO system at around 1400°C
(1): four-phase region of ThO$_2$+MgO$\cdot$Al$_2$O$_3$+Al$_2$O$_3$+SrO$\cdot$6Al$_2$O$_3$
(2): three-phase region of ThO$_2$+Al$_2$O$_3$+SrO$\cdot$6Al$_2$O$_3$
(2) Reactor burnup study

In the reactor burnup and safety analyses, the core burnup calculation was carried out to accurately estimate the Doppler and void reactivity coefficients of zirconia fuel (PuO$_2$-stabilized ZrO$_2$-Al$_2$O$_3$) PWR. Figure 5 shows the plutonium transmutation characteristics obtained by the 2-dimensional core calculation. As much as 83% of total plutonium and 98% of Pu-239 is transmuted, and the quality of plutonium becomes very poor in the spent fuels. A total of 0.9 tonne of plutonium would be denatured every year on the assumption of a 1 GWe PWR operating at 80% availability.

As shown in TABLE VI, the void and Doppler reactivities for PuO$_2$-ZrO$_2$(Y,Gd)-Al$_2$O$_3$ fuel are negative both at the beginning and at the end of burnup cycle. The reactivities and $\beta_{\text{eff}}$ are, however, very small at the beginning of cycle. The safety analysis study of zirconia fueled PWR now in progress will confirm the effect of them on reactivity accident event.

To avoid the negative effects of small reactivity coefficients of the zirconia fuel, the examination has been started for two modified fuel systems of thoria-zirconia fuels PuO$_2$-(Th,Zr)O$_2$-Al$_2$O$_3$-MgO and zirconia fuels PuO$_2$-ZrO$_2$(Yb)-Al$_2$O$_3$-MgO-(W metal). A heterogeneous core will also be examined where these fuels are partially charged, and the reactor safety and operation control are studied.

4. SUMMARY

The fundamentals of the rock-like fuels and their burnup characteristics have been studied as scheduled. The benefits in the use of the process are indicated below.

(1) Economical and technological advantages: The process consists of MOX fuel production facilities, LWR and HLW repository. They are conventional technologies. It is not necessary to build new facilities. Accordingly, the process will be accomplished by use of present technology and existing infrastructures with minimum investment in facilities. It will provide for a favorable net profit in the generation of electrical power.
(2) Environmental safety: The rock-like fuels are multi-phase fuels, that are chemically and geologically stable. The spent fuels also consists of stable minerals. They are not soluble in nitric acid by conventional way and would be stable under weathering for geological period.

(3) Proliferation resistance: The chemical and geological stability of the fuels will provide a barrier for Pu proliferation (chemical barrier). Burnup calculations shows that nearly 98% of Pu-239 could be transmuted in LWR. A total of 0.9 tonne of plutonium would be denatured every year by the operation of a 1 GWe PWR at 80 % availability.

(4) R&D time: By taking advantage of conventional technologies, the time required for the research and development for the process would be relatively short. After engineering R&D in Phase II, the disposition operation could be started.

ACKNOWLEDGEMENT

The authors would like to thank Mr. S. Matsuura, the director general of the Tokai Research Establishment of JAERI, for his valuable discussions on the R&D. They are indebted to Mr. K. Ouchi for the experiments.

REFERENCES


COMBINING AN ACCELERATOR AND A GAS TURBINE MODULAR HELIUM REACTOR FOR NEAR TOTAL DESTRUCTION OF WEAPONS GRADE PLUTONIUM

A.M. BAXTER, D. ALBERSTEIN
General Atomics, San Diego, California, USA

Abstract

Fissioning the excess inventory of weapons-grade plutonium (WG-Pu) in a reactor is an effective means of rendering this stockpile non-weapons useable. The enormous energy content of the plutonium can be released by the fission process to produce valuable electric power. No fission option has been identified that can destroy more than about 72% of the WG-Pu without repeated reprocessing and recycling, presenting additional opportunities for diversion. However, the gas turbine modular helium-cooled reactor (GT-MHR) combines high plutonium destruction and electrical production efficiency with favorable economics in an inherently safe system. Accelerator driven sub-critical assemblies offer almost complete WG-Pu destruction, but achieve this goal only by using circulating molten salt solutions of plutonium, with difficult engineering problems and safety implications. By combining the GT-MHR with an accelerator-driven sub-critical MHR assembly, the best features of both systems can be merged to achieve the near total destruction of WG-Pu in an inherently safe, diversion-proof system. In this concept WG-Pu in the form of ceramic coated plutonium-oxide particles in graphite fuel blocks are irradiated to about 65% total plutonium burnup in GT-MHR modules. The discharged fuel elements are then placed directly in sub-critical GT-MHR modules with an accelerator-driven neutron source located in the central reflector of each sub-critical module. Depending on the plutonium destruction desired, more than 99.9% Pu-239 and more than 90% total plutonium destruction can be achieved. The discharged graphite fuel elements can be disposed of directly in standard multi-purpose canisters without additional processing. The modular concept minimizes the size of each unit so that both the GT-MHR and the accelerator would be straightforward extensions of current technology. No molten salt solutions of plutonium are required, and the fuel is always in a solid encapsulated form.

1.0 THE WEAPONS PLUTONIUM ISSUE

Plutonium can be used as a basic material in a fission weapon or as a trigger for a fusion (thermonuclear) weapon. The threat posed by the diversion of plutonium for such purposes has received increased attention as a result of the strategic arms reduction treaty (START) agreements which will make surplus tens of metric tons of weapons-grade plutonium in both the United States and Russia. The safe, permanent disposition of this material presents a difficult challenge.

Fissioning the excess inventory of weapons plutonium in a reactor appears to be the most effective means to render the stockpile non-weapons useable. Material can be consumed and the isotopic composition of the remaining material degraded to make it a less attractive weapons material. Also, the fission process leads to dilution of the material with highly radioactive by-products. Further, the enormous energy content within the plutonium is released by the fission process and can be captured and converted to produce valuable electric power. However, the degree of degradation induced during irradiation is an important consideration.
No fission option has been identified that can accomplish the destruction of more than about 72% of all of the plutonium without the repeated reprocessing and recycle of the irradiated fuel from the reactor. Even if reprocessing and recycle is allowed, the time required to accomplish total destruction levels (> 95%) is very long, in some scenarios exceeding 100 years.

Of all the fission options, the Gas-Turbine Modular Helium Reactor (GT-MHR) can provide the maximum plutonium destruction (up to 72%) without reprocessing, and can generate electricity at the highest efficiency, in an inherently safe manner.

The discharged fuel elements from the GT-MHR modules, containing the remaining plutonium, can be placed in a similar, subcritical core and further irradiated using a neutron source provided by an accelerator proton beam striking a target in the center of the subcritical core. Almost total destruction of the plutonium (up to 99.99% Pu-239 and 90% total Pu) can be achieved without recycle in this manner, and the discharged graphite fuel elements can be disposed of directly in standard spent nuclear fuel multipurpose canisters without further processing (Ref 1).

2.0 THE GT-MHR/ACCELERATOR COMBINATION

The destruction of weapons plutonium using the GT-MHR/accelerator combination would be accomplished in the following manner:
- First the weapons plutonium would be irradiated to high burnup in GT-MHR modules. 90% of the original Pu-239 and 65% of the total plutonium would be destroyed in this "deep burn" step.
- Next the discharged fuel elements would be placed in a subcritical GT-MHR core which is driven by an accelerator through a target located in the central annulus (AD-MHR). The fuel elements would be used "as is", and no reprocessing of the fuel would be required before irradiation in the AD-MHR. These discharged fuel elements would be irradiated to obtain the desired final plutonium destruction using the accelerator/target as a neutron source. Over 99% Pu-239 and 83% total

Table I
THE GT-MHR/ACCELERATOR COMBINATION FOR PLUTONIUM DESTRUCTION

<table>
<thead>
<tr>
<th>Basic Unit</th>
<th>3 GT-MHRs + 1 AD-MHR</th>
</tr>
</thead>
<tbody>
<tr>
<td>GT-MHR Power Level</td>
<td>600 MW(t)</td>
</tr>
<tr>
<td>GT-MHR Fuel Cycle</td>
<td>1/3 core per GT-MHR/year</td>
</tr>
<tr>
<td>AD-MHR Power Level</td>
<td>600 MW(t) to 207 MW(t) at constant beam power</td>
</tr>
<tr>
<td>AD-MHR Fuel Cycle</td>
<td>Full core per AD-MHR year</td>
</tr>
<tr>
<td>tonnes of WG-Pu destroyed per year</td>
<td>0.8</td>
</tr>
<tr>
<td>Net Electrical Power</td>
<td>975 MW(e) per year per MT</td>
</tr>
</tbody>
</table>
Figure 1: AD-MHR PLUTONIUM DISPOSITION SYSTEM LAYOUT
plutonium destruction can be achieved in less than one year of irradiation in the AD-MHR; and over 99.99% Pu-239 and 90% total plutonium destruction during a three year burnup.

- Finally, the fuel elements discharged from the AD-MHR core could be disposed of directly in a geologic repository as spent nuclear fuel without further processing.

This system achieves near total destruction of weapons plutonium without reprocessing and with minimum potential for diversion of the material.

The current GT-MHR design for "deep burn" of plutonium utilizes a three year fuel cycle in which one-third of the core is discharged each year. Since the AD-MHR is batch loaded, combining the annual discharge from three GT-MHR modules into a single AD-MHR core permits the destruction over 99% of the Pu-239 in a total of 4 years. A schematic layout is shown in Figure 1, and basic parameters are given in Table I.

The small modular design of the overall system means that the beam power requirements for the accelerator are relatively small so that it represents a straightforward extension of current technology. The combined system of 3 GT-MHRs and 1 AD-MHR has a net electrical output of almost 1000 MW(e). The accelerator requires only 72 MW(e), and with the very high efficiency of the gas turbine, the AD-MHR averages 195 MW(e) annually.

Both reactor types use the plutonium oxide coated fuel particle which has been successfully tested to about 747,000 MWD/Mt, and up to 1400°C, in irradiations carried out in the Peach Bottom reactor in the United States, and in the DRAGON and Studsvik reactors in Europe (Ref 2-4).

The thermal neutron spectrum in the MHR core is hard enough to successfully use the large, 0.46 eV, Er-167 neutron absorption resonance. Thus this material, in the form of coated particles of natural erbium oxide, is used as a burnable poison to control excess reactivity and ensure a negative core temperature coefficient under all conditions. No fertile material required, and no additional plutonium is created in the irradiation process, thus the system truly maximizes plutonium destruction and does not support a plutonium recycle economy.

The MHR, being graphite moderated and helium cooled, is capable of operating at sufficiently high temperatures that the helium coolant can be used in a direct Brayton cycle to drive a gas turbine which converts thermal energy to electricity at almost 50% efficiency. This results in excellent economics for the system when the electricity produced from the system is sold to the grid, and produces minimum thermal discharge to the environment.

3.0 DESCRIPTION OF SYSTEM COMPONENTS

3.1 GT-MHR Plant Design

The GT-MHR module arrangement is shown in Figure 2, and the basic parameters of the core are summarized in Table II. The components for each module are contained within a steel vessel system which includes a reactor vessel and a power conversion vessel. These vessels are in a side-by-side configuration connected by a cross vessel. The vessel system is sited underground in a concrete silo, which serves as an
Figure 2: GT-MHR MODULE ARRANGEMENT
independent high pressure containment structure. The reactor vessel is made of high
strength modified 9Cr-1Mo-V alloy steel and is approximately 8.5 m diameter and
about 23.8 m high. It contains the annular reactor core, the core supports, control rod
drives, refueling access penetrations, and a shutdown cooling system. The reactor
vessel is surrounded by a reactor cavity cooling system which provides totally passive
safety-related decay heat removal. The shutdown cooling system located at the
bottom of the reactor vessel provides decay heat removal for refueling and
maintenance.

<table>
<thead>
<tr>
<th>Table II</th>
<th>BASIC PARAMETERS OF THE GT-MHR</th>
</tr>
</thead>
<tbody>
<tr>
<td>Core Power Level</td>
<td>600 MW(t)</td>
</tr>
<tr>
<td>Core Power Density</td>
<td>6.58 W/cc</td>
</tr>
<tr>
<td>Core Coolant &amp; Pressure</td>
<td>Helium @ 7.07 MPa</td>
</tr>
<tr>
<td>Core Inlet Temperature</td>
<td>490°C</td>
</tr>
<tr>
<td>Core Outlet Temperature</td>
<td>850°C</td>
</tr>
<tr>
<td>Core Shape</td>
<td>Cylindrical Annulus</td>
</tr>
<tr>
<td>Core Size:</td>
<td></td>
</tr>
<tr>
<td>Height</td>
<td>7.93 m (10 blocks)</td>
</tr>
<tr>
<td>O.D.</td>
<td>4.86 m</td>
</tr>
<tr>
<td>I.D.</td>
<td>3.0 m</td>
</tr>
<tr>
<td>Number of Fuel Elements in Core</td>
<td>1020</td>
</tr>
<tr>
<td>Fuel Cycle Length</td>
<td>3 years</td>
</tr>
</tbody>
</table>

The reactor core consists of an assembly of hexagonal prismatic graphite blocks. The
fuel blocks are arranged in three annular rings. The center and outer portions of the
core comprise unfueled removable graphite reflector blocks. The core assembly is
surrounded by a steel core barrel and contained inside the uninsulated reactor vessel.
When operating as a plutonium burner (called a PC-MHR), the fuel cycle is a once-
through three-year cycle with one-third of the core refueled approximately every 11
months.

The power conversion vessel is also made of 9Cr alloy steel and is approximately 7.6 m
in diameter and about 22.9 m high. This vessel houses the turbomachinery, a plate-fin
recuperator, and a helical coil water-cooled intercooler and precooler. The
turbomachinery includes a generator, a turbine, and two compressor sections
submerged in helium, all mounted on a single shaft supported by magnetic bearings.

A simplified flow diagram of the GT-MHR is shown in Figure 3. The helium coolant
exits the reactor core at 850°C and 7.02 MPa, flows through the center hot duct within
the cross vessel, and is expanded through the turbine. The turbine directly drives the
electrical generator and the high and low pressure compressors. The helium exits the
turbine at 510°C and 2.56 MPa and flows through the plate-fin recuperator. This
compact and highly efficient device keeps as much energy as possible within the cycle,
Figure 3: GT-MHR FLOW SCHEMATIC

contributing to the high plant efficiency. The helium then passes through the precooler, and enters the low pressure compressor at 33°C, where it is compressed to 4.35 MPa. To maintain high pumping efficiency the gas is sent through a second cooler before going through the high pressure compressor. From there it passes through the recuperator where it picks up heat from the helium exiting the turbine, flows through the outer annulus of the cross vessel, up past the reactor vessel walls, and finally flows down through the core, entering at 490°C and 7.07 MPa, to complete the loop.

3.2 The AD-MHR and Target

As shown in Figure 4, the AD-MHR core is very similar to the GT-MHR core. The primary differences are the provision for an accelerator target for neutron production by
spallation, and a surrounding pressure vessel, both located in the central graphite reflector as shown in the figure; and the fact that the system would always be subcritical. The central graphite reflector serves to moderate the spallation neutrons and produce a thermal flux spectrum in the fuel annulus. Table III provides a summary of the basic parameters. The reactor core components, fuel, and all the power conversion system components are identical to those in the GT-MHR and are described in the previous section.

### Table III

**BASIC PARAMETERS OF THE AD-MHR & ACCELERATOR**

<table>
<thead>
<tr>
<th>Core Power Level:</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Maximum</td>
<td>600 MW(t)</td>
</tr>
<tr>
<td>Average</td>
<td>410 MW(t)</td>
</tr>
<tr>
<td>Minimum</td>
<td>210 MW(t)</td>
</tr>
<tr>
<td>Fuel Cycle Length</td>
<td>300 days</td>
</tr>
</tbody>
</table>

**Accelerator:**

<table>
<thead>
<tr>
<th>Accelerator:</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Proton Beam</td>
<td>800 Mev, 33 mA (avg)</td>
</tr>
<tr>
<td>Beam Power</td>
<td>28 MW</td>
</tr>
<tr>
<td>Power Requirements</td>
<td>72 MW(e)</td>
</tr>
<tr>
<td>Neutron Source Strength</td>
<td>~ 6 x 10^{18} n/s</td>
</tr>
</tbody>
</table>

![AD-MHR CORE LAYOUT](image-url)
Several options are available for the accelerator target in the AD-MHR. However, based on studies by Los Alamos National Laboratory, the best design appears to be one using molten lead flowing through the target, cooled by helium. The lead has a very low parasitic absorption cross section for neutrons, can handle the energy deposition from the accelerator without damage, and does not have stress or other problems arising from the spallation process (Ref 5).

The target would fit into the central graphite column space in the AD-MHR. The beam window separating the proton beam vacuum tube from the lead target would be located at the top of the vessel, well above the core. The beam would then pass through a low pressure helium tube before striking the lead target. The highest neutron production intensity would be at the top of the target, with the production rate decreasing linearly with depth. The range of the proton beam would be about one meter in the target.
Calculations show that this neutron production distribution leads to a satisfactory power distribution in the sub-critical core assembly. About 60% of the proton beam energy is deposited as heat in the target, and the lead would be liquid at operating temperatures; it would be cooled by helium. The flow rate of the lead through the system would be about 0.5 meters/second. The 800 MeV protons from the accelerator would produce 20 to 25 high energy neutrons (~ 15 MeV) per proton incident on the lead target.

### 3.3 Accelerator

The accelerator design requirements are significantly less than for other applications being considered, and, as shown in Table IV, represent reasonable extensions of current technology.

**Table IV**

**ACCELERATOR DESIGN REQUIREMENTS COMPARED TO CURRENT TECHNOLOGY**

<table>
<thead>
<tr>
<th>COMPONENT</th>
<th>STATUS</th>
<th>REQUIREMENT</th>
</tr>
</thead>
<tbody>
<tr>
<td>Injector</td>
<td>&gt; 100 mA demonstrated</td>
<td>≤125 mA</td>
</tr>
<tr>
<td>RFQ</td>
<td>70 mA, 0.6 MeV demonstrated</td>
<td>≤125 mA</td>
</tr>
<tr>
<td>Drift Tube Linac</td>
<td>100 mA pulsed configuration operated at LANL. 250 mA at CERN</td>
<td>250 mA peak, pulsed operation</td>
</tr>
<tr>
<td>Coupled Cavity Linac</td>
<td>17 mA in operation at LAMPF</td>
<td>30 to 250 mA</td>
</tr>
<tr>
<td>RF Generators</td>
<td>1 MW klystrons available from industry</td>
<td>1 MW Tubes</td>
</tr>
</tbody>
</table>

For the AD-MHR concept protons are produced and accelerated to an energy of about 20 MeV in two separate, 125 mA peak, current beams before being combined in a funnel for acceleration in the drift tube linac. In this stage they are accelerated to an energy of 800 MeV and reach a peak current of 250 mA before striking the lead target. A 42% efficiency in converting AC power input to beam power is achieved by pulsed operation. Each proton pulse is 1.11 ms long, and the pulse frequency is 120 pps. At this pulse rate, calculations show that a normal, steady-state neutron flux level is achieved in the AD-MHR core, because of the long average neutron lifetime.

### 4.0 RESULTS

Figure 5 shows the plutonium destruction capability of the AD-MHR using plutonium fuel elements discharged from the GT-MHR after a 3 year burnup. As can be seen from the figure, less than a three year exposure in the AD-MHR core (900 days) results in 90% total plutonium destruction and 99.99% Pu-239 consumption. Detailed dimensional core burnup calculations show acceptable power distributions in the system throughout this exposure time.
Even a single year burnup (300 days) results in > 99% Pu-239 destruction and 83% overall plutonium consumption. Figure 6 graphically compares the WG-Pu destruction achieved in the PC-MHR and AD-MHR as compared to other options being considered for dealing with this material. As discussed in the next section, these levels of destruction are more than adequate to prevent any further weapons use of the plutonium.

![Figure 6: COMPARISON OF DISPOSITION OPTIONS TO CONSUME AND DEGRADE PLUTONIUM](image)

5.0 MEASURES OF PROLIFERATION RESISTANCE

Table V summarizes the diversion resistance of the spent plutonium fuel from both the PC-MHR and AD-MHR, including the isotopic composition of the fuel relative to the original WG-Pu. As can be seen from the table, the AD-MHR fuel cycle offers an extremely high level of resistance to diversion and proliferation. The plutonium content of each spent fuel element is very low (about one-tenth a kilogram), and the plutonium is much more difficult to recover from spent PC-MHR fuel than from spent MOX fuel. Since over 150 AD-MHR fuel elements are required for a critical mass, it would be very difficult to divert a sufficient quantify of this fuel to obtain a weapons-useful quantity of plutonium; the technology for separating plutonium from MHR spent fuel has not been developed; and the isotopic mixture of the spent fuel makes it unattractive for weapons applications.

The radiation level in the AD-MHR discharged plutonium is extremely high, even after it has been separated from the fission products. The internal heat generation rate in the plutonium, and the spontaneous neutron emission rate are both large enough to preclude the use of this material as a weapon even by a country with sophisticated weapons technology.

6.0 CONCLUSIONS

The gas-turbine modular helium reactor can provide a very high level of weapons-grade plutonium destruction, without the need for recycle, in an inherently safe system which provides maximum efficiency for electrical energy production, and minimum environmental impact. This system is also very cost effective because of its high thermal efficiency.
### Table V

**DIVERSION RESISTANCE OF AD-MHR DISCHARGED PLUTONIUM**

<table>
<thead>
<tr>
<th>Composition, % of WG-Pu</th>
<th>WG-Pu</th>
<th>LWR Discharge</th>
<th>PC-MHR Discharge</th>
<th>AD-MHR Discharge</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu-238</td>
<td>0%</td>
<td>0.1%</td>
<td>0.1%</td>
<td>0.3%</td>
</tr>
<tr>
<td>Pu-239</td>
<td>94%</td>
<td>47.9%</td>
<td>9.8%</td>
<td>0.7%</td>
</tr>
<tr>
<td>Pu-240</td>
<td>6%</td>
<td>18.7%</td>
<td>10.7%</td>
<td>5.0%</td>
</tr>
<tr>
<td>Pu-241</td>
<td>0%</td>
<td>10.6%</td>
<td>11.6%</td>
<td>4.3%</td>
</tr>
<tr>
<td>Pu-242</td>
<td>0%</td>
<td>4.0%</td>
<td>3.3%</td>
<td>6.8%</td>
</tr>
</tbody>
</table>

**Reflected Sphere:**

<table>
<thead>
<tr>
<th>Critical Mass, Kgs</th>
<th>8.0</th>
<th>9.6</th>
<th>15.2</th>
<th>20</th>
</tr>
</thead>
<tbody>
<tr>
<td>Elements req'd</td>
<td>N/A</td>
<td>1</td>
<td>55</td>
<td>150</td>
</tr>
<tr>
<td>Volume, litre</td>
<td>N/A</td>
<td>170</td>
<td>4,977</td>
<td>13,575</td>
</tr>
</tbody>
</table>

**Weapons usefulness**

<table>
<thead>
<tr>
<th>Weapons usefulness</th>
<th>extreme</th>
<th>useful</th>
<th>very low</th>
<th>not useful</th>
</tr>
</thead>
</table>

By combining the GT-MHR with an accelerator driven subcritical assembly, this high efficiency, minimum environmental impact, and inherent safety can be retained while providing near total destruction level of weapons plutonium without recycle.

### REFERENCES


PHYSICAL AND TECHNICAL-ECONOMIC ASPECTS OF WEAPON-GRADE PLUTONIUM UTILIZATION IN HTGRs

A.I. KIRYUSHIN, N.G. KUZAVKOV, N.G. KODOCHIGOV, A.S. KUDRYASHOV, Yu.P. SUKHAREV
OKBM, Nizhny Novgorod

N.N. PONOMOREV-STEPNOI, N.E. KUKHARKIN, E.S. GLUSHKOV, V.N. GREBENNIIK
RRC KI, Moscow

Russian Federation

Abstract

The current works on the program of reduction in the strategic nuclear armaments will lead to release of large stock-piles of weapon-grade plutonium (WPu). Storage of these quantities of weapon-grade plutonium requires complicated equipment and large economic expenses.

The problem of plutonium effective utilization requires urgent solution. There are some ways for that and HTGRs are a good base for plutonium burning.

In this paper the estimations of physical and technical-economic characteristics of HTGR with 200 MW power are presented. As a fuel the weapon-grade plutonium or plutonium together with different dilutions (U-238, Th-232) are used.

To guarantee of reactor safety requirement properties (to ensure a large negative temperature coefficient of reactivity at all temperatures) erbium as a burn able poison is used.

The difference between spectral characteristics of the reactor with pebble bed core (VGM) and the MHTGR type reactor with prismatic fuel blocks leads to more deeper Pu-239 burning in VGM reactor at the more less average burn up.

The comparison of weapon-grade Pu burning effectiveness in HTGR with its utilization in the other reactor types shows the advantage of HTGRs usage.

It is shown that HTGR allows to provide for a very deep, more than 80 %, Pu-239 burning and fuel element arrangement features facilitate the problem of irradiated fuel storage.

1. INTRODUCTION

The current works on the program of reduction in the strategic nuclear armaments will lead to release of large stock-piles of weapon-grade plutonium primarily in the USA and Russia (about 200 tons according to the estimates).

Storage of these quantities of weapon-grade plutonium requires complicated equipment and large economic expenses and can not be considered as a final stage of the program on nuclear armament reduction.

The present paper considers the problem of reduction (or full destruction) of the stock-piles of weapon-grade plutonium and its efficient application.

There is a number of ways of this problem solving, and one of the possible solutions of this problem is to use of the weapon-grade plutonium as a fuel for nuclear power reactors for electricity and heat production.
1. reactor
2. pressure vessels unit
3. intermediate heat exchanger
4. steam generator
5. gas circulator
6. surface cooling system

7. fuel circulation system
8. small absorber balls system
9. helium purification system
10. relief valve
11. steam-turbine plant

FIG. 1. VGM reactor plant.
In the present paper the comparison of the effectiveness of weapon-grade plutonium burning in various type of reactors is given.

The investigation carried out in the USA and Russia showed that the high-temperature helium-cooled reactors are most suitable for burning the weapon-grade plutonium, where more than 80% Pu-239 may be burnt up.

Using of coated particles with Pyc-Sic coatings as a fuel elements in HTGR facilitates the problem of irradiated fuel storage.

The comparison of weapon-grade plutonium utilization in VGM reactor (Russia) and MHTGR reactor (USA) is given as well.

2. THE MAIN TECHNICAL CHARACTERISTICS OF VGM AND MHTGR.

The high-temperature gas-cooled reactors (HTGR) are the advanced trend in nuclear power ensuring the high safety characteristics and unique possibilities of using both for electricity generation and for direct use of thermal power.

The main HTGR characteristics and advantages are determined by the following factors:
- thermal efficiency, small heat releases to the environment, low consumption of cooling water;
- high nuclear safety ensured by the inherent characteristics: chemically inert one-phase helium coolant, high heat capacity of the graphite core, high stability of fuel element based on fuel particles with multilayer coatings, high negative temperature coefficient of reactivity;
- low specific activity of the circuit and low level of radioactivity release to the environment;
- minimum excess reactivity margin (due to application of the principle of continuous refueling with spherical fuel elements or using of burn able poison).

In Russia concepts and designs of reactor plants with HTGR have been developing for many years within a wide range of power and for various purposes, extensive R&D efforts have been made for this direction.

2.1. VGM reactor plant with HTGR module concept HTGR.

The reactor plant (fig. 1) consists of:
- the reactor with a pebble bed core including control and shutdown systems;
- the vessel block consisting of the reactor vessel, the vessel for heat exchanging equipment and the cross vessel;
- high temperature heat exchanger;
- steam generator;
- gas blower;
- system providing fuel circulation, loading and reloading of the fuel elements;
- surface cooling system.

Technical solutions for reactor systems provide reactor operation at the temperatures of 750°C or 950°C. At the operating temperature of 750°C all the heat from the reactor is rejected in the steam generator and steam is used in the steam turbine unit. At the operating temperature of 950°C they start using intermediate heat exchanger (IHX) and the intermediate circuit, where there can be placed steam generator, gas turbine or...
experimental devices for the demonstration of reactor heat application.

VGM reactor system has the following parameters (in brackets the values are given for outlet temperature of 750 °C) (see table 1):/1):

Table 1.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Reactor thermal power</td>
<td>200(215) MW</td>
</tr>
<tr>
<td>SG thermal power</td>
<td>124(217) MWT</td>
</tr>
<tr>
<td>IHX thermal power</td>
<td>77 MWT</td>
</tr>
<tr>
<td>Primary circuit helium parameters:</td>
<td></td>
</tr>
<tr>
<td>core inlet/outlet temperature</td>
<td>300 °C/ up to 950(750 °C)</td>
</tr>
<tr>
<td>pressure</td>
<td>5,0 MPa</td>
</tr>
<tr>
<td>Intermediate circuit parameters:</td>
<td></td>
</tr>
<tr>
<td>IHX outlet inlet temperature</td>
<td>290 °C/ up to 900 °C</td>
</tr>
<tr>
<td>pressure</td>
<td>5,2 MPa</td>
</tr>
<tr>
<td>Steam parameters:</td>
<td></td>
</tr>
<tr>
<td>temperature</td>
<td>540 °C</td>
</tr>
<tr>
<td>pressure</td>
<td>17,2 MPa</td>
</tr>
<tr>
<td>Core:</td>
<td></td>
</tr>
<tr>
<td>height/diameter</td>
<td>9,4/3,0 m</td>
</tr>
<tr>
<td>average power density</td>
<td>3 MW/m</td>
</tr>
<tr>
<td>fuel</td>
<td>UO</td>
</tr>
<tr>
<td>enrichment for U-235</td>
<td>2</td>
</tr>
<tr>
<td>average burn up</td>
<td>8 %</td>
</tr>
<tr>
<td>average fuel residence time</td>
<td>76000 MWdays/tU</td>
</tr>
<tr>
<td>number of reshuffling cycles of fuel elements</td>
<td>950 days</td>
</tr>
<tr>
<td>Surface cooling system:</td>
<td></td>
</tr>
<tr>
<td>number of independent channels</td>
<td>3</td>
</tr>
<tr>
<td>thermal power at emergency cooling</td>
<td>1.15 MW</td>
</tr>
<tr>
<td>Reactor power change rate</td>
<td>100-50-100 %</td>
</tr>
<tr>
<td>Lifetime</td>
<td>40 years</td>
</tr>
</tbody>
</table>

Fuel elements contain the fuel particles of spherical form with diameter of 0,5 mm surrounded by the multilayer coating of pyro-carbon and silicon carbide. Coated particles are evenly distributed within the graphite sphere of 50 mm in diameter. Graphite sphere containing fuel matrix surrounded with the graphite layer of 5 mm in thickness (see. fig. 2).

2.2. Main characteristics and features of MHTGR (USA).

The technical solutions of the design were mainly based on the domestic experience of designing, construction and operation of the Peach Bottom and Fort St.Vrain reactors.

The main characteristics of the 450 MWT MHTGR design are listed in Table 2.

Standard refractory of coated particle fuel, as shown in figure 3, is used in the MHTGR.

Plutonium utilization is not a new application for the HTGR. Highly enriched, (88 % fissile) coated, plutonium oxide fuel particles were designed, fabricated and irradiated in the Peach
Bottom HTGR project in the late 1960s and early 1970s. The test fuel was irradiated to burn ups in excess of 700000 MWd/t. Similar tests in Europe of plutonium particles manufactured by Belgonucleaire for the Dragon Project also yielded similar successful results.
Table 2.

<p>| | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Power</td>
<td>450 VWT</td>
</tr>
<tr>
<td>Temperature helium</td>
<td></td>
</tr>
<tr>
<td>inlet of the core</td>
<td>288 °C</td>
</tr>
<tr>
<td>outlet of the core</td>
<td>705 °C</td>
</tr>
<tr>
<td>Helium pressure</td>
<td>7.05 MPa</td>
</tr>
<tr>
<td>Fuel cycle</td>
<td>LEU (~20 % enrichment)</td>
</tr>
<tr>
<td>Number of fuel columns</td>
<td>84</td>
</tr>
<tr>
<td>Power density</td>
<td>6 MW/m</td>
</tr>
<tr>
<td>Reactor vessel diameter</td>
<td>23.7 ft</td>
</tr>
<tr>
<td>Electrical power</td>
<td>173 MW</td>
</tr>
<tr>
<td>Efficiency</td>
<td>38.4 %</td>
</tr>
</tbody>
</table>

3. NEUTRON AND PHYSICAL ASPECTS OF WEAPONS-GRADE PLUTONIUM UTILIZATION

Any fissile material (U-235, Pu-239, U-233) both in separate particles or mixed with fertile materials (U-238, Th-232) can be used as a fuel in HTGR. The HTGRs are very flexible with respect to the fuel cycle. The ratio of the number of graphite nuclei to that of fissile material nuclei, determines the neutron spectrum in the HTGR. Amount of fertile material and the degree of the core heterogeneity can be varied within a wide range changing the quantity of the fissile (fertile) material and fuel particle size, which is difficult in the case of the other type reactors.

In estimation of the neutronic characteristics of the VGM reactor with weapons-grade plutonium loaded into the fuel elements some of versions were considered: use of pure plutonium without dilution, plutonium diluted with Uranium-238 (initial enrichment with Pu - 21 % and 10 %), and Thorium-232 (initial enrichment with Pu - 10 %). In all versions the once-through fuel cycle (without chemical processing) was assumed.

The analysis of the results obtained showed that the initial loading 0.5 g of Pu-239 per a fuel element is optimal taking into account assurance of the necessary worth of the control rods located outside the core (in the side reflector).

The parameters of VGM fuel with use of weapons-grade plutonium for various versions of its application in comparison with the design for the uranium fuel are presented in table 3. For comparison this table also lists the similar data for American reactor MHTGR obtained by G.A. It follows from the analysis of the table 3 data that VGM reactor can be effectively used for destroying of weapons-grade plutonium simultaneously with electricity generation.

At the VGM thermal power of 200 MW the annual output of one reactor is \((\frac{9}{2} \times 0.8) 615 \text{ MW} \cdot \text{yr}\), with about 260 kg of the weapons-grade plutonium destroyed within the same period.

Therefore for destroying 50 tons of the weapons-grade plutonium over a period of 40 years about 18 VGM reactors will be needed with the electricity output of 45 GWe-yr.

The HTGR reactor fuelled with plutonium-239 has the positive temperature effect, which is undesirable from the safety point of view. To assure the negative temperature effect the reactor fuel elements must be added with poisons having resonances near the
Table 3
Fuel parameters of VGM Reactor for use of weapons-grade plutonium

<table>
<thead>
<tr>
<th>Value of Parameter</th>
<th>initial option VGM 235</th>
<th>Option 1</th>
<th>Option 2</th>
<th>Option 3</th>
<th>Option 4</th>
<th>MHTGR (USA)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Charge, g/MWdt</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>235 U 8% Pu 238 U 92%</td>
<td>0.94</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Pu 239</td>
<td>-</td>
<td>1.27</td>
<td>1.0</td>
<td>1.10</td>
<td>1.0</td>
<td>1.68</td>
</tr>
<tr>
<td>Pu 240</td>
<td>-</td>
<td>0.08</td>
<td>0.06</td>
<td>0.07</td>
<td>0.06</td>
<td>0.11</td>
</tr>
<tr>
<td>Pu 239</td>
<td>-</td>
<td>1.35</td>
<td>1.06</td>
<td>1.17</td>
<td>1.06</td>
<td>1.79</td>
</tr>
<tr>
<td>Pu enrichment of Pu 239 %</td>
<td>-</td>
<td>94</td>
<td>94</td>
<td>94</td>
<td>94</td>
<td>94</td>
</tr>
<tr>
<td>Discharge, g/MWdt</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>235 U</td>
<td>0.128</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>233 U</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>0.100</td>
<td>-</td>
</tr>
<tr>
<td>Pu 239</td>
<td>0.056</td>
<td>0.090</td>
<td>0.077</td>
<td>0.178</td>
<td>0.050</td>
<td>0.174</td>
</tr>
<tr>
<td>Pu 240</td>
<td>0.045</td>
<td>0.081</td>
<td>0.100</td>
<td>0.146</td>
<td>0.090</td>
<td>0.23</td>
</tr>
<tr>
<td>Pu 241</td>
<td>0.024</td>
<td>0.102</td>
<td>0.100</td>
<td>0.151</td>
<td>0.090</td>
<td>0.25</td>
</tr>
<tr>
<td>Pu 242</td>
<td>0.020</td>
<td>0.086</td>
<td>0.077</td>
<td>0.065</td>
<td>0.070</td>
<td>0.04</td>
</tr>
<tr>
<td>Pu 239</td>
<td>0.145</td>
<td>0.359</td>
<td>0.354</td>
<td>0.540</td>
<td>0.300</td>
<td>0.70</td>
</tr>
<tr>
<td>Pu content of Pu 239 %</td>
<td>38</td>
<td>25</td>
<td>22</td>
<td>33</td>
<td>17</td>
<td>25</td>
</tr>
<tr>
<td>Fuel burn up, GWD/t</td>
<td>78</td>
<td>730</td>
<td>210</td>
<td>90</td>
<td>100</td>
<td>560</td>
</tr>
<tr>
<td>Pu loading, kg/yr (ν = 0.8)</td>
<td>-</td>
<td>75</td>
<td>60</td>
<td>65</td>
<td>60</td>
<td>215</td>
</tr>
<tr>
<td>Destruction of Pu 239</td>
<td>-</td>
<td>93</td>
<td>92</td>
<td>84</td>
<td>95</td>
<td>90</td>
</tr>
<tr>
<td>Pu destruction total %</td>
<td>-</td>
<td>73</td>
<td>67</td>
<td>54</td>
<td>72</td>
<td>60</td>
</tr>
</tbody>
</table>

The calculations showed that in insertion of 0.1 g of Er into each VGM spherical fuel element the negative reactivity coefficient of

\[ -7 \times 10^5 \text{ l/degree} \]

is attained at the operating temperature, reactivity margin in this case is about 7 % \( \Delta k/k \) at room temperature.

Although the delayed neutron fraction for Pu-239 is essentially lower than that for U-235 (\( \beta_{\text{eff}} = 0.0021 \) instead of 0.0065) it is expected that the slow thermal response of the core, the single-phase coolant and the large margin between the normal and limiting fuel temperature (\( ^\circ 400 \)) ensure the reactor safety.
4. THE COMPARISON OF THE EFFICIENCY OF WEAPONS-GRADE PLUTONIUM DESTRUCTION IN POWER REACTORS OF THE DIFFERENT TYPES.

The net fraction of Pu-239 destroyed at burner NPR's depends on the following factors:
- fuel burn up (i.e., the fraction of fission products relative to fissile atoms (fima) or to all heavy nuclides (fifa));
- the impact of radiation capture losses in the fuel characterized by $\chi$ - parameter ($\chi = \frac{N_c}{N_f}$, $N_c$, $N_f$ - rates of capture and fission reactions).

Maximum burn up in the different projects considered was taken as follows:
- for VGM reactor the fima value $\approx 73\%$;
- for VVER-1000 - 3.4 $\%$;
- for BN - 1600 - 10 $\%$, (see table 4).

The comparison performed shows that VGM has the advantages against the other reactors - burners in both parameters:
- the burn up may reach 730000 MWd/t and more due to the implementation of multi-layer protective coatings;
- the fraction of parasitic absorption in fuel $\alpha' = 0.6$ due to thermalization spectrum peak being displaced towards Pu resonance at 0.3 ev.

The rate of Pu destruction due to transmutation to higher isotopes via radiation capture ($n,\gamma$) is 2-3 times higher than for the other reactor considered.
The content of Pu in irradiated fuel is reduced from 94% (fresh WPu) to:
- 25% - VGM;
- 40% - VVER;
- 85% - BN.

Table 4. Comparison of WPu destruction efficiency in various power reactors

<table>
<thead>
<tr>
<th>Characteristics (Discharge fuel)</th>
<th>NPR's type</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>VGM</td>
</tr>
<tr>
<td>238 U, kg/t</td>
<td></td>
</tr>
<tr>
<td>239 Pu, kg/t</td>
<td>66,7</td>
</tr>
<tr>
<td>240 Pu</td>
<td>60,0</td>
</tr>
<tr>
<td>241 Pu</td>
<td>75,5</td>
</tr>
<tr>
<td>242 Pu</td>
<td>63,7</td>
</tr>
<tr>
<td>Pu kg/t</td>
<td>265,3</td>
</tr>
<tr>
<td>Fission Products, kg/t</td>
<td>734,0</td>
</tr>
<tr>
<td>Pu destruction, %</td>
<td>93,0</td>
</tr>
</tbody>
</table>

Co-generation of electricity at WPu burners NPR's (for 80% capacity factor) during 40 years

| Power, GW(t). | 18x0,2 | 2 x 3,225 | 5 x 4,3 |
| Net efficiency, % | 38,4 | 31 | 37,2 |
| Power, GW(e) | 1,382 | 2,0 | 8,0 |
| Fuel burnup: |
| % fima | 73 | 3,4 | 10,0 |
| % fifa | 77,6 | 77,3 | 83,3 |
| Co-generation of electricity, GW(e) year per 50 t Pu, no recycling | 45,0 | 65,0 | 255,0 |

CONCLUSION

1. In case of the fuel cycle with the single-act irradiation of WPu (without recycling) the module VGM burner-reactor possesses the advantage:
   - in Pu-239 incineration efficiency per one cycle (the burn up may reach 730000 MWD/tPu), it removes the irradiated fuel out of the sphere of probable military use;
   - the efficiency of NPR with VGM is higher than that for some other types of reactors-incinerators due to VGM maximum electricity production in burning of a given amount of weapons-grade plutonium.
2. The developed technology of multi-layer coatings provides tightness of particles and the absence of chemical
interaction between the fission products and the structural materials. It facilitates of solving the problem of irradiated fuel storage.

REFERENCES


LIST OF PARTICIPANTS

BELGIUM

Bairiot, H. (Co-chairman)  FEX
                Lijsterdreef 24
                B-2400 Mol

CANADA

Meneley, D.A.  AECL CANDU
                2251 Speakman Drive
                Mississauga L5K B2

FRANCE

Garnier, J-C.  CEA/CEN-Cadarache
                Direction des Reacteurs Nucléaires
                B.P. 211
                13108 Saint-Paul-lez-Durance Cedex

Lecocq, A.B.J.  EURiWA
                Rue des Fauvette 27
                F-91400 Orsay

Rehbinder, S.  NUSYS
                Rue Christope Colomb 9
                F-75008 Paris

GERMANY

Krüger, W.  BfS Berlin - Karlishorst
                KT 3

ITALY

Landeyro, P.A.  C.R.E. CASACCIA
                Via Anguillarese 301
                I-00100 Rome

Lombardi, C.  Dept. of Nuclear Engineering
                Politecnico di Milano
                Ponzio 84/3
                I-20138 Milano

Mazzola, A.  Dept. of Nuclear Engineering
                Politecnico di Milano
                S. Donato (Mi) 20097

JAPAN

Furukawa, K.  Tokai University
                Kitakaname 1117
                Hiratsuka, Kanagawa 259-12
Ishikawa, M.  
Power Reactor & Nuclear Fuel Development Corporation  
Narita 4002  
Oharai-machi 311-13

Mitachi, K.  
Toyoshashi University of Technology  
Tempaku-cho 1-1  
Toyohashi 441

Muromura, T.  
Japan Atomic Energy Research Institute  
Tokai-mura, Ibaraki-ken 319-11

Yamashita, K.  
Japan Atomic Energy Research Institute  
Naritachyo 3607  
Ohairai-machi 311-13

RUSSIAN FEDERATION

Baiburin, G.G.  
All-Russian Scientific and Research Institute of Inorganic Materials (ARSRIIM)  
Rogov 5a  
123479 Moscow

Belov, S.B.  
Experimental Design Bureau of Machine Building (EDBMB)  
Burnakovsky proezd 15  
60363 N. Novgorod

Dekusar, V.M.  
Institute of Physics & Power Engineering (IPPE)  
249020 Bondarenko sz. 1  
Obninsk, Kaluga Region

Ivanov, E.A.  
Institute of Physics & Power Engineering (IPPE)  
249020 Bondarenko sz. 1  
Obninsk, Kaluga Region

Kagramanian, V.S.  
(Scientific Co-ordinator)  
Institute of Physics & Power Engineering (IPPE)  
249020 Bondarenko sz. 1  
Obninsk, Kaluga Region

Kazaritsky, V.D.  
Institute for Theoretical & Experimental Physics  
25, B. Cheremushkinskaya  
117259 Moscow

Krivitski, I.Y.  
Institute of Physics & Power Engineering (IPPE)  
249020 Bondarenko sz. 1  
Obninsk, Kaluga Region

Koshkin, I.  
Ministry of Atomic Energy of the Russian Federation (MINATOM)  
Staromonetny pereulok 26  
109180 Moscow

286
Kovalevski

Kudryavsev, E.

Murogov, V.M. (Chairman)

Naumov, V.S.

Pshakin, G.

Rogozkin, B.D.

Smetanin, E.Ya.

Sukharev, Yu. P.

Zabudko, L.M.

Ziabletzev, D.N.

**SWITZERLAND**

Chawla, R.

**UNITED KINGDOM**

Page, R.J.
USA

Alberstein, D. General Atomics
3550 General Atomics Court
San Diego, CA 9212

ORGANIZATIONS

Magill, J. CEC/Joint Research Centre
Institute for Transuranium Elements
Postfach 2340
76125 Karlsruhe
Germany

Sukhanov, G. International Atomic Energy Agency
(Scientific Secretary)
Wagramerstrasse 5
P.O. Box 100
A-1140 Vienna
Austria

Rinejski, A. International Atomic Energy Agency
(Co-Scientific Secretary)
Wagramerstrasse 5
P.O. Box 100
A-1140 Vienna
Austria

Takamatsu, M. International Atomic Energy Agency
Wagramerstrasse 5
P.O. Box 100
A-1140 Vienna
Austria

Zaribas, N. OECD/NEA
Boulevard des Iles 12
F-92130 Issy-les-Moulineaux
QUESTIONNAIRE ON IAEA-TECDOCs

It would greatly assist the International Atomic Energy Agency in its analysis of the effectiveness of its Technical Document programme if you could kindly answer the following questions and return the form to the address shown below. Your co-operation is greatly appreciated.

Title: Unconventional options for plutonium disposition
Number: IAEA-TECDOC-840

1. How did you obtain this TECDOC?

[ ] From the IAEA:
   [ ] At own request
   [ ] Without request
   [ ] As participant at an IAEA meeting
[ ] From a professional colleague
[ ] From library

2. How do you rate the content of the TECDOC?

[ ] Useful, includes information not found elsewhere
[ ] Useful as a survey of the subject area
[ ] Useful for reference
[ ] Useful because of its international character
[ ] Useful for training or study purposes
[ ] Not very useful. If not, why not?

3. How do you become aware of the TECDOCs available from the IAEA?

[ ] From references in:
   [ ] IAEA publications
   [ ] Other publications
[ ] From IAEA meetings
[ ] From IAEA newsletters
[ ] By other means (please specify)
[ ] If you find it difficult to obtain information on TECDOCs please tick this box

4. Do you make use of IAEA-TECDOCs?

[ ] Frequently
[ ] Occasionally
[ ] Rarely

5. Please state the institute (or country) in which you are working:

Please return to: R.F. Kelleher
Head, Publishing Section
International Atomic Energy Agency
P.O. Box 100
Wagramerstrasse 5
A-1400 Vienna, Austria