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Thorium fuel utilization: Options and trends

Proceedings of three IAEA meetings held in Vienna in 1997, 1998 and 1999

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FOREWORD

A series of three IAEA meetings on utilization of thorium fuel were held in Vienna over a period of three years, 1997–1999:

- Advisory Group Meeting on Thorium Fuel Cycle Perspectives, Vienna, 16–18 April 1997.
- Advisory Group Meeting on Thorium Fuel Utilization: Options and Trends, Vienna, 28–30 September 1998.
- Technical Committee Meeting on Utilisation of Thorium Fuel; Options in Emerging Nuclear Energy Systems, Vienna, 15–17 November 1999.

The purpose of the meetings was to assess the advantages shortcomings, and options of the thorium fuel under current conditions, with the aim of identifying new research areas and fields of possible co-operation within the framework of the IAEA Programme on Emerging Nuclear Energy Systems. Apart from current commercial reactors, the scope of the meetings covered all types of evolutionary and innovative nuclear reactors, including molten salt reactors and hybrid systems.

For the convenience of readers, the titles of papers presented at the 1997, 1998, 1999 meetings are marked by 1, 2 and 3 asterisks (, ,) respectively.

The IAEA would like to thank all participants and authors of papers for their valuable contributions to the success of the meetings. The IAEA officers participating in the organization the above mentioned meetings were: V. Arkhipov, V. Onoufriev, J-S. Choi and A. Stanculescu. The IAEA officer responsible for the publication was A. Stanculescu.

EDITORIAL NOTE

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CONTENTS

III. NUCLEAR AND REACTOR PHYSICS, NUCLEAR DATA AND CORE DESIGN

IV. TECHNOLOGY AND DESIGN OPTIONS

V. PROBLEMS TO RESOLVE: SAFETY, NON-PROLIFERATION, ENVIRONMENT PROTECTION

SUMMARY

Background

Even though thorium was considered, since the beginning of the nuclear power development, to be the nuclear fuel to follow uranium, the use of thorium-based fuel cycles has been studied on a much smaller scale as compared to uranium or uranium/plutonium cycles. The technology to utilize thorium in nuclear reactors was thought to be similar to that of uranium. Although based on boundary conditions and needs quite different from the present ones, those studies have permitted, however, to identify many incentives for the use of thorium fuel. Thorium resources are larger than those of uranium, and neutron yields of ^{233}U in the thermal and epithermal regions are higher than for ^{239}Pu in the uranium/plutonium fuel cycle. The introduction of the thorium-based nuclear fuel cycle would therefore vastly enlarge the fissile resources by breeding ^{233}U. Large thorium deposits in some countries, coupled with a lack of uranium deposits in those countries is another strong incentive for the introduction of thorium-based nuclear fuel cycles. Other reasons identified in past studies are the potential for fuel cycle cost reduction, the reduction in ^{235}U enrichment requirements, safer reactor operation because of lower core excess reactivity requirements, and safer and more reliable operation of ThO_2 fuel as compared to UO_2 fuel at high burnup due to the former's higher irradiation and corrosion resistance.

The TMI and Chernobyl accidents, and growing long-lived radioactive waste issues provided new incentives for the use of thorium-based fuel cycles, given their potential for reducing the production of plutonium and higher actinides, as well as the possibility for a more effective incineration of plutonium and long-lived radiotoxic isotopes. On the other hand, the thorium fuel cycle has some disadvantages when compared with the uranium fuel cycle, which were also recognized from the very beginning of thorium-fuel related activities, more specifically: the thorium-^{233}U fuel cycle is characterized by a much stronger gamma radiation level than the uranium-plutonium cycle, and therefore handling during fabrication requires more care; nuclear reactions by neutron absorption and decay schemes for thorium-based fuels are more complicate; longer water storage time for the spent fuel is needed due to higher residual heat; potential difficulties in down stream spent fuel reprocessing.

Against this background, the participants in several consultants meetings held by the IAEA in 1994–1996 expressed the view that the thorium fuel cycle deserves further serious consideration and detailed evaluation of its potential advantages. To implement these recommendations, several activities were planned. Among them, a series of three meetings: the Advisory Group Meetings on Thorium Fuel Cycle Perspectives (Vienna, 16–18 April 1997) and on Thorium Fuel Utilisation: Options and Trends (Vienna, 28–30 September 1998), and the Technical Committee Meeting on Utilisation of Thorium Fuel; Options in Emerging Nuclear Energy Systems (Vienna, 15–17 November 1999).

The first Advisory Group Meeting focused on physics aspects of thorium fuelled cores, and discussed ensuing advantages and disadvantages of the thorium fuel cycle. In line with the conclusions of the first Advisory Group Meeting, the second one addressed mainly technological aspects of the thorium fuel utilization. Apart from current commercial reactors, the scope of the Technical Committee Meeting covered all types of evolutionary and innovative nuclear reactors, including Molten Salt Reactors and Hybrid Systems.

IAEA activities

Thorium fuel cycle related activities carried out by the IAEA include the preparation of status reports on advanced nuclear power technologies development, conduct of technical information exchange meetings, and collaborative R&D performed within the framework of Co-ordinated Research Projects (CRPs).

Status report on thorium-based fuel cycle

In the framework of IAEA activities on the use of thorium as nuclear fuel, a report on the performance of the thorium cycle entitled A fresh look at the thorium fuel cycle was drafted in 1991 and distributed as Working Material. As a follow up action, the preparation of a report on the status of the thorium-based fuel cycle was initiated to describe the state of the art of the thorium cycle, and to indicate areas which need further investigations. The report includes the general overview and summary of the thorium-based fuel cycle concepts, and contributions from the various countries/groups presenting in detail their concepts. A draft of the status report was reviewed and thoroughly discussed by a group of experts. IAEA-TECDOC-1155, Thorium Based Fuel Options for the Generation of Electricity: Developments in the 1990s, was published in 2000.

Co-ordinated Research Project (CRP) on the Potential of Thorium Based Fuel Cycles to Constrain Plutonium and to Reduce Long-term Waste Toxicities

This CRP examines the different fuel cycle options in which plutonium can be recycled with thorium to incinerate the plutonium, or replace it with materials that are less controversial. The potential of the thorium matrix is examined through computer simulations. Each participant chooses his own fuel cycle, and the different cycles are compared on the basis of certain predefined parameters (e.g. annual reduction in the plutonium inventory). The toxicity accumulation and the transmutation potential of thorium-based cycles for current, advanced, and innovative nuclear power reactors including hybrid systems is investigated. The CRP was launched in 1996, and 9 institutions from China, Germany, India, Israel, Japan, the Republic of Korea, the Netherlands, the Russian Federation, and the United States of America are participating. The final report of the CRP will be published by the IAEA in 2002.

Co-ordinated Research Project (CRP) on the Use of Thorium Based Fuel Cycles in Accelerator Driven Systems (ADS) to Incinerate Plutonium and to Reduce Long-term Waste Toxicities

The purpose of the CRP is to assess the uncertainties of the calculated neutronic parameters of a simple model of a thorium or uranium fuelled Accelerator Driven System (ADS) in order to reach a consensus on the calculational methods and associated nuclear data. The participants identified a number of issues which should be discussed to have a better understanding of the ADS and they agreed that some points are to be reviewed at a later stage through comparisons of the different approaches and tools used by the different groups. Computer code validation against experiments is also envisaged. The CRP was launched in 1996, and 10 institutions from Belarus, the Czech Republic, France, Germany, Italy, the Netherlands, the Russian Federation, Spain, Sweden, and CERN are participating. The final report of the CRP will be published by IAEA in 2002.

Status of thorium fuel option development in the Member States
(based on statements of the participants in the meeting in 1999)

Canada
Atomic Energy of Canada Ltd (AECL) has a very comprehensive program of work on thorium fuel cycles under way.

AECL has investigated many techniques for thorium fuel fabrication and has fabricated hundreds of thorium-based fuel elements. A number of full-sized, thorium CANDU fuel bundles (uranium-thorium, plutonium-thorium and pure thorium) have been irradiated at full power for years in the NRU research reactor. High quality thorium fuel bundles have been produced and the program of fuel production is continuing.

Measurements of fundamental physical quantities important to the physics of thorium fuel cycles have been undertaken in the ZED-2 critical assembly. Recently, a large code-development program was completed, allowing the performance of full-core fuel management simulations which accurately model the flux-dependence of thorium-based fuels. Lattice-cell and full-core fuel management studies have been performed of a variety of schemes for exploiting thorium fuel, and more studies are continuing. The properties of spent thorium fuel as a waste product have been studied and evaluated with respect to the Canadian nuclear fuel waste management program.

France
Rather than a continuous programme on thorium-based fuels, CEA had occasional involvement in thorium-related subjects. These include (in the 60's) an irradiation of thorium subassemblies in a power reactor, followed by reprocessing tests. More recently, CEA's involvement was mainly in the field of neutronics calculations of cores containing thorium, in view of either plutonium burning or long term thorium cycle sustainability:
- PWR with thorium-plutonium fuel or Th-^{233}U closed cycle
- Fast reactors with thorium-plutonium fuel or Th-^{233}U closed cycle
- ADS (like the Energy Amplifier, or molten salt reactors).

Furthermore, an irradiation with a thorium-plutonium pin is foreseen in the Phenix fast reactor; and a core configuration including Th elements is currently discussed for the MASURCA critical mock-up: this would e a subcritical core, fed by a 14 MeV pulsed neutron source, as part of the MUSE programme at MASURCA.

Finally, CEA has been involved in the 4th framework programme of the European Community (action named "Thorium as a waste management option"), providing fast reactor calculations. Similarly, CEA will be involved in an action of the 5th framework programme about thorium.

Germany
Germany, in the past, was one of the protagonists in view of thorium-based fuel. Its use was concentrated for a long time on the AVR (a pebble-bed high temperature research reactor) in Juelich and on the THTR (Thorium High Temperature Reactor), a 300 MW(e) pebble-bed prototype reactor.

The AVR was operated for more than 2 decades using HEU-Th-fuel. The reactor was operated with a coolant outlet temperature of 950°C, and the fuel achieved burnups of more than 140 000 MW·d/tHM. In the wake of the international non-proliferation requirements the decision was made to switch over to LEU-fuel.

The THTR was also operated with HEU-Thorium mixed oxide fuel, until it was shutdown mainly for political-economic reasons. Thus, a broad industrial experience existed in Germany in the field of the fabrication of thorium-based fuel. The Institute of Chemical Technology of the Research Center in Juelich was involved in the development of the so-called Thorex-process (thorium extraction). In the meantime, there has been a political decision to stop the fabrication and reprocessing of thorium fuel, and research activities are supposed to be concentrated on questions of reactor safety and on waste treatment.

However, since getting rid of plutonium seems to be a real political concern also in Germany, R&D institutions are now having a fresh look at the thorium fuel cycle the rationale being mainly to avoid the production and to burn plutonium. There is the intention to resume work on the thorium cycle, based on the experience gained in the past. A proposal issue of the bi- and tri-lateral co-operations on the reduction of weapons-grade plutonium was submitted to the EC for a test irradiation under PWR conditions. This irradiation is planned to start in 2001.

India

India has been pursuing a steady programme of thorium fuel cycle activities. The country is currently engaged in the design of an advanced heavy water reactor (AHWR) with the aim of utilising thorium for power generation. The aim is to produce 75% of the power from thorium. This reactor incorporates several advanced passive safety features while utilising the expertise existing on pressurised heavy water reactors (PHWRs).

India has a continuous programme of irradiating thorium in research reactors. Thoria rods have been irradiated in the annulus of the CIRUS reactor. Several fuel assemblies have been irradiated in the in-pile test loops for testing and qualifying advanced fuels of (U,Pu)MOX and (Th,Pu)MOX, and many such irradiations are in the pipeline. The research reactor PURNIMA, utilising ^{233}U fuel, has been developed for neutron investigation studies. The 30 KW research reactor KAMINI, which uses plate type ^{233}U-Al alloy as fuel is currently in operation.

Thoria bundles have been used for power- flattening in the initial core of the standardised Indian PHWR and the RAPS type of PHWR. There is an ongoing programme for continuous irradiation of thorium in PHWRS.

All the fuel cycle aspects of thorium are under study. A programme of post irradiation examination (PIE) of the spent fuel from the Thorium irradiations in the research and power reactors has been initiated. The issues of back end of the thorium fuel cycles are also being addressed with particular reference to the handling of highly radioactive spent thorium fuel.

Japan

There has been no authorized activity on the Th fuel cycle in Japan, since the government intends to promote the U-Pu fuel cycle. Activities are restricted to basic studies mainly in universities. However, there are many activities in Japanese universities. Some of them are supported by Grant-in-Aid for Scientific Research from the Ministry of Education, Science, Sports and Culture. Recently, a preliminary study was initiated to investigate the thorium fuel cycle and the accelerator driven subcritical reactor. This study was supported by research for the Future Program from the Japan Society for the Promotion of Science (JSPS). However, it encountered great difficulty to get a sufficient budget for the full promotion of research mentioned above, since the thorium fuel cycle is now out of scope in the present long term plan for the atomic energy development in Japan. At the Kyoto University Research Reactor Institute (KURRI) a future plan has intensively been investigated since 1997 to promote the

joint programme among university researchers. In this future plan, a basic study on the thorium fuels cycle is considered to be a main research objective as well as that on the accelerator driven system (ADS). The Kyoto University Critical Assembly (KUCA) is expected to be an important facility to perform critical experiments by purchasing the denatured ^{233}U fuel from the United States, if possible. The reactor physics experiments related to the ADS are also expected to be performed in the KUCA to establish a new neutron source for the joint programme as a substitute for the Kyoto University Research Reactor (KUR). In the Japan Atomic Energy Research Institute (JAERI) a study on the development of the rock-type fuel containing Th is performed for the transmutation of TRU elements.

At present, the discussion on the new long term plan for the atomic energy development in Japan is underway in the Atomic Energy Committee. Since there are only few energy resources in Japan, it is inevitable to extend available energy resources including fertile ones in order to match future energy demands. From this point of view, we are expecting that some positive statements for the development of the thorium fuel cycle will be included in the document of new long term plan for the atomic energy development in Japan.

Republic of Korea
As Korea has not yet decided with respect to the future of spent fuel recycling technology development, comprehensive plans or activities regarding thorium fuel cycle applicable to power reactor are not available yet. Meanwhile, as a part of the long term national nuclear R&D program, the advanced PWR project incorporates design simulation studies of various ThO_2-UO_2 mixed cores. The main objectives of these studies are on uranium resource savings and less radiotoxicity. Another national project performed at the Korea Atomic Energy Research Institute, which deals with the possibility of thorium fuel cycle, is the HYPER project, an accelerator-driven subcritical system. Molten salt or metallic fuel with thorium are considered as candidate materials.

In universities, also supported y the government, there are a variety of basic research projects associated with thorium composition materials as the fuel for PWR, CANDU, ADS and MSR. Development of the AMBIDEXTER concept is a typical case of this.

Netherlands
The Netherlands believe that the thorium cycle offers challenging options for nuclear waste reduction, both at the front end of the fuel cycle and at the back end of it. The main interest of the Netherlands is to attain data for application of the thorium cycle in existing LWRs but also, on the long term, in ADS and fast reactors. Emphasis lays on the burning of plutonium along with thorium with the aim to reduce the lifetime of nuclear waste.

This so-called thorium-assisted plutonium burning has to be considered as the pursuit of two objectives at the same time. On the short term, the objective refers to the burning of plutonium at rates that are much higher than in existing $(U,Pu)O_2$-MOX fuelled LWRs. On the long term, however, possibilities are created to embark on a self sustaining thorium cycle when time is ready for it.

To achieve a high Pu/TRU burning, the irradiation behaviour of Th/Pu fuel at high burnup has to be examined. In addition to that vital data for geological disposal, scenarios need to be determined, and new waste minimising routes for reprocessing of thorium-based fuel need to be addressed.

As the Netherlands is aware of the fact that aforementioned subjects cannot be investigated on its own, our country adheres to the strategy to do this research in the framework of the European Union. This can be illustrated by the fact that the Netherlands joined the EU 4th framework programme and performed the co-ordination of the project called "Use of Thorium Cycle as a Waste Management Option". As a continuation of this programme the Netherlands and other member countries submitted a proposal for the 5th framework, which bears the title "Thorium Cycle: Development steps for PWR and ADS Applications". If this proposal will be accepted, the Netherlands will perform the co-ordination of the project and in addition will carry out calculations on Th-fuelled cores, Th-fuel pellet fabrication, generation of essential nuclear data, irradiation in the High Flux Material Testing Reactor and postirradiation experiments.

Russian Federation

In the Russian Federation, research work on the thorium fuel cycle began nearly at the same time as the work on the uranium based one. Of course, the relative scales of these research works were incomparable. But recently the peculiarities, problems and perspectives of the thorium-based fuel cycle have been discussed more actively. Minatom's institutes, the Kurchatov Institute, and the other institutes of the Russian Federation, virtually have not stopped independent research in the area of the thorium cycle, and, are currently supporting them by new works on reactor concepts, the physics of thorium systems, and technology. The list of recent publications in this area can be found in the paper presented at this meeting.

Conceptual investigations on the thorium fuel cycle are performed in the Kurchatov Institute (VVERT reactor, MSR, HTGR), IPPE (WWER type reactors, FR, MSR), VINIEF, ITEF (HWR, ADS). The technological problems of the thorium fuel cycle are studied and developed in the Belarus Institute, Radium Institute, IPPE, Kurchatov Institute, NIIAR.

The experimental studies were conducted on critical facilities and power reactor. Considerable work is still pending: analysing the experimental results, obtained from critical facilities and reactors, as well as establishing the models for checking the nuclear data libraries and the calculation methods employed.

Because of the present budget difficulties, the institutes focused only on the theoretical and calculational investigations of the thorium fuel cycle, including conceptual core designs, and the handling of experimental data which was obtained previously. The experimental investigations on fuels is performed on a laboratory scale using only samples of materials.

Up to now there were no national or industrial programmes on the thorium fuel cycle. To co-ordinate research performed separately, MINATOM entrusted in 1999 the State Scientific Centre of the Russian Federation - Institute of Physics and Power Engineering with the duties of head organization of the nuclear industry as regards the problems of the thorium fuel cycle.

Turkey

In Turkey, where recovering the ~1% thorium content of the ore is not economic by itself, but in conjunction with rare earths could be viable. The Turkish Atomic Energy Authority is highly interested in thorium-based fuel studies. In the past it has produced thoria and thoria-urania fuel pellets, and studies on thorium-based fuels are continuing. Urania and urania-gadolinia fuels could be prepared by a sol-gel process to form pellets for sintering. These could then be coated with B_4C as a non-interactive five-micron layer by hydrogen reduction of a carbon tetrachloride-boron trichloride mixture. Such a layer could be more effective than BN with diminished production of ^{14}C.

United Kingdom

On thorium fuel-cycle development:

BNFL remains unconvinced by current arguments for using thorium fuels except in special circumstances, and for operational reasons cannot use existing uranium plant in the UK to fabricate them. However, through its interest in Westinghouse, the company is linked with studies on thorium possibilities. In principle it does not rule out more active future involvement subject to demonstration of clear benefits with technical and economic viability.

BNFL is also a partner in proposals under the European Commission 5th Framework Nuclear Fission Programme for work on (a) a reprocessing flowsheet and (b) an accelerator-driven HTR-type minor-actinide burner.

On specific projects:

As a partner in two proposed 5th Framework studies, BNFL proposes respectively to (a) advise and assist in the development needed to transform laboratory studies into a potential industrial reprocessing operation, and (b) apply its expertise in waste management to preparing irradiated HTR-type fuel compacts for disposal.

On advantages of the thorium cycle:

BNFL recognises that thorium is proposed as an effective matrix for the purpose of utilising plutonium constructively, an objective that the company actively supports and practises in other ways. BNFL does not believe that any reduction in civil plutonium stocks within the foreseeable future can significantly affect the issue of weapon proliferation, and the use of thorium for this purpose presents significant new operational difficulties.

Thorium might serve as a matrix for fissioning or transmuting minor actinides if this should be required. The necessity and case for this objective, or for preferring thorium to other matrices, both remain to be established.

In the long term thorium may well provide a means of resource extension, in parallel with fast-reactor developments rather than as a substitute for them. Meanwhile the most valuable contribution to industry that it can make appears likely to be in extending fuel irradiation where recycling of recovered fissile and fertile values is considered impracticable or undesirable.

United States of America

DOE is conducting four projects involving use of the thorium fuel cycle. All four projects are based on an once-through, proliferation resistant, high burnup, long refuelling cycle use of thorium in a light water reactor. Three of these projects are part of the Nuclear Energy Research Initiative (NERI) program. These are: “Advanced Proliferation Resistant, Lower Cost Uranium-Thorium Dioxide Fuels for Light Water Reactors”, with INEEL as the lead organization; “Fuel for a Once-Through Cycle (Th, U)O2 in a Metal Matrix”, with ANL as the lead; and “A Proliferation Resistant Hexagonal Tight Lattice BWR Fuel Core Design for Increased Burnup and Reduced Fuel Storage Requirements”, with BNL lead. The fourth project is “The Radkowsky Thorium Fuel (RTF) Project”, also under BNL lead. Also of interest is the DOE Accelerator Transmutation of Waste (ATW) program. The ATW Roadmap for a five year R&D program on the ATW has received final approval in mid-November 1999.

European Commission
EURATOM 5th Framework Programme for Research and Training in the field of Nuclear Energy (1998 – 2002).

The EURATOM 5th Framework Programme (FP5) comprises two distinct parts:
1) a "direct" action; and
2) an "indirect" action.

The indirect action is the main mechanism for Community research, development and demonstration activities, and is mainly executed by calls for proposals, whereas the direct action is complementary to the indirect action and is carried out by the Joint Research Centre (JRC).

The indirect action programme comprises a key action on controlled thermonuclear fusion and a key action on nuclear fission. It also includes generic research in the fields of radiological sciences, support for research infrastructure, training activities and accompanying measures.

The major topics of research in the key action on nuclear fission are:
1) Operation Safety of Existing Installations,
2) Safety of the Fuel Cycle,
3) Safety and Efficiency of Future Systems, and
4) Radiation Protection.
5) The generic research on Radiological Sciences comprises topics on Radiation Protection and Health,
6) Environmental Transfer of Radioactive Material,
7) Industrial and Medical Uses and Natural Sources of Radiation; and
8) Internal and External Dosimetry.

The support for research infrastructure includes
1) Access to Large Scale Facilities,
2) Networks, and
3) Data Bases and Tissue Banks.

Training activities includes:
- Training Fellowships,
- Special Training Courses,
- Grants for Cooperating with Third Countries, and
- Research Training Networks.

The total budget for the nuclear fission programme in FP5 is set at 281 million Euro.

Conclusions

The papers and statements of the delegates presented at the three IAEA meetings of which the proceedings are summarized in this publication, underline existing and growing interest in many IAEA Member States to investigate the potential of advanced thorium fuel cycles and the related reactor technologies.

The papers addressed the main physics aspects of thorium fuelled reactor cores, assessed advantages and disadvantages of thorium fuel utilization, presented the various options and concepts under investigation, and reviewed remaining problems and uncertainties linked to thorium fuel utilization and reactor technologies based on thorium fuel. Presentations, statements and discussions identified as main reasons for the renewed interest in thorium fuel cycles two issues: the potential to incinerate plutonium and reduce actinide production, on the one side, and better material properties and fuel behaviour, on the other side. However, the most important conclusion to be drawn is that there is a need for a unified systematic approach in assessing thorium fuel utilization: a methodology (metrics) to evaluate the performance parameters of the thorium fuel cycle must be developed. This methodology would define the performance parameters matrix as well as the algorithms for the evaluation.

Consensus was also reached on the necessity to develop and maintain a database of all available information relevant to thorium fuel cycles, their utilization and related reactor technology. It was also pointed out that there are considerable uncertainties with regard to future thorium fuel prospects and associated reactor technology development needs.

I. INTERNATIONAL AND NATIONAL CONCEPTS ON FUTURE THORIUM BASED NUCLEAR FUEL CYCLES IN COMPARISON WITH OTHER FUEL CYCLES

BRAZILIAN EXPERIENCE ON THORIUM FUEL CYCLE INVESTIGATIONS*

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Abstract. Brazilian systematic investigations on the use of thorium fuel cycles in nuclear power reactors started in 1965. During the 60's and early 70's the work was mainly concentrated on the thorium utilization in heavy water reactors (HWRs). This work was performed in the framework of a cooperation agreement with the, French CEA. In the frame of the International Nuclear Fuel Cycle program, Brazil started in 1979 an R&D program on the thorium utilization in pressurized water reactors (PWRs), within the scope of a Brazil-German cooperation agreement. This program lasted for almost ten years. The activities of both programs wore performed at the CDTN nuclear technology development center, in Belo Horizonte. More recently two Brazilian institutions restarted again investigations on the thorium fuel cycle. At CNEN, work is being pursued to further investigate the possible utilization of thorium in a PWR (Angra 1). The purpose of the paper is to comment briefly the main results of the past work and to give emphasis on the description of the recent results and new plans related to the work on thorium utilization in power reactors.

1. INTRODUCTION

The major incentives for thorium use in power reactors in Brazil were and still are:

- the estimated large thorium resources of the country (see Appendix), and
- the improved fissile fuel utilization of the thorium fuel cycle in thermal reactors (due to the higher η value of the ^{233}U as compared to that of ^{239}Pu), which results in a better utilization of the uranium reserves as well as in a reduction of the uranium enrichment requirements.

Today, the non-proliferation characteristics of the thorium fuel cycle, for instance:

- the contamination of the ^{233}U by the ^{232}U and its daughter products, some of them being hard gamma emitters;
- the capability of the ^{233}U to be denatured with ^{238}U; and
- the reduction of plutonium buildup, if considered a future mix of thermal reactors operating in the U/Pu cycle and of Th-fueled reactors (and possibly even the burning of plutonium).

add a new incentive.

2. PAST EXPERIENCE ON THORIUM FUEL UTILIZATION

The most important activities in Brazil aiming at the introduction of thorium-fuelled power reactors in the long term were developed in two different occasions by different institutions, in the framework of two projects in close cooperation with international partners, as shown in Table I.

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* 1997 meeting.

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Table I. Brazilian Projects on Thorium Fuelled Power Reactors

Period	Institutions	Project	Country partner
From mid 1965 to mid 1970	CNEN & UFMG	"Instinto/Toruna"	France
From mid 1979 to mid 1988	NUCLEBRÁS	"Thorium Utilization in PWRs"	Germany

CNEN - Comissão Nacional de Energia Nuclear (Brazilian National Nuclear Energy Commission);

UFMG - Universidade Federal de Minas Gerais (Federal University of Minas Gerais);

NUCLEBRÁS - Empresas Nucleares Brasileiras S.A.

The Instituto de Pesquisas Radioativas - IPR (Institute for Radioactive Research), in Belo Horizonte, later renamed Centro de Desenvolvimento da Tecnologia Nuclear - CDTN (Nuclear Technology Development Center), was in charge of both projects.

2.1 "Instinto/Toruna" Project

The first project was developed by the so-called Thorium Group, in the framework of a cooperation agreement with the French Commissariat à l'Energie Atomique - CEA. It was motivated by the results of a long term study of fuel requirements [1], one of the tasks of the Study's Committee for the first Brazilian Power Reactor, created by the Presidency of the Republic in 1965 and coordinated by the Brazilian National Nuclear Energy Commission CNEN. This project was ambitious and aimed at the development of an indigenous thorium-fuelled pressurised heavy water reactor concept with prestressed concrete reactor vessel. It was scheduled in the three phases shown in Table II.

Table II. "Instinto/Toruna" Project Phases and Objectives

Phase	Main Objective
From 1966 to 1967	Evaluation of a thorium-fuelled pressurised heavy water reactor - PHWR concept ("Instinto Project")
From 1968 to 1971	Research and Development and conceptual design of a natural uranium fuelled PHWR concept ("Toruna Project")
From 1971 on	Development of a PHWR prototype (eventually)

The studies of the first phase were completed with the release of a Final Report with positive conclusions and recommendations which led to the continuation of the project [2].

The second phase was oriented towards the conceptual design of a natural uranium fuelled PHWR, required for the production of fissile material (plutonium) for the startup of the thorium fuel cycle. This work was concluded also with a Final Report [3, 4]. This phase contemplated also a strong effort on research and development in different areas, such as fuel technology, core physics design, reactor and plant thermal-hydraulics design, reactor vessel design, materials and components testing, and fuel cycle economics. Formation and training of personnel had a major incentive. Several facilities and laboratories were designed and implemented at the IPR institute: the heavy-water subcritical facility ("Capitu"), the

experimental thermal-hydraulic loop ("CT1"), the Fuel and Materials Laboratory, and the Components Testing Laboratory.

The continuation of this project in its third phase, which included a greater R&D effort (design and construction of a critical facility and a high power thermal-hydraulic loop) aiming at the design and (eventually) the construction of a PHWR prototype, was not implemented, due to the decision of adopting the pressurised light water reactor - PWR for the Brazilian first nuclear power plant (NPP). The reasons for the choice of the PWR for Angra 1 and the following NPPs, as well as a short description of the "Instinto/Toruna" project, which may be considered as an important contributor to the genesis of the Brazilian NPP program, are given in reference [5]. The main results of this project may be summarised as:

- the assessment and own development of know-how related to the technology and economics of water-cooled reactors, which made it possible to give a strong support to the Brazilian NPP implementation program, based on light water reactors; and, perhaps one of the most fruitful results,
- the formation of a staff highly qualified in different aspects of the analysis and design of nuclear power plants, later requested by the different organisations involved in the implementation of the nuclear program (Angra 1 and Angra 2/3 NPPs; formation and training of personnel; and research and development).

2.2 R&D Program on the Thorium Utilization in PWRs

The second project was developed in the framework of a cooperation agreement between Brazil and Germany. The motivation was directed towards the improvement of knowledge in the field of thorium utilisation in power reactors aimed at fulfilling the "Governmental Agreement on Cooperation in the Field of Science and Technology" (1969) and the "Memorandum of Understanding between the Kernforschungszentrum Jülich, GmbH - KFA and the Empresas Nucleares Brasileiras S.A - NUCLEBRÁS" (1978). The "International Nuclear Fuel Cycle Evaluation - INFCE", organized at the same occasion (1977), was an incentive to the development of the joint CDTN, KFA Jülich, Siemens A.G./Group KWU and Nukem GmbH R&D program. The general objectives of the program were:

(a) to analyse and prove the thorium utilisation in PWRs,
(b) to design the PWR fuel element and reactor core for the different thorium fuel cycles,
(c) to manufacture, test and qualify Th/U and Th/Pu fuel elements under operating conditions, and
(d) to study the closing of thorium fuel cycles by reprocessing of spent thorium-containing PWR fuel elements.

Technology transfer by joint work on the different tasks presented another major objective of the program. Three phases were foreseen at the start of the program (Table III).

The first phase was concluded in 1983 and the results were published in a Final Report [6]. The program was terminated in the second phase, by mid 1988, after nine years of successful cooperation before entering the pathfinder demonstration phase with a $(Th,U)O_2$ fuel bearing test assembly in a commercial PWR (Angra 1) [7].

Table III. Phases and Objectives of the "R&D Program on Thorium Utilisation in PWRs"

Phase	Main Objective
From 1979 to 1982	Adaptation of the existing methods and technologies to PWR thoria fuels and irradiation testing of $(Th,U)O_2$ fuel rods in a test reactor
From 1982 to 1986	Research and development effort concentrated on the demonstration of the behaviour of $(Th,U)O_2$ fuel in a power reactor
From 1986 on	Demonstration of $(Th,Pu)O_2$ fuel

The results of the program, discussed deeply in the program Final Report [8], confirmed in detail that the developed thoria based fuels, produced by merging of the standard light water reactor palletising process with the chemical ex-gel process developed for the high temperature reactor fuel, can be used in present PWRs. No changes in the fuel assembly and in the core design are needed. This holds both for $(Th,U)O_2$ and $(Th,Pu)O_2$ fuels in 3- and 4-batch operation. The latter shows high burnup potential beyond the four-cycle scheme. In this case, the inserted fissile plutonium is strongly depleted and the once-through put-away cycle becomes very attractive. As far as the technology development and transfer for the Th/U fuel are concerned, the program objectives were accomplished. However, large scale demonstration of Th/U fuel in a power reactor, fabrication and qualification of Th/Pu fuel as well as closing the fuel cycle would require substantially more effort. The main conclusions of this Brazilian-German joint program were:

(a) The utilisation of thorium in PWRs presents a long-term option providing in some respects interesting results. The most attractive application of Th-based fuels is the use of recycle plutonium in an extended burnup once-through fuel cycle.
(b) From the point of view of cooperation and technology transfer, the program experience showed the importance of using hardware oriented goals, clear definitions of required outputs and sufficient communication including joint work on interacting tasks.

2.3. Other Activities

Other activities on thorium utilisation took place in other Brazilian institutions also. For instance, from mid 60's to end of the 70's, the Instituto de Pesquisas Energéticas e Nucleares - IPEN (Institute for Energy and Nuclear Research), in São Paulo, has developed experimental activities on thorium-fuel technology [9, 10]. In the 70's, the IPEN spent a great deal of effort in a series of studies of the High Temperature Reactor concept [e.g. 11, 12]. It has, in addition, developed some activities (M.Sc. thesis work) on thorium utilisation in PWRs and in gas cooled fast breeder reactors (feasibility study of a subcritical assembly and evaluation of thorium metal blankets) [13, 14].

The Instituto de Estudos Avançados - IEAv (Institute of Advanced Studies) of the Centro Tecnológico Aeroespacial - CTA (Aerospace Technology Center), in São José dos Campos, spent some effort in the 70's and 80's in studying thorium-fuelled both gas-cooled and sodium-cooled fast breeder reactor concepts [15, 16, 17].

3. RECENT ACTIVITIES AND PLANS

Several activities involving thorium utilisation in power reactors are either under way or being discussed.

3.1 Determination of Fuel Diffusion Properties

The transport properties of nuclear materials are very important to better understand the different phenomena influenced or controlled by them, such as oxidation and reduction, sintering, mechanical deformation at high temperature (fluence), grain growth, and densification. For the fuel designer it is necessary the knowledge of the autodiffusion and the heterodiffusion coefficients, in order to simulate properly, by using complex computer models, the fuel behaviour under power reactor conditions.

In addition to that, the published values of the transport properties present, in general, a high dispersion, which makes difficult the establishment of criteria to select the diffusion coefficients to use in the fuel design calculations. This was the motivation for the CDTN research center and the Department of Physics of the Universidade Federal de Ouro Preto - UFOP (Federal University of Ouro Preto) to join their efforts, since 1992, to develop a research aiming at the determination of the diffusion properties of nuclear materials manufactured in Brazil. In a first step, the anionic and cationic diffusion properties of the following ceramic materials are being studied:

- uranium dioxide fuel: UO_2,
- uranium/gadolinium mixed oxide $(U,Gd)O_2$, a burnable poison,
- uranium/cerium mixed oxide $(U,Ce)O_2$, a simulator of uranium/plutonium mixed oxide $(U,Pu)O_2$,

This step, from which first results are already available [e.g. 18 and 19], will be followed by the study of the diffusion properties of

- thorium oxide, thorium/uranium and thorium/cerium mixed oxides: ThO_2, $(Th,U)O_2$ and $(Th,Ce)O_2$.

Due to the multidisciplinary aspects and the complexity of the techniques involved in the determination of these properties (e.g. making use of the secondary ions mass spectroscopy technique - SIMS), this project has gained the support of other laboratories, in particular of the French laboratories: Laboratoire des Composés Non-Stoechimétriques, Université Paris XI, Orsay, and Laboratoire de Physique des Solides, Centre National de Recherche Scientifique - CNRS, Belleville-Meudon.

3.2 Energy Scenarios and Thorium-Fuelled MSBRs

Investigations of scenarios for long term energy requirements and the introduction of breeder reactors in Brazil are being studied at the School of Engineering's Nuclear Engineering Department (DEN) of the Federal University of Minas Gerais - UFMG, in Belo Horizonte [20].
Considering the fact that the primary energy consumption per capita is a good index of the overall development of a country, since it reflects all desirable components of the development (e.g., the social, economical and technological components), the study made assumptions for three long term global energy demand scenarios (Table IV):

Table IV. World Energy Demand Scenarios

Average Energy Demand	1990	2015	2060 I	2060 II	2060 III
kW per capita	2.55	2.48	3.0	4.0	5.0

The following assumptions were made for Brazil:

(1) The population growth projection in the period 1990-2100 is summarised in Table V; it was assumed that the population becomes stabilised in about 280 million inhabitants by the end of the next century.

(2) In order to achieve a minimum reasonable development of the country in the long term, the primary energy consumption per capita projection in 2060 was assumed to be *at least* equal to the global average energy consumption per capita of the conservative Scenario I (Table IV).

With these assumptions the total long term energy requirements were derived, as shown in Table V. The most important Brazilian energy resources are hydro, nuclear (uranium and thorium) and biomass (with a great production yield due to the very favourable location in the tropics). Fossil resources, oil in particular, are rather limited.

The study considered the indigenous energy resources and reserves presented in Table VI:

From the results showed in the above tables the following conclusions are derived:

(1) The nuclear energy that could be produced by uranium and thorium, *if totally used*, is by far the largest non-renewable energy resource (not only in Brazil but worldwide also);

(2) The long term energy requirements cannot be supplied by hydro power and by indigenous fissile fuels only, if reactors of poor fuel utilisation (like the LWRs) are considered[*].

(3) Use of biomass and of uranium and thorium *in breeder reactors* seem to be essential in order to fulfil the forecasted energy demand.

Following these conclusions, the DEN started to make a comparison between different breeder reactor concepts as possible candidates for the long term electricity supply. Part of this work is being done as a series of M.Sc. thesis work, which includes also energy analysis of the nuclear fuel cycles.

Although the development of the sodium-cooled fast neutron breeder reactor (FBR) has had a strong support in several countries, culminating with the operation of several prototypes, demonstration reactors, and even of a large nuclear power plant, the full commercialisation of this reactor concept is not expected to occur earlier than the second quarter of the next century. Taking this into account, the DEN considered that the molten salt-cooled thermal breeder reactor concept (MSBR), originally developed in the USA [22] and now being pursued in Japan [23], could be a possible breeder reactor candidate for the long term energy supply.

[1] The available Brazilian uranium reserves amount to 301,490 tU_3O_8. About 2/3 are measured and indicated reserves and 1/3 are inferred ones.

Table V. Brazil - Population Growth and Energy Consumption Scenarios

Year	Population, 10^6 inhabitants	Energy Consumption, kW/cap	Yearly Total Energy Requirements, TW/a	Q/a [1]	Total Cumulative Energy Requirements, Q Period Total	
1989	147	1.84	0.27	8.062	-	-
2025	226	2.48	0.56	16.72	427	427
2060	264	3	0.79	23.64	768	1,195
2100	280	3	0.84	25.08	974	2,169

[1] Q - Quad: 1 Q = 10^{15} Btu.

Table VI. Brazil - Indigenous Energy Resources

Resources	10^6 tEP	Q	TW
Depletable (Fossil) - Total [a,b]	**4,894**	**209.7**	**7.08**
Sustainable - Nuclear (Fission) [a]**:**			
– Uranium in light water reactors (LWRs)	1,342	57.5	
– Uranium in LWRs with recycling (+ 35%)	1,813	77.6	
– Uranium in breeder reactors [c]	107,360	4,600	
– Thorium in breeder reactors [d]	214,720	9,200	
– **Nuclear (fission) - Total**		**14,012**	**473,08**
Renewables:			
– Hydro [f]	271.0/a	11.4/a	0.38/a
– Biomass - excluding liquid fuels	200.0/a	8.4/a	0.28/a
Renewables - Total [g]		**63.0/a**	**2.13/a**

[a] Source: BEN-1990 [21].
Remark: Other energy sources, sustainable and renewables, were not considered due to its present low potential
[b] Oil, natural gas, coal "in situ", shale and tar (assuming 100% resource recovery).
[c] Uranium reserves taken from BEN, assuming 70% recovery and 20% recycling losses. The uranium reserves may be higher, since only part of the country has been surveyed (aerogeographical survey: 60%; terrestrial survey: 40%).
[d] Assuming 20% recycling losses. Thorium reserves were assumed to be at least twice those of uranium. As it is known, Brazil is a country with high thorium resources, but no systematic survey has been done until now.
[f] Assuming that all hydro potential will be used.
[g] Including ethanol (without co-products or sugar production), vegetable oil, hydrocarbons (from Euphorbias in semi-arid regions), and assuming, for each case considered, a land use equivalent to 10% of the national territory. Energy products from biomass may use land not useful for agriculture.

A comparison between the FBR and the MSBR is now under way, considering the different characteristics of both concepts, including:

- core physics (conversion and breeding factors, doubling time, initial fissile inventory),
- thermodynamic efficiency,
- fuel cycle flexibility,
- safety,
- economics, and
- non-proliferation aspects.

Preliminary results of this comparison have recommended to follow up the MSBR development abroad, as well as to start more detailed investigations of this reactor concept as a possible alternative to the FBR.

3.2 Proposal of a Sodium-Cooled Research Reactor

Recently, as a continuation of the studies mentioned in 2.3 above, the IEAv made a proposal to study a low power sodium-cooled research reactor with a moderated neutron spectrum. This would be a first step of using sodium in a reactor system in Brazil, looking ahead towards the long term penetration of fast reactors, possibly using thorium [24].

3.3 Proposal of Thorium-Fuelled PWR Lattice Experiments

The IPEN institute and the Centro Tecnológico da Marinha (Navy Technology Center) in São Paulo - CTM-SP made a joint proposal to CDTN, now under discussion, of continuing the investigations of the thorium fuel cycle, with emphasis on fuel development and on lattice experiments with ThO_2 and/or $(Th,U)O_2$ fuel in their critical facility IPEN/MB-01 [25]. This critical facility was designed for experiments with typical PWR lattices, but it is quite flexible to accept different geometries and different fuel/absorber materials.

Preliminary reactivity calculations were performed for lattices containing ThO_2 and $(Th,U)O_2$ rods, with the uranium enriched at 5 w/o. Three square arrays with rods containing thorium inserted in the central part of the UO_2 lattice were investigated (Table VII).

Table VII. Thorium Rod Arrays for Lattice Experiments

Material	Rod Array	Approx. number of rods[a]
ThO_2	8 × 8	64
$(Th, 25w/oU)O_2$	10 × 10	100
$(Th, 50w/oU)O_2$	12 × 12	144

[a] Some positions of the central part of the lattice are occupied by control and safety rods.

The cross sections of the fuel cells were generated with the HAMMER-TECHNION code [26] and the effective multiplication factors (k_{eff}) were calculated with CITATION [27]. These number of rods correspond to the maximum allowable, since an increase in the thorium containing region in any of the three arrays would make the core subcritical. These calculations were performed in order to estimate the materials requirements.

The general objective of the proposed lattice experiments is to get acquainted with mixed Th/U cores, by means of measuring, e.g.:

- neutron spatial flux distributions in mixed Th/U cores;
- temperature reactivity coefficients;
- reactivities of ThO_2 and of $(Th,U)O_2$ rods;
- reaction rates inside these rods.

To perform the proposed lattice experiments a great effort should be spent in the production of Th and Th/U fuel pellets and of the fuel rods. A cost/benefit analysis of this proposal shall be made before a final decision.

4. BARRIERS FOR THE INTRODUCTION OF THORIUM FUEL CYCLE

The major barrier to the introduction of the thorium fuel cycle in Brazil is the fact that the startup of the thorium fuel cycle will require an initial inventory of fissile material. This means that, either an enrichment facility with the capability for the production of highly enriched uranium or the closing of the U/Pu fuel cycle with the implementation of a reprocessing plant for obtaining plutonium, is required. Both alternatives face a series of problems, which can be summarised as:

- technology development,
- high capital investment, and
- nonproliferation policy.

In this respect it is worth to mention that the nuclear power policy in Brazil is following the recommendations of the Commission for the Evaluation of the Brazilian Nuclear Program, established in 1986 by a Decree of the President of the Republic. The Final Report with the evaluation and the recommendations of the Commission was published by the Brazilian Academy of Sciences in 1990. One of the recommendations states: "To postpone the implementation of the spent fuel reprocessing project as it was originally planned, in view of its high costs and because it is not required in the medium term." As a consequence of this recommendation, the NUCLEBRÁS' Reprocessing Project was interrupted and the staff was dispersed [28].

In practice, this means that the Brazilian government does not include the closing of the nuclear fuel cycle as a medium term option. This of course will change in the long term, particularly if the need of using thorium as an energy source becomes a requirement.

5. CONCLUSIONS

From the effort already spent in Brazil on investigating the thorium use in power reactors and from the gained experience, it can be concluded that:

- The utilisation of thorium in power reactors presents an important long-term option.
- The long-term energy requirements cannot be supplied by hydro power and by indigenous fissile fuels only, if reactors of poor fuel utilisation (like the LWRs) are considered alone. Use of biomass and of uranium and thorium in breeder reactors seem to be essential in order to fulfil the forecasted energy demand.
- The most attractive application of thoria-based fuels in pressurised water reactors is the use of recycled plutonium in an extended burnup once-through fuel cycle. However, the large scale demonstration of thorium containing fuel in a power reactor, the fabrication and qualification of thorium/plutonium fuel as well as the closing of the fuel cycle would require substantial effort of R&D and high investment costs.
- The benefits of working in partnership with the share of experience and costs and the importance of interacting tasks and good communication have been demonstrated.
- The adoption of well defined hardware oriented goals, clear definitions of the outputs and continuous effort are important factors for the success of the project.
- The systematic follow up of the worldwide developments oriented towards the thorium utilisation in power reactors is important in order to subsidise future decisions. This also applies to the demonstration and developments of breeder reactors.

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APPENDIX

THORIUM RESERVES IN BRAZIL

A recent survey was made for the "Diagnosis Project" of the Brazilian Association of Metallurgy and Materials - AMB [29]. Reference [25] gives the following data for thorium reserves in Brazil adopted in this survey:

Table A-I. Thorium Reserves in Brazil

Reserves, in t ThO_2	**Indicated**		**Inferred**	
	1972	**1992**	**1972**	**1992**
Brazil	10,000	606,000	20,000	700,000

Source: IAEA

The Brazilian thorium reserves are not easily comparable to other international categories, whose extraction and processing costs range from US$10 to US$20 per kg of ThO_2. In Brazil, thorium resources are considered those deposits of detritic monazite along the coast (monazite sands) which can be explored economically for the production of monazite and its associates. Rare earth salts (as main product) and thorium oxide (at a cost lower than US$20/kg ThO_2) are obtained from the monazite. This last reference presents also the Table A-II for the natural potential resources of thorium in Brazil:

Table A-II. Thorium Potential Resources in Brazil

Occurrence	**Associated Mineral**	**Average Content, %**	**Resource, t ThO_2**	
			Measured	**Estimated**
Coastal deposits	Monazite	5	2,250	-
Morro do Ferro (State of MG)	Thorite and others	1 to 2	35,000	-
Barreiro, Araxá (MG)	Pyrochlore	0.09		1,200,000
Área Zero, Araxá (MG)	Pyrochlore	0.09	30,000	-
Alluvial and pegmatite deposits	Monazite	5	3,000	2,500
Total			73,750[a]	1,202,500

[a] Including 3,500 t of monazite sand of Indústrias Nucleares do Brazil S.A.- INB (in the States of RJ, ES)

THORIUM FUEL-CYCLE STUDIES FOR CANDU REACTORS*

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Abstract. The high neutron economy of the CANDU reactor, its ability to be refuelled while operating at full power, its fuel channel design, and its simple fuel bundle provide an evolutionary path for allowing full exploitation of the energy potential of thorium fuel cycles in existing reactors. AECL has done considerable work on many aspects of thorium fuel cycles, including fuel-cycle analysis, reactor physics measurements and analysis, fuel fabrication, irradiation and PIE studies, and waste management studies. Use of the thorium fuel cycle in CANDU reactors ensures long-term supplies of nuclear fuel, using a proven, reliable reactor technology.

1. INTRODUCTION

The CANDU® reactor has an unsurpassed degree of fuel-cycle flexibility, as a consequence of its channel design, excellent neutron economy, on-power refuelling, and simple fuel bundle [1]. These features facilitate the introduction and full exploitation of thorium fuel cycles in CANDU reactors in an evolutionary fashion.

The thorium fuel cycle in CANDU reactors is of strategic interest for several reasons:

- The amount of energy that can be extracted from mined uranium can be significantly extended using thorium fuel cycles; in the limit, the self-sufficient equilibrium thorium (SSET) cycle is independent of natural uranium and of any external supply of fissile material [2, 3].
- The once-through thorium (OTT) cycle in CANDU reactors provides an evolutionary approach to exploiting some of the energy potential of thorium without recycling [4]. The optimal OTT cycle is economical today, both in terms of money and in terms of uranium resources. This cycle creates a mine of valuable ^{233}U, safeguarded in the spent fuel, which may be recovered in the future.
- The abundance of thorium in the earth's crust is about 3 times that of uranium; thus the thorium fuel cycle ensures a long-term supply of nuclear fuel. Countries with abundant thorium reserves can enhance both the sustainability of nuclear power and their degree of energy independence.
- In thorium fuel, ^{233}U is produced in-reactor through neutron capture in ^{232}Th, and subsequent beta decay of ^{233}Th and ^{233}Pa. The concentration of fissile ^{233}U in the spent fuel is about 5 times higher than that of ^{239}Pu in spent natural uranium UO_2 fuel. This isotope of uranium is a very valuable fissile material because of the high number of neutrons produced per neutron absorbed (η) in the thermal neutron spectrum of CANDU reactors.
- The thermal conductivity of ThO_2 is about 50% higher than that of UO_2 over a large temperature range, and its melting temperature is 340ºC higher than that of UO_2. As a consequence, fuel-operating temperatures will be lower than those of UO_2, and all thermally activated processes—such as diffusion of fission gas from the fuel— will be decreased. Fission-gas release from the fuel should be lower than for UO_2 operating at similar ratings.

* 1998 meeting.

- ThO_2 is chemically very stable, and it does not oxidize—a benefit for normal operation, postulated accidents, and in waste management.
- ^{232}Th produces fewer minor actinides than ^{238}U does. The resultant lower radiotoxicity of spent thorium fuel is claimed by some to be a benefit in waste management. However, in an engineered geological disposal vault, the actinides contained in used fuel are not a significant contributor to radiological risk [5], and this benefit is judged to be small.

To ensure the viability of the CANDU reactor in the long term, AECL maintains an ongoing program on thorium fuel cycles. This program includes fuel-cycle studies, reactor physics measurements, development of reactor physics methods, fabrication of thorium fuels, fuel irradiation in the NRU research reactor at the Chalk River Laboratories (CRL), post-irradiation examination (PIE) of irradiated fuel, and assessments of fuel performance and waste management.

2. THORIUM FUEL CYCLES IN CANDU REACTORS

Historically, the main reasons for interest in thorium cycles have been, globally, to extend the energy obtainable from natural uranium and, locally, to provide a greater degree of energy self-reliance. That ^{233}U has the highest value of η for thermal neutron absorption of any of the commonly obtainable fissile nuclei is also significant because it introduces the possibility of a self-sustaining, or near-breeding, cycle in an already-developed and demonstrated thermal reactor. These possibilities are of particular interest to countries that have thorium reserves but lack uranium reserves because they hold out the possibility of energy independence using a single reactor type. More recently, some of the other noted advantages of ThO_2 as a fuel material have become prominent in evaluations of methods for dispositioning military plutonium.

Since thorium itself does not contain a fissile isotope, neutrons must be initially provided by adding a fissile material, either within or outside the ThO_2 itself. How this is done defines a variety of thorium fuel cycle options in CANDU reactors; there are, however, 2 broad classes: recycling options (those in which the ^{233}U is recycled into fresh fuel) and the once-through cycles (where the thorium-containing fuel passes once through the reactor and is either disposed of or stored for possible future recycling).

2.1. Thorium cycles involving recycling

Thorium cycles involving recycling are discussed by Veeder and Didsbury [6]. The authors used a special version of the WIMS-AECL [7] lattice code to analyze and compare the resource utilization of various CANDU reactor fuel cycles. These cycles included once-through natural uranium and slightly enriched uranium (SEU) cycles, as well as reprocessing cycles, based both on uranium and thorium. In the uranium cycles, plutonium from uranium fuel was recycled with either natural uranium or depleted uranium, and in the thorium cycles, the initial fissile material was either ^{235}U or plutonium. The results show that natural-uranium savings of up to 55% are possible by recycling plutonium in uranium cycles. For thorium cycles, the largest improvements in natural-uranium utilization are realized in replacement generating units that inherit the ^{233}U produced in the units initially using thorium. For such systems in equilibrium, savings in natural-uranium requirements of up to 90% compared with the cost of the once-through natural-uranium cycle are indicated.

The ultimate uranium-conserving fuel cycle would be the SSET cycle, in which no fissile topping material would be required, and which, in equilibrium, would require no natural uranium. The ^{233}U concentration in the recycled fresh fuel matches the ^{233}U concentration in the spent fuel. Further improvements in neutron economy would be required to achieve this.

2.2. Once-through thorium cycles

Fuel recycling is a demanding and potentially expensive technology to implement. For this reason OTT cycles have been investigated, for instance, by Milgram [4]. He studied, in a relatively simple way, CANDU-reactor-based cycles in which the fissile material required to support the buildup of ^{233}U in pure thorium bundles was provided in separate "driver" fuel bundles, containing in this case SEU. Such a system allows very different burnups and feed rates for the 2 fuel types (a higher burnup would be required in general for the thorium bundles than for the driver fuel), and it facilitates a search for combinations of feed rates, burnups, uranium enrichment, and neutron flux level that leads to cycles that are potentially economic compared with once-through uranium cycles, without taking any credit for the ^{233}U produced. (The term economic is here used to denote economy expressed in terms of either resource utilization or money.)

In general, the attempt was made to demonstrate the existence of cycles in which the utilization of natural uranium was as good or better than once-through SEU cycles and that, in addition, produced ^{233}U in the discharged thorium fuel. The study indicated that such cycles do indeed exist— although their implementation would pose many technical challenges, not the least of which would be finding refuelling schemes that produced acceptable power distributions in the reactor.

However, the flexibility provided by on-power refuelling in the CANDU reactor opens up many possibilities for fuel management in a once-through fuel cycle. Separate channels could be refuelled with ThO_2 bundles and with SEU. Alternatively, the same channel could be refuelled with both ThO_2 and SEU in any particular arrangement; during refuelling, the ThO_2 bundles could be reshuffled back into the core to ensure that they reach the required burnup.

If the ThO_2 bundles removed from the reactor were left to sit for a few weeks, or temporarily placed in low-flux regions of the core before being reinserted into the channel, the ^{233}Pa would decay to ^{233}U, thereby increasing the reactivity and energy derived from the fuel. Although fuel management in the OTT cycle will be challenging, the CANDU on-power refuelling system provides the means required to meet that challenge.

The benefit of the OTT cycle therefore is that it produces a mine of valuable ^{233}U in the spent fuel—at little or no extra cost—that is available for recovery at a time predicated by economic or resource considerations. Although the study by Milgram [4] focused on the use of SEU as the "driver" fuel, other fuels could also be considered, for example, "DUPIC" fuel [8] from reprocessed spent PWR fuel or even natural uranium [9].

2.3. Thorium as a carrier for plutonium annihilation

A special class of once-through fuel cycles that has recently received attention is one that uses ThO_2 as a matrix material for the annihilation of military plutonium [1]. Such Pu/ThO_2 cycles would achieve a very high efficiency in plutonium destruction. One bundle configuration considered employed ~2.6% weapons-derived plutonium in ThO_2 in a modified CANFLEX

[10] bundle, comprising a large central graphite displacer surrounded by 35 fuel elements in the 2 outer fuel rings. A burnup of 30 MW·d/kg heavy element (HE) was achieved, and >94% of the fissile plutonium was destroyed.

Good neutron economy is the key to high efficiency in plutonium destruction with ThO_2. Of course, ^{233}U is produced, through neutron capture in ^{232}Th, and partially burned in situ. This material is safeguarded in the spent fuel with all the proliferation-resistant barriers that spent fuel affords. The spent Pu-ThO_2 fuel would be simply stored until a decision was taken to recycle the contained ^{233}U and thorium, based on economic and resource considerations, and the availability of a proliferation-resistant recycling technology for ThO_2. Of course, high-enriched uranium (HEU) could also be used as the fissile component in ThO_2, as an option for dispositioning military-derived HEU.

2.4. Proliferation-resistant thorium fuel cycles

As has been discussed, to obtain the full energy potential from the thorium cycle in the longer term requires recycling of the ^{233}U. This approach embodies the environmental 3Rs of *reduce, reuse and recycle.* However, special consideration must be given to non-proliferation issues. Conventional reprocessing involves the production of separated fissile material, with the associated proliferation risks that are addressed in the framework of international safeguards. One degree of proliferation resistance is provided in thorium fuel cycles by the presence of ^{232}U in the spent fuel. The ^{232}U renders the ^{233}U less attractive for diversion because of its copious emission of alpha particles and the penetrating 2.6 MeV gamma ray associated with ^{208}Tl in the ^{232}U decay chain.

The absence of a commercially established facility for recycling thorium fuel opens up the possibility of incorporating the highest degree of proliferation resistance in the design of a new fuel recycling facility from the start. It is possible to conceive of recycling options for thorium that have a higher degree of proliferation resistance than conventional reprocessing does. Moreover, the high neutron economy of CANDU reactors facilitates such options because the fissile requirements are small compared with those of a light-water reactor (LWR). In recycling the used ThO_2 fuel, only the high-neutron-absorbing rare-earth fission products need to be extracted, and the ^{233}U can be recycled, along with thorium and other fission products and actinides. The intense radiation fields, and the absence of separated fissile material would be important proliferation-resistant features. A simplified flow sheet may have economic benefits. Remote fabrication technology would be required for the recycled fuel, providing yet another proliferation barrier. The simple, small CANDU fuel bundle would facilitate remote fabrication. Except for the SSET cycle, new fissile material would be required to "top up" the fissile content of the recycled material, to maintain the desired burnup. Using "denatured" uranium for the fuel "topping", in which the concentration of ($^{233}U + {}^{235}U$) in ^{238}U is less than a critical value [typically, $^{238}U \geq (6\times{}^{233}U + 4\times{}^{235}U)$], provides another degree of proliferation resistance, through isotopic dilution of the fissile component. This feature of course entails a penalty in uranium utilization compared with using HEU as the topping material. However, this penalty is not large compared to the significant improvement in uranium utilization over the natural-uranium cycle [6].

2.5. An evolutionary approach to thorium fuel cycles in CANDU

One possible strategy that would enable a country to move towards a CANDU thorium cycle would be the following. The reactor would be initially fuelled with natural-uranium fuel, facilitating localization of the fuel-fabrication technology. SEU could then be introduced, lowering fuelling costs even further, improving uranium utilization, and reducing the quantity of spent fuel. (Alternatively, a country with a dual CANDU–PWR reactor mix could utilize the DUPIC fuel in the CANDU reactor). From there, the OTT cycle would be introduced. The fuel-cycle parameters (such as uranium–thorium feed rates, uranium enrichment, burnups) would be so chosen that uranium utilization and fuel-cycle economics are comparable to those of SEU.

The spent fuel would be simply stored until a decision was taken to recycle the contained ^{233}U and thorium, predicated on economic and resource considerations and the availability of proliferation-resistant recycling technology. In the recycling stage, some fission products would be removed from the spent thorium fuel (particularly the neutron-absorbing rare-earth elements), and the thorium, ^{233}U, and remaining fission products would be blended with "denatured" UO_2 to form fresh fuel. After irradiation, that fuel would be stored and eventually recycled, again topped with denatured UO_2 to maintain an economic burnup.

3. FUEL-MANAGEMENT SIMULATIONS

3.1. Introduction

Two methods can be used to introduce the OTT fuel cycle into existing CANDU reactors. The first is a "mixed-core" approach where a large number of driver channels containing enriched-uranium fuel are used to support a relatively small number of channels dedicated to thorium irradiation. Because of the disparity in reactivity and power output between driver channels and thorium channels, very sophisticated fuel-management schemes will be required to shape the channel and bundle power distributions in the mixed core, to achieve the nominal reactor power output. This approach is theoretically feasible, but its practicality has not been investigated in detail.

An alternative approach is to fuel the whole core with mixed-fuel bundles, which contain both thorium and enriched-uranium fuel elements in the same bundle. This "mixed-fuel bundle" approach is a practical means of utilizing thorium in existing CANDU reactors, while keeping the fuel and the reactor operating within the current safety and operating envelopes established for the natural-uranium fuel cycle.

3.2. Options for burning thorium in CANDU reactors

Two options have been examined for burning thorium fuel in an existing CANDU 6 reactor. In Option 1, only one fuel type was used throughout the entire core, and the reactor's adjuster rods were removed. The reference fuel design is a CANFLEX fuel bundle with 1.8 wt % slightly enriched UO_2 fuel in the outer 35 elements and natural ThO_2 fuel in the inner 8 elements. The initial fissile content was chosen to maximize the burnup of the thorium fuel elements, without exceeding the current limits on maximum channel and bundle power.

The second option illustrates the flexibility of existing CANDU reactors to accommodate both thorium fuel and adjuster rods. In Option 2, each of the 3 regions shown in Figure 1 contains a different type of thorium fuel bundle. The fuel in the 196 outer-region channels is the same as that used in Option 1. The fuel in the 124 inner-region channels is identical to that in the outer-region channels, except that the central ThO_2 element contains 6.0 wt % of gadolinium to shape the flux distribution. The gadolinium-doped bundles are used only in the inner region, which is under the influence of the adjuster rods.

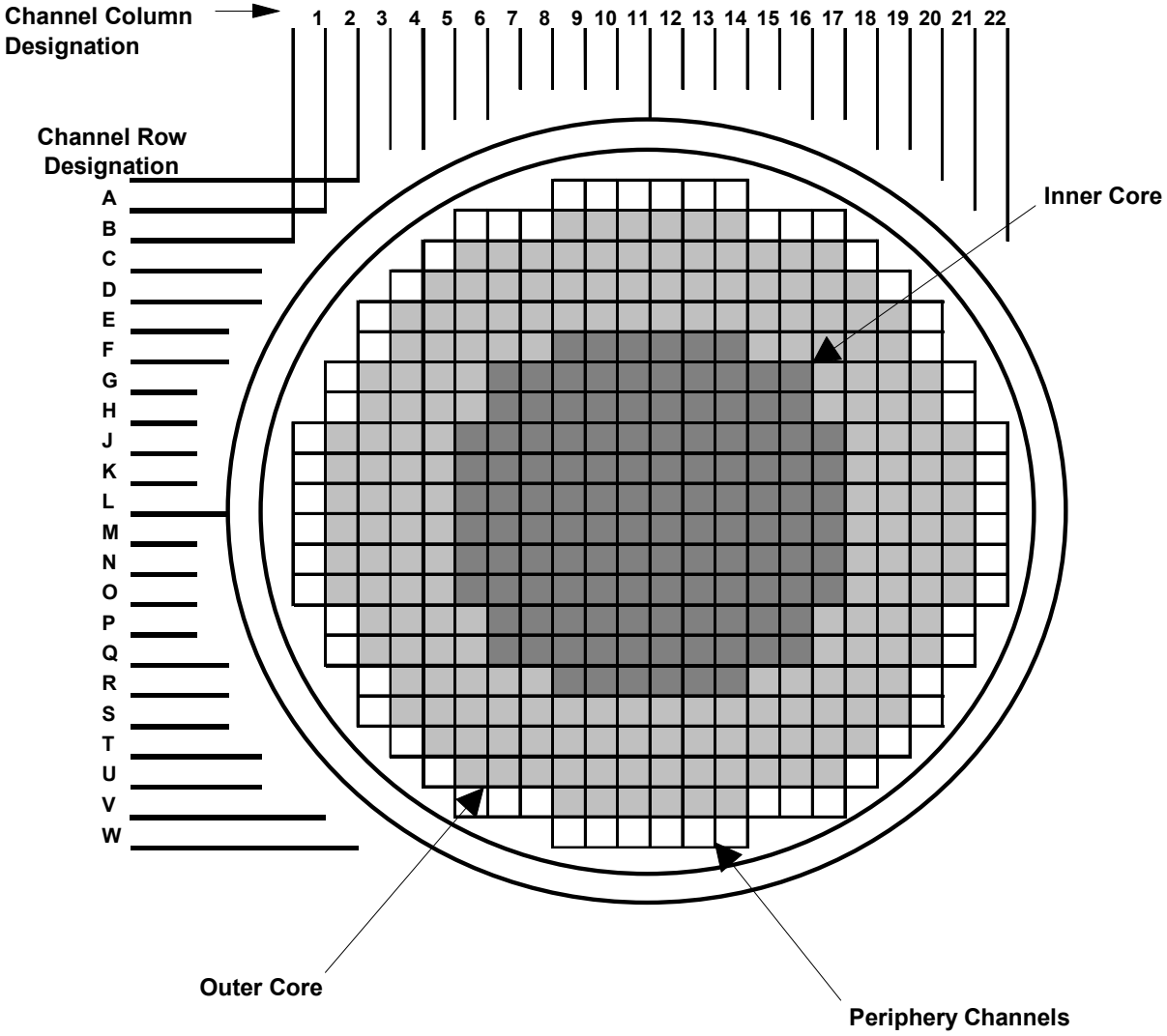

Figure 1. Reactor Core Model of a CANDU 6.

The 60 peripheral channels in Option 2 contain thorium bundles designed to achieve burnups of over 50 MW·d/kg HE. These high-burnup thorium bundles use natural ThO_2 in all 43 fuel elements. However, the initial fissile content in the outer 35 elements is increased from 0 wt % to 1.7 wt % using 20 wt % enriched uranium. These high-burnup thorium bundles are strategically located at the edge of the core to utilize a large percentage of the leakage neutrons to produce power. This arrangement significantly increases the amount of thorium fuel in the core and improves the overall fuel efficiency of the thorium-burning reactor.

3.3. Lattice properties of mixed-thorium fuel

Three types of fuel were used for these two studies: the mixed SEU and ThO_2 bundles, gadolinium-doped mixed bundles and high-burnup ThO_2 bundles. The variation of lattice k-infinity, and fissile content as a function of bundle average burnup are shown in Figures 2 and 3 for lattices of each of these 3 fuel types. Although natural-UO_2 and natural-ThO_2 fuel bundles were not used in this study, their physics properties are also shown in these figures for comparison purposes.

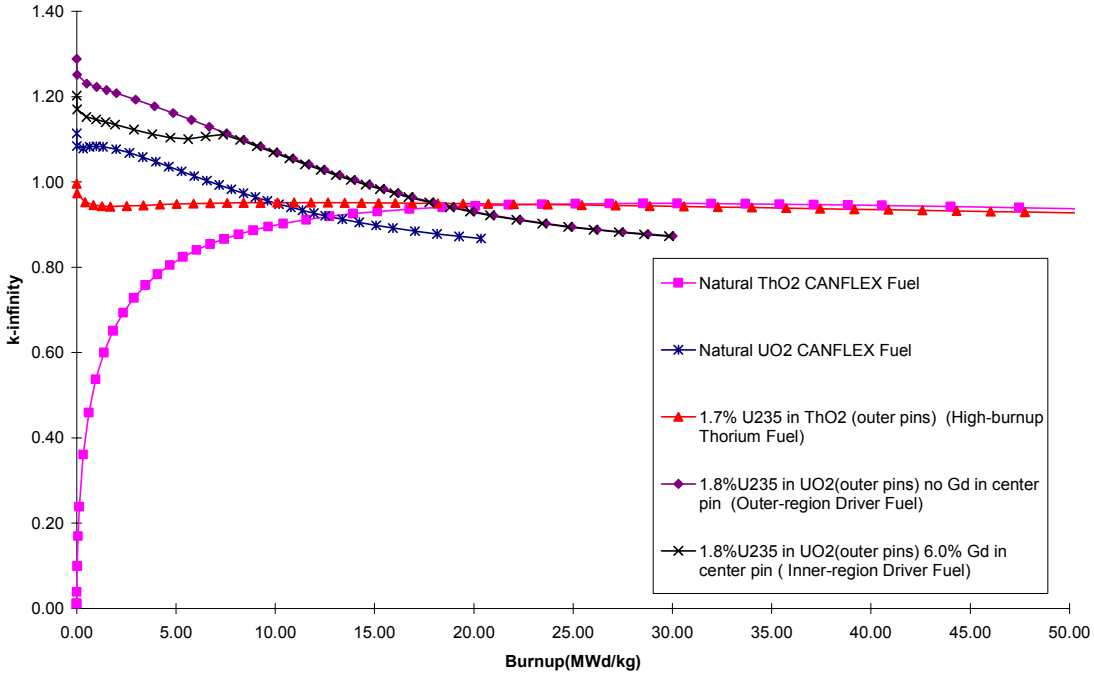

Figure 2. Lattice k-infinity vs. fuel burnup.

The initial fissile content of the high-burnup thorium bundles has been carefully chosen so that the depletion rate of the fissile material is almost the same as the conversion rate of the fertile ^{232}Th into fissile ^{233}U. Consequently, the reactivity and the fissile content of the high-burnup thorium bundles are almost constant throughout the entire lifetime of the bundles.

The main purpose of the gadolinium is to shape the axial flux distributions so that the resulting bundle flux and power distributions are similar to those in the thorium-burning reactor without adjuster rods. The gadolinium effectively eliminates the bundle power distortion caused by the adjuster rods. As expected, the effect of gadolinium on lattice reactivity is evident only during the initial stage of the fuel's lifetime. The fast burnout rate of gadolinium suppresses the reactivity of the fresh bundle without causing significant burnup penalty over the lifetime of the fuel. This effect also reduces the channel and bundle power ripples caused by refuelling. The presence of a neutronic poison, gadolinium, in the central element also reduces coolant-void reactivity [11]. This approach results in a significant reduction in the core-averaged coolant-void reactivity.

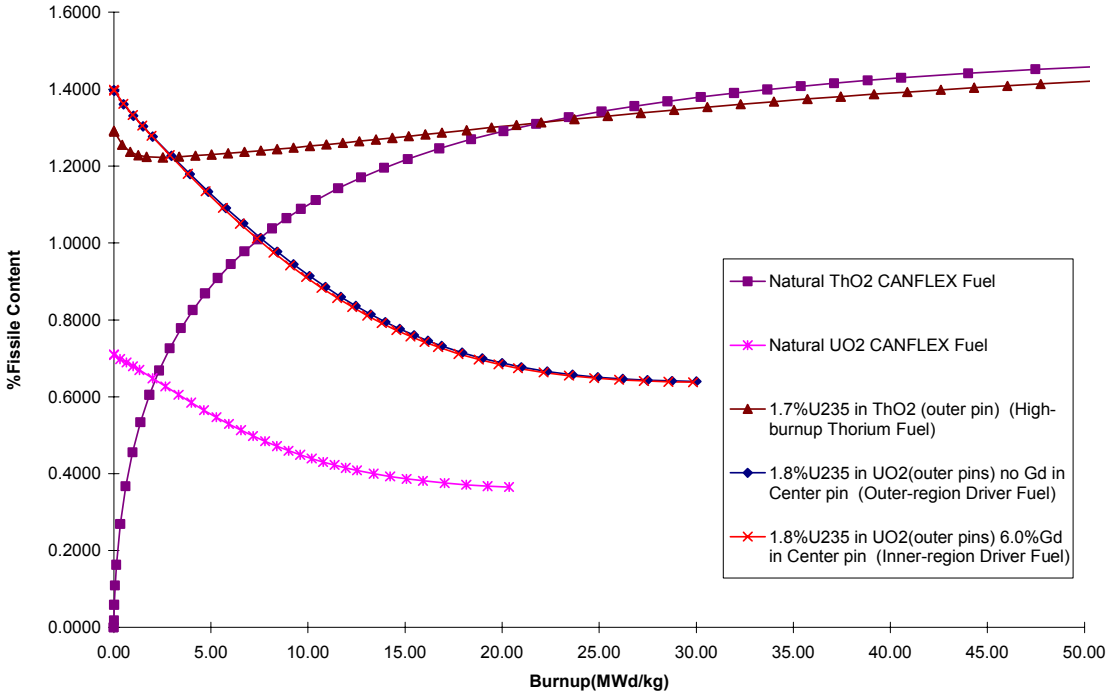

Figure 3. Fissile content vs. fuel burnup.

3.4. Characteristics of thorium-burning CANDU reactors

The RFSP code [12] was used to perform time-average core calculations for Options 1 and 2 using a uniform 2-bundle-shift fuelling scheme. Instantaneous core calculations were conducted using randomly generated age patterns. The channel power distributions for the 2 thorium-burning reactors are very similar to those of a typical natural-uranium-fuelled CANDU reactor. The axial power distributions in the thorium-burning reactors are flatter than those in a natural-uranium CANDU and are skewed towards higher power at the coolant-inlet end. This skewed axial power profile should improve the thermalhydraulic performance.

The fuelling rates and the maximum channel and bundle powers for both options are well within the limits established for current CANDU reactors using natural-uranium fuel. The coolant-void reactivity is also significantly reduced from that of a natural-uranium reactor, under comparable conditions. Option 1 gives 21% better uranium utilization than does a natural-uranium-fuelled CANDU reactor. About half of the improved fuel efficiency is due to the removal of the adjuster rods. The other half can be attributed to the energy produced in the thorium fuel. Option 2, which uses the existing adjuster rods, gives 14% better uranium utilization than does a natural-uranium-fuelled CANDU reactor, with the additional advantage of a significantly lower coolant-void reactivity.

3.5. Conclusions

The current study represents only a first look at practical fuel-management strategies for the OTT fuel cycle. Two options for implementing the OTT fuel cycle in existing CANDU reactors were identified. For both options, the uranium utilization is better than that of the natural-uranium fuel cycle. The reactor and the fuel perform within existing envelopes, without requiring major modification to the current reactor design. Coolant-void reactivity is significantly lower than that of a natural-uranium reactor under comparable conditions.

4. REACTOR PHYSICS ASPECTS OF THORIUM FUEL CYCLES

4.1. General considerations

The reactor physics calculational and nuclear data requirements for thorium cycles are similar to those that are known for the more familiar uranium–plutonium cycles, except that different nuclides play the important roles. Thus the fertile material is ^{232}Th instead of ^{238}U, but its neutron capture cross section, particularly in the resonance region, must be equally well understood. The fissile nuclide that is bred is primarily ^{233}U (rather than ^{239}Pu), and it is preceded by 2 precursors, ^{233}Th and ^{233}Pa, (rather than ^{239}U and ^{239}Np). The capture and fission cross sections of all these nuclides are important to varying degrees, but one interesting point is that the neutron capture cross section of ^{233}Pa is of more significance than that of ^{239}Np because of the relatively long half-life (27 d) of ^{233}Pa, which allows neutron capture to compete effectively with beta decay at the neutron flux levels that are to be expected in a thorium-fuelled reactor. This process leads to flux-dependent production rates of ^{233}U, which in turn can lead to interesting reactivity transients as flux levels vary. Because of this effect, an extra requirement is imposed on the codes used for reactor calculations. This requirement is the consideration that the composition of the fuel is no longer a function of its burnup alone but also of the flux it was exposed to throughout its irradiation [13]. AECL is producing combinations of its existing codes that will allow this type of calculation to be performed. In addition there is a need for a program to evaluate the appropriate nuclear data for thorium cycles, and, if necessary, to perform additional cross-sectional measurements, for validating the codes for calculations of thorium-containing reactor lattices.

4.2. Validation measurements for CANDU lattices

In the late 1980s and early 1990s AECL completed 2 sets of measurements in the ZED-2 critical facility on 36-element bundles of CANDU-type fuel containing thorium as the fertile material. The first set of measurements was on fuel consisting of a uniform mixture of ThO_2 and PuO_2 in which Pu was about 2 wt % of the mixture. The second was on similar-geometry oxide fuel in which the fissile material was ^{233}U at 1.23 wt % of the ThO_2-$^{233}UO_2$ mixture.

Insufficient fuel was available to create uniform critical cores of the fuels, so the substitution technique was used to measure lattice buckling. In this technique, a uniform critical reference core is first created, and its critical size is measured. For these measurements the reference core consisted typically of 55 rods, each containing five 28-element natural-UO_2 CANDU bundles, arranged in a hexagonal lattice. Into the central region of this lattice 1, 3, 5, and finally 7 rods, each containing 5 bundles of the thorium-containing fuel are successively substituted, the critical size of the reactor being measured for each geometry. An analysis involving a calculational model of the reactor is used to derive the buckling of the substituted fuel lattice for each substitution geometry, and an extrapolation gives the best estimate of the buckling of a critical core of the substituted fuel. Measurements were completed for both fuel types at 2 lattice pitches, 31 cm and 24.5 cm, with 3 different coolants in the fuel channels: heavy water, void (air), and light water. In addition, the change in critical size of the reactor was measured as the water coolant and fuel in the 7 substituted rods were heated to 300ºC.

In addition to the preceding measurements, which can be viewed as integral measurements of the neutron balance in thorium-fuel lattices, detailed reaction rate distribution measurements were made. These measurements were performed in and around the fuel bundle at the centre of the substituted region by irradiating and then counting pieces of foil material containing the

isotopes of interest. Foils were placed between the pellets of representative elements in each ring of the bundle, as well as throughout the remainder of the cell. The materials that were irradiated and counted included thorium metal, ^{233}U-Al alloy, ^{235}U-Al alloy, ^{239}Pu-Al alloy, all of which are materials actually occurring in the fuel. In addition, various other neutron-indicating materials were irradiated (^{63}Cu, ^{55}Mn, ^{197}Au, ^{176}Lu, and ^{115}In). These materials are used as neutron spectrum indicators; for instance, ^{63}Cu and ^{54}Mn have essentially 1/v cross sections, whereas ^{197}Au and ^{115}In have a large capture resonance in the epi-thermal region, and ^{176}Lu has a capture resonance in the thermal region.

These measurements are all designed to provide data for validating calculations for thorium-fuelled CANDU lattices performed with lattice–cell codes like WIMS-AECL. The analysis of the substitution measurements to yield lattice buckling has not been completed although a program to do so is currently in place. However the reaction rate data for the ^{233}U-Th fuel were compared with WIMS-AECL calculations [14, 15].

5. FABRICATION AND IRRADIATION EXPERIENCE WITH THORIA FUEL

Many experiments were performed to assess the irradiation performance of thorium fuels. In addition to research reactor irradiations, a number of power reactors were fuelled with thoria, including Elk River [16], Indian Point [17, 18] and Shippingport [16]. This section examines methods that have been used to fabricate thoria fuels and their in-reactor performance.

5.1. Materials

Thorium in the form of ThO_2 is derived from thorium oxalate. In this process oxalic acid ($H_2C_2O_4$) is added to a thorium nitrate ($Th(NO_3)_4$) solution to precipitate thorium oxalate ($Th(C_2O_4)_2$). The product typically contains up to 6 waters of hydration. The final step is calcination of the thorium oxalate to form ThO_2. Most experience indicates that such powders are not reactive or uniform enough for a feed powder in a ceramic process [16]. As a result, it is usual to include a milling stage at some point in the fabrication process.

In addition, past irradiations have often required that a fissile component be added to the (fertile) thoria matrix. Ideally, the fissile component should be as uniformly distributed throughout the fertile matrix as possible. A number of techniques have been investigated to generate the required distribution.

5.2. Fabrication processes

A number of routes have been used to fabricate $(Th/U)O_2$ and $(Th/Pu)O_2$ fuel, including dry-powder blending/milling, wet-powder blending/milling and co-precipitation. These processes are described in Sections 5.2.1 through 5.2.3.

5.2.1. Dry-powder blending

Dry blending can be done by a number of methods, including mixing in a V-blender, dry-ball milling, vibratory milling and jet milling. These dry methods tend to be dusty and thus have associated health concerns. There are also problems with uniformity on a micron scale, and intensive mixing is required. The resulting pellets are generally of good quality.

Fuel for the Shippingport light-water breeder reactor (LWBR) was fabricated by dry-powder blending [19, 20]. Early attempts at pellet fabrication yielded low densities (90% to 94% theoretical density, TD) and improved comminution (jet milling, or micronizing) was used in conjunction with higher sintering temperatures, to increase the density to 96% TD.

AECL also investigated dry-blending to fabricate thorium fuels. In this case, previously ground thoria powders were mixed with highly enriched uranium powders and were dry-ball-milled before pressing and sintering. Between 1981 and 1983, six bundles of $(Th/Pu)O_2$ fuel were fabricated in the Recycle Fuel Fabrication Laboratory (RFFL) at the CRL. In 1982, one bundle of ThO_2 fuel was fabricated, and between 1982 and 1985 a number of bundles and elements of $(Th/^{233}U)O_2$ and $(Th/Pu)O_2$ were fabricated. Densities achieved were typically 96% TD.

5.2.2. Wet-powder blending

As is the case with dry blending, there are a number of wet-mixing methods, including wet-ball milling and attrition milling. The advantage of attrition milling is a fast throughput, compared with ball milling (1 h of attrition milling is comparable with 1 d of ball milling). Wet processes have the advantage of not being dusty.

The fuel for the Indian Point reactor was wet-blended [18]. It was eventually sintered to a density of 93% TD (significantly lower than other thoria fuels described here).

The fuel pellets used as reference fuels in AECL's WR 1 irradiations were manufactured using a process of attrition milling, pan drying (forming cakes), granulation, pressing and sintering. This fuel tended to have a granular microstructure that resulted in fission-gas releases higher than expected for thoria (although still typical of releases found in UO_2). After irradiation, this granular microstructure was still evident.

5.2.3. Co-precipitation

There are a number of methods of co-precipitating UO_2 and ThO_2 [16, 21]. AECL experience has focused on the addition of ammonia to nitrate solutions. In this process, uranium and thorium are dissolved into a nitrate solution to form $UO_2(NO_3)_2$ and $Th(NO_3)_4$. Ammonia is added to the solution to precipitate $(NH_4)U_2O_7$ (ammonium diuranate - ADU) and $Th(OH)_4$ (thorium hydroxide). The precipitate is calcined to form blended UO_2 and ThO_2 powder, which is subsequently processed into fuel pellets. The microstructure and quality of these pellets was generally very good.

5.3. Irradiation experience

As reported by 16], the Elk River reactor in Minnesota used a mixed $(Th/U)O_2$ fuel containing 5.27 and 4.36 mole percent uranium enriched to 92% ^{235}U. To enhance sintering, 0.4 wt % CaO or TiO_2 was added to the ThO_2 component.

This step resulted in fuel pellets having densities of 94% TD. Homogeneity of the fuel was poor. The fuel was irradiated to approximately 8 MW·d/kg HE. Neutron radiography of the spent fuel indicated little cracking, and fission-gas release (Kr and Xe) was approximately 0.2%.

The Indian Point Reactor Number 1 was a 270 MW(e) PWR [16, 17]. In one core load the fuel was $(Th_{1-X}/U_X)O_2$ fuel pellets, having values of x between 0 and 0.09. The uranium was enriched to 93 mole percent ^{235}U. Only one core load was used because of the high cost of the enriched uranium. The maximum burnup of the fuel was 32 MW·d/kg HE. The measured fission-gas release was 1 to 2%.

The last core charge to the Shippingport PWR was thoria, and the reactor was operated as a LWBR [22]. The reactor was shut down in 1982 October after 1200 effective full-power days of operation. The fuel for the reactor was ThO_2 and $(Th/U)O_2$ with the uranium in solid solution in the thoria. Pellet densities were typically 96% of TD. Maximum fuel burnup was 60 MW·d/kg HE. To accommodate this high burnup, the fuel design included 45° chamfer angles [23].

The fuel development program, in support of the Shippingport reactor, was performed by Westinghouse Electric Corporation's Bettis Atomic Power Laboratory (Bettis), and irradiations were performed in a number of reactors. The fission-gas release was typically between 0.1% and 2% [24, 25].

AECL conducted a program to develop thoria fuels for use in CANDU reactors, from the mid-1970s to the mid-1980s. Test irradiations were performed in the WR 1 reactor at the Whiteshell Laboratories of AECL. Fission-gas release from “standard pellets” in these experiments was somewhat higher than the Bettis results, typically between 1 and 15%.

In addition to the WR 1 irradiations, AECL also irradiated four 19-element bundles in the NPD reactor between 1977 and 1987. The fuel in two of these bundles contained 2.6 wt % UO_2 (enriched to 93 wt % ^{235}U), and the other two contained 1.45 wt % UO_2. Fission-gas release in these bundles was typically 0.4% to 0.5%.

Overall, thoria fuels have been found to exhibit in-reactor performance superior to the in-reactor performance of UO_2 operating under similar conditions. This improved performance is a result of material properties of thoria (higher thermal conductivity, greater chemical inertness), but it is important that the microstructure of the fuel be of a high enough quality (free of residual granules) to benefit from these features.

6. WASTE-MANAGEMENT ASPECTS OF THORIA FUELS

6.1. Introduction

Thoria-based fuels are also appealing from a waste-management perspective because ThO_2 is chemically stable and almost insoluble in groundwater. In this section, we explore waste-management aspects of thoria-based fuels and compare some of the key chemical and physical properties of UO_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 [26]. This section is based on a previous, more detailed comparison of factors affecting the disposal of $(Th/Pu)O_2$ and UO_2 fuels [27].

6.2. Chemistry of thoria

6.2.1. Redox chemistry

By far the most important chemical difference between ThO_2 and UO_2 is 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, 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, so long as the uranium or other fissile component is either present in solid solution or is effectively encapsulated by thoria. 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 leachability 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 to 175°C, because at higher temperatures oxidation of UO_2 to U_3O_8 in defected elements can cause powdering of the fuel matrix and splitting of the fuel cladding ([28], and references therein). Matrix oxidation is not an issue with thoria-based fuels. Moreover, the thoria structure can easily accommodate oxidation of minor solid-solution components such as U and Pu. Thus fuel oxidation is unlikely to be a concern during dry storage of thoria-based fuels, and hence the maximum storage temperature would be limited by some other factor, probably cladding degradation [29].

6.2.2. Aqueous chemistry

The solubility of crystalline thoria in aqueous solution 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 [30]. 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. No credible aqueous or geochemical process has been identified that would greatly accelerate ThO_2 fuel-matrix dissolution under disposal conditions [27].

6.2.3. Compatibility of actinides with thoria

Thoria crystallizes with the fluorite structure, as do all other actinide dioxides. 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. 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

6.3. 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-to-sheath gap of the fuel. 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 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.

6.3.1. 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 fuel-to-sheath 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 [31]. 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 is; it also has a higher melting point and slower cation diffusion. Therefore, for a given power rating and fuel geometry, it would be expected to run cooler and undergo less grain growth.

Fission-gas release rates are expected to be somewhat smaller for thoria-based fuels than for UO_2 fuels that have comparable geometry, microstructure and power history. This conclusion is based on the lower diffusion rate for xenon in ThO_2 than UO_2 [32, 33] and the smaller burst release in ThO_2 [34].

The expected low fission-gas release rates from thoria-based fuels are supported by in-pile experiments on ThO_2 and $(Th/U)O_2$ fuel assemblies. Goldberg [24, 25] 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 significantly lower than for UO_2 under comparable operating conditions.

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 [35, 36], and thus we expect the release of these fission products to be lower for a thoria-based fuel than for UO_2. Jones et al. [37] reported low fission-gas releases for $(Th/U)O_2$ fuels, and they also noted that fission-product release from defected thoria elements was 1 to 2 orders of magnitude lower than for UO_2. Experimental data obtained by Matzke [38] supports this notion; he found that the release of Br, Cs and Rb from thoria was generally slower than from UO_2.

6.3.2. Diffusion properties of thoria

Diffusion of fission products in UO_2 and ThO_2 remains poorly understood, but generally appears to involve uranium or thorium 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 in UO_2 - roughly paralleling the difference in cation lattice diffusion [31, 38, 39, 40, 41].

Fission-product migration in-reactor involves further complexity; indeed, Matzke (1980) has suggested that 5 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 for UO_2 fuels.

6.4. Reactor operation

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, ThO_2 fuels should exhibit little or no grain growth. CANDU reactors have flexibility in fuel management and fuel design that can ensure that ThO_2 fuels would operate at similar or lower linear power ratings, compared with the corresponding ratings of current UO_2 fuel.

For example, the 43-element CANFLEX bundle reduces peak ratings by about 20% compared with the peak power ratings of the 37-element bundle [10]. Hence there is a real possibility that ThO_2 fuel could be operated in CANDU reactors with minimal fission-gas release.

6.5. Conclusions

The high degree of chemical stability and the low solubility of thoria make irradiated thoria-based fuels attractive as waste forms for direct geological disposal. Moreover, 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.

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 PIE 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.

7. SUMMARY

The high neutron economy of the CANDU reactor, its ability to be refuelled while operating at full power, its fuel channel design, and its simple fuel bundle provide an evolutionary path for allowing full exploitation of the energy potential of thorium fuel cycles in existing reactors.

AECL has done considerable work on many aspects of thorium fuel cycles, including fuel-cycle analysis, reactor physics measurements and analysis, fuel fabrication, irradiation and PIE studies, and waste-management studies.

Use of the thorium fuel cycle in CANDU reactors ensures long-term supplies of nuclear fuel, using a proven, reliable reactor technology. Those same CANDU features that provide fuel-cycle flexibility also make possible many thorium fuel-cycle options.

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STUDIES ON THORIUM FUEL CYCLE IN JAPAN*

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Abstract. The official policy of Japanese government concerning nuclear energy is to pursue ^{238}U-Pu fuel cycle. On one hand, a large number of university professors, together with some of the research staffs of Japan Atomic Energy Research Institute (JAERI), has performed several research projects on the thorium fuel cycle as the future nuclear energy with the support of Grant-in-Aid for Scientific Research by the Ministry of Education, Science and Culture. These projects ended in 1993,but several activities have been continued after that.

1. INTRODUCTION

The work in Japan until the end of 1995 was reported in [1], therefore, this report is concentrated on the activities since then mainly concerning neutronics.

2. NUCLEAR DATA

To study the fission process, the simultaneous measurements of masses of fission fragments, kinetic energies and the number of prompt fission neutrons (υ(m)) for each fission fragments of ^{233}U have been performed by Nishino et al. [2] using the super mirror neutron guide tube system of Kyoto University Research Reactor Institute (KURRI). Figure 1 shows the experimental set up. This method measures the energy of a fragment FF1 at the same time measuring the emission angle θ and the energy of the other fission fragment FF2. The energies were measured by time of flight method. The fission neutrons were detected by a liquid scintillator (NE213). The ^{233}U sample was electroplated on a Ni foil (90 μg/cm^2). The sample thickness was 140 μg/cm^2. Figure 2(a) shows the average neutron multiplicity from each fission fragment as a function of the fragment mass. The neutron multiplicity for light and heavy fragment groups is 1.49 and 1.01, respectively, which means about 60% of the fission neutrons have their origin in the light fragments. This value is very close to that of 58% for ^{235}U(n_{th},f) [3]. In the same figure, the corresponding data by Milton and Apalin et al. [4] are shown in comparison. Figure 2(b) shows the average total neutron multiplicity, which is the sum of the neutrons from both fragments.

The measurement of fission cross section of ^{231}Pa which is one of the nucleus to produce ^{232}U whose existence is one of the problem in the reprocessing, was carried out by K.Kobayashi [5] et al. using a lead spectrometer coupled to the 46MV electron LINAC at KURRI. The ^{231}Pa sample was electroplated on an Al plate of 0.5mm thick. The diameter of the sample was 20mm. As the reference sample highly enriched ^{235}U(99.91%) was prepared by the same method as the ^{231}Pa sample. Figure 3 shows the slowing down spectrometer and Figure 4 shows the cross sections deduced using the ^{235}U fission cross section of ENDF/B6. The evaluation of JENDL3.2 is mostly within the experimental error but a little higher between 0.3 and 0.7 eV than the experimental value.

* 1998 meeting.

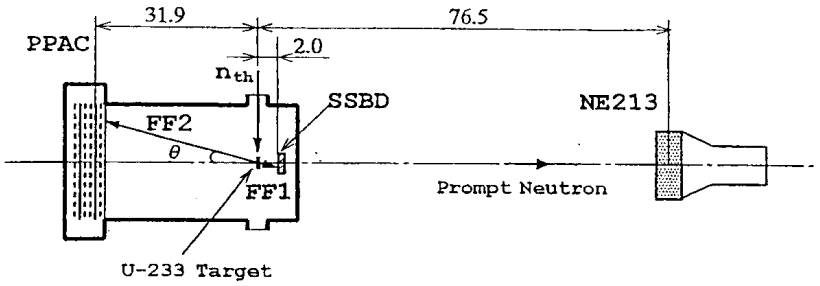


Fig.1 Experimental arrangement for simultaneous measurement of neutrons and fission fragments for $^{233}U(n_{th},f)$

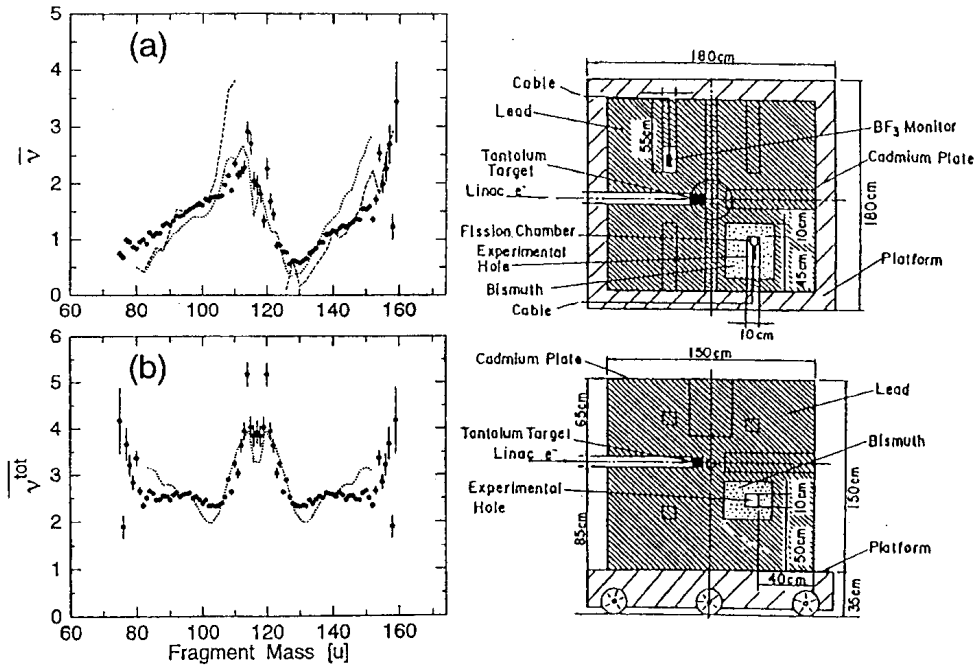


Fig.2 (a) Average neutron multiplicity versus fragment mass.

(b) Average total neutron multiplicity, ν_{tot} as a function of fragment mass.

Fig.3 Cross sectional view of Kyoto univers Leads Slowing Down Spectro-meter used j ^{231}Pa fission cross section measurement.

3. CRITICAL EXPERIMENT

Critical Experiments of polyethylene moderated core fuelled with thorium and highly enriched uranium (Th-EU) have been carried out for several years at KURRI. Table I shows the names of constructed assemblies and H/^{235}U and ^{232}Th/^{235}U atomic number ratios. Since 1996, B3/8"P30EU-Th-EU and B4/8"P24EU-Th-EU-EU [6] assemblies were constructed. In these cores, the criticality, the flux distributions and the reactivity distributions of ^{232}Th and ^{233}U samples were measured (Figure 5) and analyzed with SRAC-CITATION code system which is the standard design code for a thermal reactor in Japan, though the results of the analysis is not available at present. The cadmium (Cd) ratios of 8 activation foils shown in Table II were measured at KUCA B4/8"P24EU-Th-EU core and the results were compared with those of similar natural uranium core (B3/8"P36EU-NU-EU) [7]. The analysis was carried out by SRAC-CITATION system with JENDL3.2 cross sections. The results of C/E are shown in Table III. It was found that the behavior of C/E is similar for both cores. That is C/E for Cd ratio <2 are~1.06, for those 2-8 are ~0.97 and for those >8 are ~1.07, respectively. It means that the calculated neutron spectrum for both Th and NU loaded cores should be the same.

4. NUCLEAR DESIGN

4.1. Once through burning process of plutonium

At JAERI, once-through burning process has been studied for the disposition of excess plutonium (Pu) [8]. A new stable fuel material of multi-phases is fabricated based on the conventional MOX fuel technique. After irradiation in LWR, the spent fuel would be geologically stable and becomes high level waste (HLW) without reprocessing. As one of the 2 candidate systems, PuO_2-ThO_2-Al_2O_3-MgO system is proposed. The experimental study has been made to examine phase relations of the fuel materials and the distribution of fission products. Pu transmutation characteristics were estimated by a 2-dimensional core calculation. The results are shown in Figure6. It was found that as much as 83% of total Pu and 98% of ^{239}Pu are transmuted after about 1400days of burn up. Therefore, the quality of Pu becomes very poor in the spent fuels. A total of 0.87 tonne of Pu would be denatured every year under the assumption of a 1GW(e) PWR operating at 80% of availability. If we use the PuO_2-ZrO_2(Y,Gd)-Al_2O_3-MgO fuel system, the Doppler coefficient is very small especially at the beginning of the fuel cycle as shown in Table IV. However, if we modify the fuel system to PuO_2-(Zr-Th)O_2-Al_2O_3-MgO, the Doppler reactivity is markedly improved and when half of the ZrO_2 is replaced by ThO_2 , it becomes 4 times as large as that for the case without ThO_2 as shown in Figure7.

4.2 Molten Salt Reactor

Several studies on the use of molten salt reactor (MSR) with thorium have been carried out. For instance, Osaka et al. [9], studied the two-step TRU transmutation using MSRs suitable for the TRU transmutation. They use molten salt whose initial composition is ^{7}LiF-BeF_2-ThF_4-$^{233}UF_4$-(TRU)F_X=71.64-16.00-11.85-0.31-0.15mol% in the first step and =71.80-16.00-12.0-0.2-0.0mol% for the second step. The schematic view of the reactor is shown in Figure 8, which is a 2250MW(th) single fluid single region core. The specifications of Step1 and Step2 cores are shown in Table V. The fuel volume ratio (FVR) of Step1 is 9% and that of Step2 is 9% for the first 12 years and is changed to effectively 11.7% by the use of hollow graphite. The composition of minor actinide (MA) in Step1 is shown in Table VI.

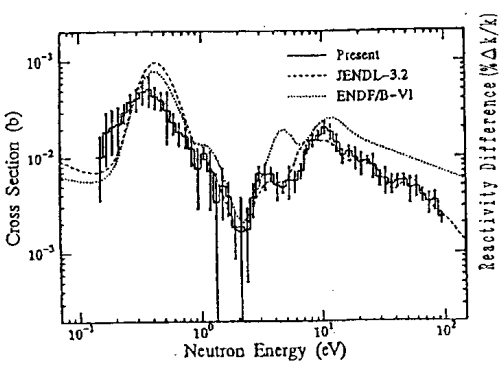


Fig.4 Result of ^{231}Pa fission cross section measurement.

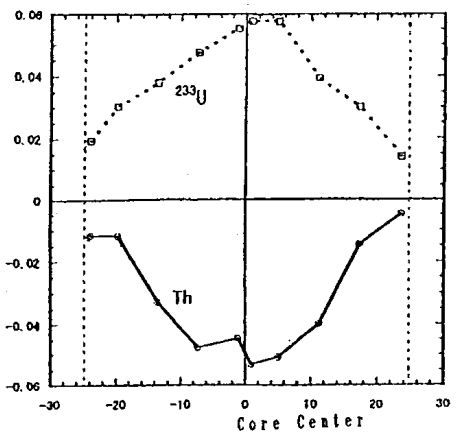


Fig.5 Measured reactivity differences of Th and ^{233}U samples at KUCA B4/8" P24EU-Th-EU-EU core

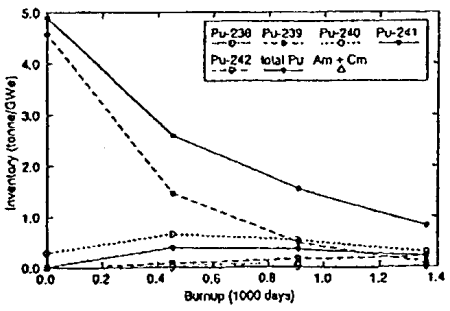


Fig.6 Burnup dependence of Pu com-position in PuO_2-Zr(Y,Gd)-Al_2O_3 fuel PWR in JAERI's once-through burning process.,

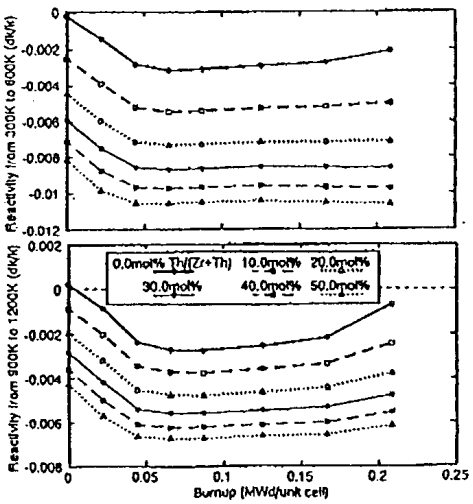


Fig.7 Doppler reactivity of PuO_2-(Zr-Th)O_2-Al_2O_3 fuel cell of PWR in JAERI's once-through burning process.

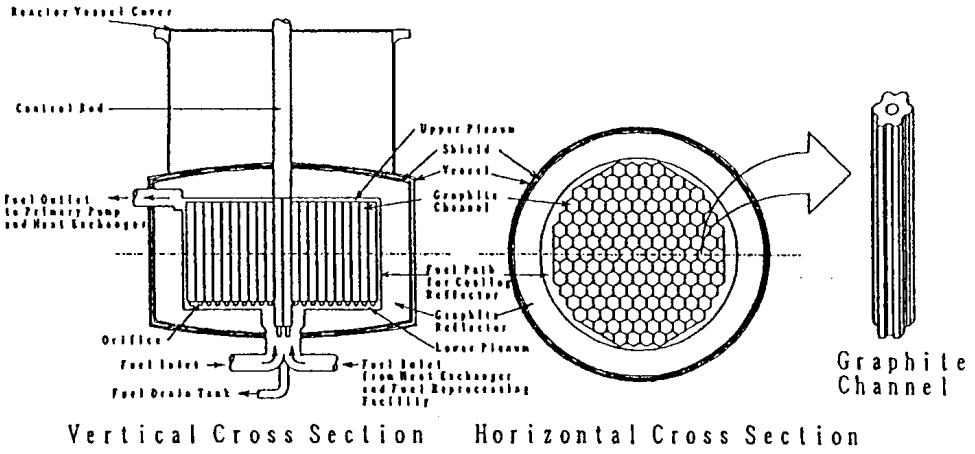


Fig.8 Schematic view of MSR model(2250MWth) adopted for 2-step minor trans- mutation study.

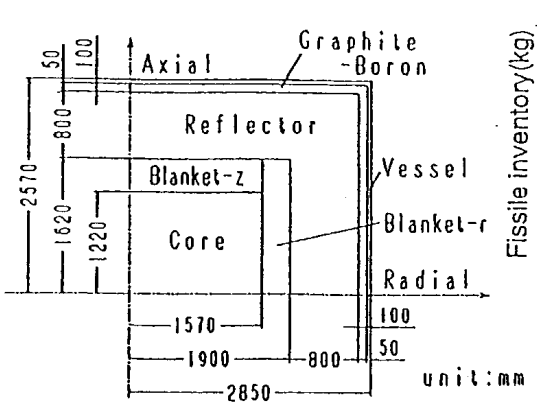


Fig.9 Core model of Pu burning MSR (FUJI-PU3) Power Station. (250MW th)

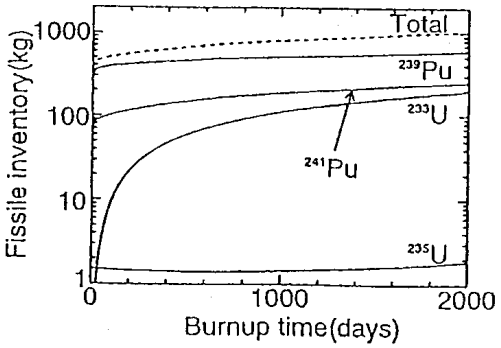


Fig.10 Time change of fissile inventories in Pu burning MSR(FUJI-PU3)

The burn up calculation was carried out by ATOM code developed for this study. In Step1, MA is continuously added by 2kg/day for 13 years, then the reactor is operated for 17 years without adding MA. Then in Step2 which is designed to be suitable for the transmutation of Cm isotopes, MA from Step1 is continuously added 0.81 kg/day for 1800 days, then TRU is burned without adding more MA. The results of the transmutation after 60 years are shown in Table VII.

Table I. Summary of polyethylene moderated and fuelled with thorium and enriched uranium cores at KUCA.

Name of assembly	$H/^{235}U$ ratio	$^{232}Th/^{235}U$ ratio
B3/8”P48EU16Th	322	12.7
B3/8”P45EU18Th	322	15.3
B4/8”P24EU-Th-EU	214	19.1
B3/8“P30EU-Th-EU	161	19.1
B4/8”P24EU-Th-EU-EU	161	12.7

Table II. Parameters of foils used in Cd ratio measurement for KUCA B4/8”P24EU-Th-EU and B3/8”P36EU-NU-EU cores.

Foil	Target nuclide	Thickness (mm)	Mass (mg)	Radius (cm)	Measured nuclide	Resonance energy(eV)
Au	Au-197	0.20	26.47	0.45	Au-198	4.90
In	In-115	1.27	121.1	0.635	In-116m	1.46
Mn	Mn-55	0.50	49.45	0.635	Mn-56	337
W	W-186	1.52	370.8	0.635	W-187	18
Dy	Dy-164	0.25	24.55	0.635	Dy-165	--
Th	Th-232	0.51	75.45	0.635	Pa-233	21.8,23.5
EU-Cu	U-235	1.10	121.4	0.50	Ce-143	--
DU	U-238	0.25	57.60	0.635	Np-239	6.77

Table III. Results of C/E values of Cd ratio for Th and NU loaded cores.

Foil	Th core	NU core
DU	1.093	1.104
Th	1.080	1.053
Au	1.061	1.032
W	0.971	0.949
In	0.957	0.945
Mn	0.950	0.969
EU	1.075	1.057
Dy	1.069	1.094

The main conclusions are:

1. Transmutation rate of total TRU including Pu is 98.2%.
2. Transmutation rate of Am is as much as 99.9% and that of Np is more than 99.9%.
3. Transmutation rate of Cm is 80.2%.
4. Total amount of TRU transmutation is 25.6ton/60years.

Table VIII shows the comparison with other types of reactor, which shows the merit of MSR for the transmutation of MA.

The study of Pu burning MSR (FUJI-PU3) Power Station is carried out by Mitachi et al. [10] in which Pu recovered from the spent fuel of LWR was supposed to be used. The reactor model is shown in Figure 9 and the fuel salt composition is

Table IV. Void and Doppler reactivities and effective delayed neutron fraction (β_{eff}) estimated for PuO_2-ZrO_2-(Y,Gd)-Al_2O_3 fuelled PWR of JAERI's once through Pu burning process.

	Moderator density	BOEC	EOEC
Void reactivity	100%	0.0	0.0
(%Δk/k)	60%	-0.332	-9.74
	30%	-3.42	-28.6
	5%	-17.7	-92.6
Doppler reactivity	Fuel temperature		
(%Δk/k)	1200K	-0.0982	-0.201
	900K	0.0	0.0
	600K	0.107	0.209
	300K	0.253	0.467
β_{eff}		2.87×10^{-3}	3.50×10^{-3}

BOEC: Beginning of the Equilibrium Cycle.
EOEC: End of the Equilibrium Cycle.

Table V. Specifications of Step1 and Step2 cores of minor actinide transmutation MSR.

Step 1	Radial distance	190 cm
	Axial distance	360 cm
	Fuel volume ratio of core	9%
	Fuel salt volume	
	Inside vessel	$5.70m^3$
	Primary loop	$9.49m^3$
	Peaking factor of thermal power	1.782
Initial composition of fuel salt		
component	^7LiF BeF_2 ThF_4	$^{233}UF_4$ $(TRU)F_x$
Ratio (mol%)	71.64 16.00 11.85	0.36 0.15

Step 2	Radial distance	190 cm
	Axial distance	360 cm
	Graphite channel	
	0 to 12 years	
	Type	Normal
	Fuel volume ratio	9%
	12 to 30 years	
	Type	with Hollow
	Hollow part ratio	25%
	Effective Fuel volume ratio	11.7%
	Fuel salt volume	
	Inside vessel	$5.70m^3$
	Primary loop	$9.49m^3$
	Peaking factor of thermal power	1.718
Initial composition of fuel salt		
Component	^7LiF BeF_2 ThF_4	$^{233}UF_4$ $(TRU)F_x$
Ratio (mol%)	71.8 16.0 12.0	0.2 0.0

Table VI. Minor actinide composition loaded for Step 1 core of MA transmutation MSR.

Nuclide	Mass (wt.%)
Np-237	49.14
Am-241	29.99
Am-243	15.50
Cm-244	4.99
Others	0.38

Recovered from spent fuel of burn up of 35GWd/t of 1,000MW(e) PWR cooled 5 years before reprocessing.

^{7}LiF-BeF_2-ThF_4-UF_4-PuF_3=69.65-22-8-0.1-0.25 mol%. The Pu composition is ^{238}Pu-^{239}Pu-^{240}Pu-^{241}Pu-^{242}Pu = 1.5-55.0-25.3-13.2-5.0 atomic %. The FVR is 0.18. The fuel salt temperature of inlet is assumed to be 840ºK and that of outlet is 940ºK. The feeding rate of Pu and Th is shown in Table 9 which is determined so as to keep the excess reactivity is in the range of 1.50~1.65%. The burn up calculation up to 2000 days was carried out with ORIGEN code where one group cross section is prepared by SRAC and RESEND code which is a computer program for reconstruction of resonance cross sections from the nuclear data file. The changes in fissile inventories in the core and the integrated amount of Pu generated in the core are shown in Figures 10 and 11, respectively. Table 10 shows the inventory and feed of heavy metals. From the table, it is concluded that 507kg of Pu is destroyed and 194kg of ^{233}U is produced during 2000 days of operation. If the reactor operation is normalized to 1GW(e) generation, MSR transmutes 925kg of Pu annually, which is larger than the amount transmuted by MOX (649kg). The net fissile uranium produced annually is 362kg.

From these experiences, we have proposed to study MSR in the IAEA Coordinated Research Program (CRP) on the Potential of Thorium-based Fuel Cycle to Constrain Pu and to Reduce Long-term Toxicity. In this study [11], the burn up characteristics of a 200MW(th) MSR consuming Pu from LWR and converting into ^{233}U was calculated. In this study, 2-region cell composed of fuel salt which flows in the central circular hole opened at the center of hexagonal moderator graphite column as shown in Figure 12 was adopted.

The fuel salt composition was selected as ^{7}LiF-BeF_2-ThO_2-PuF_3=72.16- 16.0-11.4-0.6 mol% for the initial loading with the FVR of 20% to obtain negative temperature coefficient. The core average temperature was assumed to be 900ºK. Plutonium composition is that of given for the benchmark problem of (Pu-Th)O_2 lattice of LWR of the CRP [12]. The initial atomic number densities are shown in Table XI and the reactor model is shown in Figure 13.

Table VII. Results of two-step MA transmutation for 60years.

	Loaded (kg)	First Step (kg)	Second Step (kg)	Two-step transmuted (kg)	Ratio (%)
Np-237	12,818.1	87.8	69.2	Recycled to	
Pu-238	0.0	2,509.6	95.8	Another	
Pu total	0.0	3,370.8	117.2	step 1 core	
Am-241	7,821.7	21.7	0.4	-7,821.3	99.9
Am-243	4,043.5	178.8	2.0	-4,041.5	99.9
Am total	11,885.3	201.1	2.3	-11,882.9	99.9
Cm-244	1,301.4	546.3	4.8	-1,296.7	99.6
Cm-245	67.3	224.0	2.2	-65.1	96.7
Cm-246	0.0	465.9	188.9	+188.9	-----
Cm-total	1,382.1	1,342.6	273.4	-1,108.6	80.2
Total	26,085.4	5,362.3	475.0	-25,610.4	98.2

Table VIII. Comparison of MA transmutation with other types of reactors.

		PWR	FBR	MSR Two-step
Thermal power	(MW(th))	3,424	3,000	2,250
Cycle length	(day)	840	1,080	----
TRU loaded	(ton)*	32.97	92.40	26.09
TRU transmuted	(ton)*	11.77	29.21	25.61
TRU transmutation	Ratio(%)*	35.70	31.61	98.18
TRU transmuted	(kg/1GW(th) a)*	19.37	54.84	63.24

*PWR and FBR; 6 units operation per 60years(core life is 30years).
TRU means MA (excluded Pu).
MSR; operation of 5 Step1 core and 1 step2 core.
TRU means MA and Pu.

Table IX. Feeding rate of Pu and feeding amount of Th for Pu burning MSR (FUJI-PU3)
Feeding rate of plutonium.

days	0-20	20–100	100-200	200-300	300-500	500-700	700-1000	1000-1300	1300-1600	1600-2000
g/day	6500	1300	900	850	730	750	590	620	600	610

Feeding amount of thorium

days	100	200	300	500	700	1000	1300	1600
kg	144	220	200	400	0	150	170	200

Table X. Inventories and feeds of heavy metals for plutonium burning MSR (FUJI-PU3).

	Initial Inventory (kg)	Feed 0-2000 days (kg)	Inventory 2000 days (kg)
Th total	16461	1484	17698
Pa-233	0		3.97
Pa total	0		4.67
U-233	0		194
U-235	1.52		1.86
U total	211		396
Pu-239	292	821	598
Pu-241	70.1	197	247
Pu total	531	1492	1516
Am total	0		50.0
Cm total	0		8.90

The calculation was carried out by SWAT code system that is basically the combination of SRAC-ORIGEN2 and used for the IAEA benchmark calculation. Pu was assumed to be fed continuously so as to keep k_{eff} nearly unity. The change in k_{eff} is shown in Figure 14. Although in this case, the initial decrease is too large but it could be adjusted easily. As for the changes in the atomic number densities of main isotopes, it was assumed that ^{135}Xe is completely removed,

Table XI. Initial atomic number densities (n/cm^3) of MSR used for IAEA Coordinated Research Program of Th fuel cycle perspectives.

Th-232	3.649E-03
Pu-238	1.908E-06
Pu-239	1.172E-04
Pu-240	4.539E-05
Pu-241	1.505E-05
Pu-242	9.531E-06
Li-7	2.260E-02
Be-9	5.037E-03
F-19	4.785E-02
C-12	9.226E-02

Table XII. Changes of Pu and Pa-233+U-233 inventories of MSR for IAEA Coordinated Research program of perspectives of Th fuel cycle.

Items		With Xe(kg)	Without Xe(kg)
Pu initially loaded	Pu total	250.0	250.0
	Pu fissile	184.6	184.6
Pu added until	Pu total	350.0	350.0
1620 days*	Pu fissile	258.2	258.2
Pu left at	Pu total	89.4	83.8
1620 days	Pu fissile	30.2	27.4
Pu-233+U-233 left at	1620 days	113.4	108.9
Pu added until	Pu total	490.0	490.0
2250 days	Pu fissile	361.8	361.8
Pu left at 2250 days	Pu total	80.0	77.1
	Pu fissile	27.6	26.0
Pa-233+U-233 left at	2250 days	115.9	112.4

* Pu added at 1620 days is excluded

but the results are similar for the case with ^{135}Xe except for k_{eff}. Table XII shows the change of ^{233}Pa and ^{233}U inventories at 1620days that correspond about 60MWD/kg of burn up for the initial loaded heavy metal and at 2250days that corresponds to about 86MWD/kg. This system consumes about 2.6 times of initially loaded Pu and makes the fissile Pu content from 74% to 33% in 2250days. It looks the system is promising from the standpoint to constrain Pu and to convert into ^{233}U although it had better to take out ^{233}U(+^{233}Pa) at 1620 days since after that ^{233}U is more or less works for TRU transmutation.

5. Molten Salt Accelerator Breeder Reactor

The concept of accelerator driven molten salt reactor (A-MSB) was studied by Furukawa et al [13]. Recently a plan is approved to begin preparatory work to launch the study of Th fuel cycle and a subcritical system driven by an accelerator. This year, an accelerator suitable for the energy amplifier, high energy neutron behavior in accelerator driven subcritical systems and expected results in KUCA if it were loaded with 11.5% ^{233}U enriched fuel will be mainly investigated. In this preparatory study, the research theme and problems to be solved as the energy amplifying system of A-MSB will be specified and it is determined if this work should be adopted as a subject of the Japan Society of Scientific Promotion.

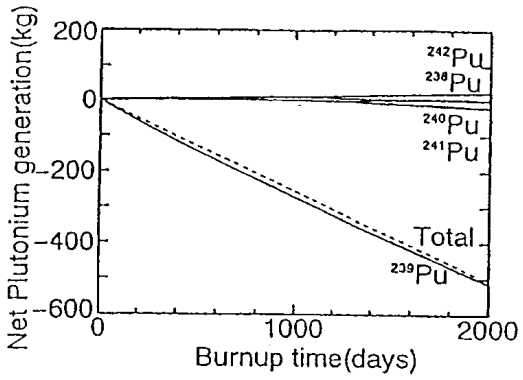
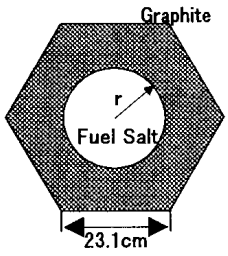


Fig.11. Time change of integral amounts of Pu isotopes generated in the core of Pu burning MSR(FUJI-PU3) from startup.



Case	V_F/V	r(cm)	X(mol %)	Y(mol %)
Case1	0.05	4.697	11.8	0.2
Case2	0.1	6.64	11.84	0.16
Case3	0.2	9.3946	11.4	0.6

(Fuel : LiF-BeF_2-ThF_4-PuF_3=72:16:X:Y mol %,Graphite : 1.840g/cm^3)

Temperature : 900k

Fig. 12 Cell model of MSR used for burn up calculation of IAEA Coordinated Research Program(RP) on Perspectives of Th fuel cycle.

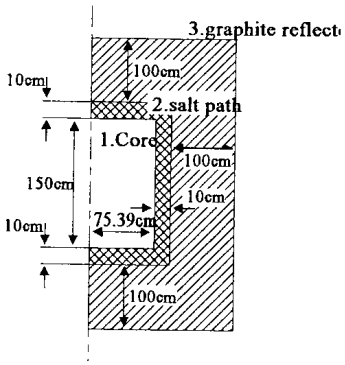


Fig.13 Reactor model of MSR(200MWth) for IAEA CRP.

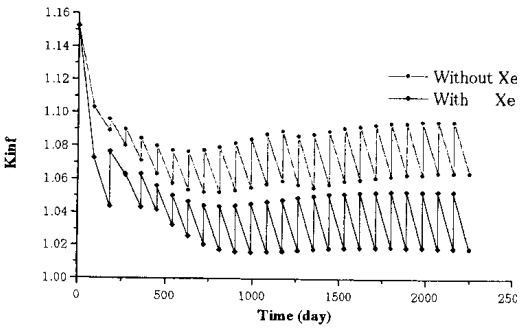


Fig.14 Change in k-inf during burn up of MSR of IAEA CRP.

6. Concluding Remarks

The present status concerning Th fuel research in Japan was reviewed stressing in neutronics aspects. Although the research was not so systematic, if the preparatory work recently approved is successful, the organized work will become possible in the future.

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THORIUM FUEL CYCLE CONCEPT FOR KAERI'S ACCELERATOR DRIVEN SYSTEM PROJECT*

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Abstract. Korea Atomic Energy Research Institute (KAERI) has been carrying out accelerator driven system related research and development called HYPER for transmutation and energy production. HYPER program is aiming to develop the elemental technologies for the subcritical system by 2001 and build a small bench scale test facility (~5MW(th)) by the year 2006. Some major features of HYPER have been developed and employed, which are on-power fueling concepts, a hollow cylinder-type metal fuel, and Pb-Bi as a coolant and spallation target material. Another fuel cycle concept for HYPER has been also studied to utilize thorium as a molten salt form to produce electricity as well as to transmute TRU elements. At the early stage of the fuel cycle, fissile plutonium isotopes in TRU will be incinerated to produce energy and to breed ^{233}U from thorium. Preliminary calculation showed that periodic removal of fission products and small amount of TRU addition could maintain the criticality without separation of ^{233}Pa. At the end of the fuel cycle, the composition of fissile plutonium isotopes in TRU was significantly reduced from about 60% to 18%, which is not attractive any more for the diversion of plutonium. Thorium molten salt fuel cycle may be one of the alternative fuel cycles for the transmutation of TRU. The TRU remained at the end of fuel cycle can be incinerated in HYPER having fast neutron spectrums.

INTRODUCTION

Most of the existing reactors in Korea utilize the low enriched uranium. Since spent fuels resulted from these reactors contain long-lived radionuclides including plutonium isotopes, Korean government has paid great attentions how to handle them. Plutonium isotopes should be either completely isolated from the biosphere due to its toxicity, or utilized effectively without any possibility for its diversion to military purpose. Though direct disposal of spent fuels in deep geological repository has been considered, it seems not to remove all the problems. Rather than just covers all of the dangerous possibilities, there must be a safe and more effective way to deal with this notorious waste.

Thus KAERI has initiated the transmutation research for minor actinides and long-lived fission products since 1992. Some feasibility studies were performed and a couple of basic guidelines were introduced to decide the type of transmutation system [1]. An accelerator driven subcritical reactor, named HYPER (Hybrid Power Extraction Reactor) was found to be promising for the transmutation purpose. HYPER research is being performed within the frame of the national long term nuclear research plan. The whole development schedule is subdivided into two phases. The design concept of the transmutation system and some basic key technologies are scheduled to be developed in Phase I (1997 – 2001). A small scale test facility (~5MW(th)) is to be designed and built in Phase II (2002 –2006). 1 GeV 16mA proton beam is designed to be provided for HYPER. HYPER is planned to transmute about 380 kg of TRU a year and produce 1000 MW(th) power. The support ratio of HYPER to LWR units producing the same power is assumed to be 1 to 5 ~6.

First of all, the fuel cycle concept was focused on transmutation of long-lived radionuclides, thorium utilization and proliferation resistance. Among various fuel types evaluated for the system, thorium molten salt fuel has been one of the choices for the purpose. The fuel cycle consists of following characteristics:

* 1998 meeting.

- Enrichment facility is not required for the fuel cycle.
- Plutonium is not separated from other TRU elements.
- TRU elements separated by pyrochemical processes are mixed with thorium as a fluoride form.
- There is no separation process for protactinium.

WHY THORIUM MOLTEN SALT FUEL IS BENEFICIAL?

The advantages of thorium fuel cycle as compared to uranium-based fuel cycle involve a significant reduction in the yield of transuranic actinides, especially plutonium, and production of ^{233}U during incinerating fissile materials [2]. The ^{233}U bred by thorium is a superior fissile material for thermal reactors than either ^{235}U or ^{239}Pu. The thorium fuel cycle has an attractive negative temperature coefficient in thermal reactors that enhance reactor safety. The production of fission products which is the main contributor to reactor poisoning is about 25% less for ^{233}U than for ^{235}U or ^{239}Pu. Finally, thorium is an abundant resource than uranium. The neutronic properties of each nuclear fuels are described in Table I [3].

Table I. Neutronic properties of each nuclear fuels

Isotope	^{235}U	^{239}Pu	^{233}U
Obtained from	Natural U	^{238}U	^{232}Th
Neutron produced per			
- Fission	2.418	2.871	2.492
- Thermal neutron absorbed	1.98	1.86	2.2
Absorption cross section, b			
- Thermal neutrons	555	1618	470
- Fast neutrons	1.5	2	2

When the number of neutrons produced per neutron absorbed in fissile material is greater than 2.0, it is theoretically possible to generate fissile material at a faster rate than it is consumed. In thermal reactors fueled with plutonium, the number of neutrons produced per neutron absorbed is less than 2.0 and breeding is impossible. For ^{233}U, on the other hand, this number is substantially greater than 2.0, and breeding is practicable in a thermal reactor.

Another advantage of using thorium molten salt fuel is that the separation of plutonium from other TRU elements is not necessary. MOX fuels for PWR and FBR are required to separate plutonium. Plutonium presents a difficult problem because it cannot be simply denatured. It has been known that almost any combination of plutonium isotopes can be made into a weapon unless the ^{242}Pu content is very large. Thus, pure plutonium separation from spent fuels should be prohibited all the time.

Molten salt fuel cycle was introduced because it gave many advantages [4]. There is no fuel fabrication process and it facilitates to remove the fission products periodically and to provide homogeneous burning of the transmuted materials. The fission products to be removed periodically will be noble gases, seminoble and noble metals.

PRELIMINARY CALCULATIONS OF THORIUM MOLTEN SALT FUEL

Preliminary calculation was made by MCNP and ORIGEN code for following conditions. Initial core of the thorium molten salt reactor had 20.9 ton of thorium and 8.89 ton of TRU to produce 1,000 MW(th) power. The average neutron flux at the beginning of cycle was assumed to be ~$5x10^{14}$ n.s/cm^2.s. The net multiplication factor at the beginning of the cycle was assumed 1.049. First removal of fission products and 100 kg of TRU addition were made after 700 days at the beginning of the cycle. Thereafter every 1,000 days fission products were removed and 80 kg of TRU were added. TRU addition was necessary to maintain the criticality. The thorium concentration and total inventory of actinides were allowed to decline naturally. The isotopic compositions of TRU at the beginning were shown in Table II with those at the end of the cycle.

Table II. Isotopic composition of TRU at the beginning and the end of thorium fuel cycle.

Nuclide	Weight Fraction (%) Beginning of Cycle	Weight Fraction (%) End of Cycle
Np-237	4.6	0.9
Pu-238	1.4	9.2
Pu-239	52.1	7.3
Pu-240	23.7	43.0
Pu-241	7.7	10.3
Pu-242	4.5	15.4
Am-241	5.0	4.2
Am-243	0.8	3.9
Cm-244	0.2	4.3

As can be seen, fissile plutonium contents in TRU were reduced significantly from 59.8% to 17.6%. Total added TRU amounted to 9,546 kg and 3,432 kg of TRU was remained at the end of the cycle. Thus 6,114 kg of TRU were consumed for 32 years which was equal to 190 kg of TRU burning every year. For thorium 8,160 kg were consumed and 1,662 kg of ^{233}U were remained in the used molten salt fuel.

From the calculation, it is obvious that the thorium fuel cycle can have a significant impact on the disposal problem of PWR spent fuels and can effectively utilize fissionable TRU elements to generate new fissile materials from thorium. The TRU production level of thorium fuel cycle was lower than that of the uranium cycle. Thus thorium molten salt fuel cycle may be one of the alternative fuel cycles for the transmutation of TRU. The TRU remained at the end of the thorium molten salt fuel cycle can be incinerated in the system having fast neutron spectrums.

PYROCHEMICAL PROCESSES FOR HYPER FUEL CYCLE

There are many possible processes to separate TRU from spent fuels. Because Purex process, though well established, is considered not to be proliferation resistant, the combination of pyrochemical processes will be employed to separate TRU from spent fuels. Since the decontamination factor of pyrochemical processes is not sufficiently high, it is well known that TRU obtained by these processes cannot be utilized for military purposes without further purification.

Fig.1 shows the pyrochemical processes for the separation of uranium and TRU from PWR spent fuels. After clad materials are removed from spent fuels, either fluorination process or direct oxide reduction process will be applied. If fluorination process is chosen, uranium hexafluoride can be separated easily by its high volatility and be converted to uranium dioxide fuels for CANDU reactors. TRU can be separated from fission products by pyrochemical processes [5]. Finally it is converted to fluoride forms and mixed with thorium molten salt fuels. If direct oxide reduction process is applied [6], electrorefining process will provide the separation of uranium, fission products, and TRU elements.

Fig.2 briefly shows the concept of flow diagrams of HYPER fuel cycle during operation. Fission products will be removed periodically and thorium, uranium and TRU will be re-circulated. No other separation processes such as protactinium separation will be considered.

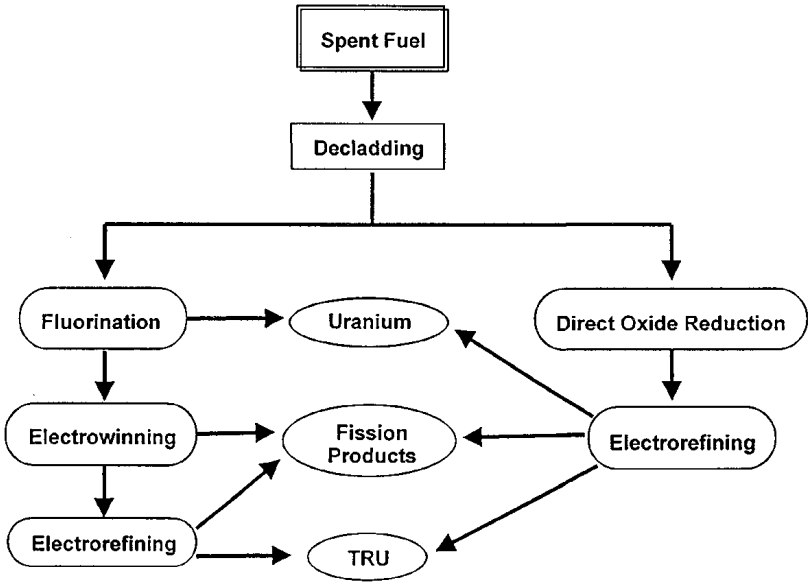

Figure 1. Pyrochemical processes for HYPER fuel cycle.

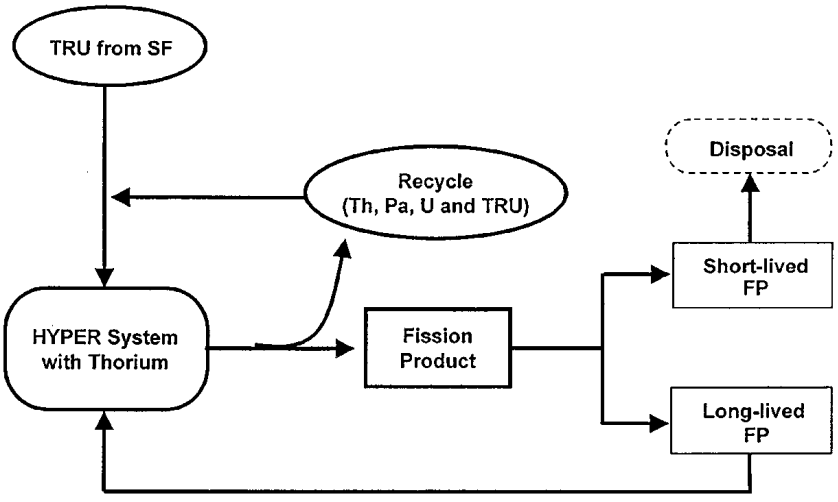

Figure 2. Fuel cycle for HYPER system.

PROLIFERATION RESISTANCE OF HYPER FUEL CYCLE

Proliferation-resistant fuel cycle is defined as one in which at each point of the cycle the fissile material is so degraded that it is not realistic to extract it and use it to produce a fission weapon.

In this fuel cycle concept, because the separation of protactinium may be not sufficiently proliferation resistant, the isolation process of protactinium would not be involved [6]. As a result, ^{233}U is always contaminated by ^{232}U and its daughter products, some of which are hard γ emitters. This makes it much more difficult to handle. By contrast, Pu as an α -emitter can be more easily diverted.

It has been known that ^{233}U is an inferior material for arsenal purpose than ^{239}Pu because nuclear weapon basically depends on fast fissions. ^{233}U can be easily denatured by the addition of ^{238}U at the beginning of the cycle if really needed. This can ensure that no weapon's grade uranium is present at any point of the thorium cycle though it is contradicted to the transmutation purpose.

The fissile plutonium isotopic composition at the end of the thorium molten salt fuel cycle is transformed to uninteresting composition as a weapon material. Thus using TRU for thorium fuel cycle may be more proliferation resistant than direct disposal of spent fuels.

HYPER FUEL CYCLE FOR TRANSMUTATION AND ENERGY PRODUCTION

Depending on the nuclear programs of each country, reactor types and fuel cycles for the transmutation may be different. In Korea where PWR and CANDU are being operated, the study on the utilization of uranium in spent fuels of PWR has been performed and Direct Use of PWR spent fuel In CANDU (DUPIC) Program is in progress. In order to utilize the plutonium, reprocessing of spent fuels and fabrication of MOX fuels for PWR in foreign country are being considered. Because Korea does not have a reprocessing plant, a combination of pyrochemical processes is being considered to separate TRU and long-lived fission products from the PWR spent fuels for ADS purpose. Fig.3 shows a brief concept of HYPER fuel cycle combined with other fuel cycles.

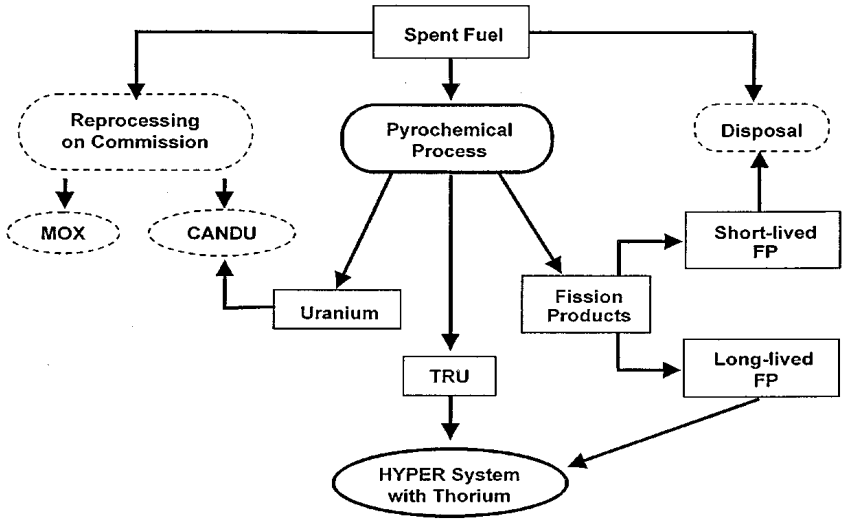

Figure 3. Concept of HYPER fuel cycle.

The uranium, which is a by-product of the pyrochemical process, would be used for CANDU fuels. And TRU will be sent to either thorium molten salt reactor for breeding ^{233}U or ADS for transmutation. In order to use thorium as a nuclear fuel, a neutron source such as ^{235}U, ^{239}Pu, or an accelerator is necessary to supply enough neutrons. In the thorium molten salt reactor, fissile materials in TRU are incinerated to produce neutrons and energy. Some of the neutrons are used to breed ^{233}U and the other neutrons are used to maintain the criticality of the molten salt reactor. The TRU remained at the end of the thorium fuel cycle can be incinerated in HYPER having fast neutron spectrums.

FUTURE DEVELOPMENT PLAN

- Further code calculations will be made for following conditions:
* Only the noble gases, seminoble and noble metals among fission products are removed.
* Where the thorium content is kept constant.
- Effective separation process of fission products from the used thorium molten salt fuel will be evaluated.
- Oxide removing process from the molten salt fuel will be studied.
- Solubility of TRU and fission products in thorium molten salt will be measured.
- Methods to control the Redox and activity conditions of molten salt fuel will be studied.
- On-line analytical techniques will be developed.

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BNFL's VIEW OF THE THORIUM DEVELOPMENT PROGRAMME*

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Abstract. The value of an innovation depends on the urgency of problems or deficiencies that it is intended to overcome, on its effectiveness in doing so, and inversely on the costs and any new difficulties that it raises. Thus industry sees no net benefit in thorium simply as a substitute for uranium except perhaps in certain areas such as India, since a general shortage is not expected for several decades. Interest is directed more at the property of thorium in generating hardly any transuranic elements apart from a little neptunium; using it would minimize additions to inventories of these elements, or increase net rates of consumption in schemes to incinerate them.

1. INTRODUCTION

Thorium is claimed to have the advantages of reducing risks of weapon proliferation associated with plutonium stockpiles; the risks to future generations from minor actinides in waste; and public opposition to the industry on the grounds of these risks.

The importance of anti-proliferation measures, long-term waste management and public acceptance is undisputed; whether in these respects thorium offers enough advantages over the uranium-plutonium cycle to warrant changing the basis of the whole industry is more questionable. On the issue of proliferation, as pointed out on previous occasions, [1,2] the considered opinion within BNFL is that any risk due to civil plutonium is already adequately controlled, depends on the existence rather than the size of the stockpile, and would not be greatly changed by foreseeable additions or reductions; while uranium-233 bred from thorium carries its own proliferation risks, diminished but not eliminated by contamination with U-232 and its decay products.

Again, in relation to long-lived wastes, the minor actinides represent only part of an acceptably small risk, shared in comparable proportions with certain fission products also produced by the thorium cycle; while substantially eliminating this risk from either cycle, even if practicable, would incur costs and operational hazards liable to outweigh any future benefit.

A further major issue is that the thorium fuel cycle involves substantial technical difficulties in the main-line process, not certain to be satisfactorily resolved on a commercial scale. Other approaches to the proliferation and waste issues therefore seem likely to remain more cost-effective for the time being. Nevertheless, technical developments, political pressures or changes in regulatory policy might alter the position, particularly in relation to states newly acquiring nuclear technology. The merits of thorium in incinerating waste or surplus plutonium could then become important. Its interesting possibilities as a fertile poison, to limit the reactivity swing in uranium or plutonium systems, also need to be recognized.

So far, most studies on thorium fuels have been rather speculative, and none the worse for that as a necessary preliminary to serious industrialization. However, it may be time to consider realistically which arguments for thorium are the most cogent, which of the various ideas for utilizing its properties are most likely to attract industrial interest, how they are most likely to

* 1998 meeting.

be effectively realized, and in what direction the inevitable costs of development would most usefully be channeled to satisfy the industrial driving criteria.

2. DRIVERS FOR ADOPTING THORIUM

Whatever may be the technical merits of thorium, it will not be widely adopted unless utilities see a commercial benefit, regulatory bodies insist, or governments make special arrangements for public-relations or other political reasons.

2.1. Commercial aspects

The significant considerations are in manufacture (materials, fabrication and recycling), utilisation and waste management. Of the materials, thorium itself is relatively cheap, while plutonium as a likely fissile component might be cost-free at least in the early decades. In fabrication, the complication of adding plutonium to a fertile matrix is at present rather more expensive than the equivalent isotopic enrichment of uranium, but for UO_2/PuO_2 the difference is expected to diminish, disappear or reverse with increasing experience and throughput. The balance of manufacturing costs is therefore uncertain, after the substantial investment needed in new plant and infrastructure. In the longer term, utilisation would presumably raise the price of thorium as a commodity and perhaps give plutonium a commercial value.

Thorium could have a particular advantage in hybrid or mixed fuels, as a fertile poison to damp down initial excess reactivity while breeding ^{233}U to replace the fissile content consumed. This could permit simplified reactor control measures or extended fuel residence.

In the reactor, there may be little to choose economically between thorium and uranium fuels. However, the process of changing to thorium would again be inevitably expensive. Even in a case claimed to be favourable, with fuel assemblies externally similar to those of conventional design and no major changes to the rest of a standard light-water reactor, [3] the control rods would apparently have to be rearranged, with whatever that might involve in outage time and discharge of partially-irradiated fuel.

Waste management issues will depend on the form of cycle. Assembly for assembly in a once-through regime, thorium would be very similar to uranium, except for extra shielding against gamma radiation from ingrown thallium-208. On the other hand, if high irradiation were achieved the number of assemblies discharged per unit of electricity generated would diminish. In a closed cycle, reprocessing difficulties would probably make thorium more expensive.

On the whole, thorium appears unlikely to offer a convincing economic advantage for the foreseeable future. The closest approach could well be as fertile neutron poison. Utilities have resisted its introduction even in India, but seem readiest to accept it in its capacity to reduce the reactivity swing during the course of a reactor cycle.[4]

2.2. Safety

Regulators would need to be thoroughly convinced that safety would not be compromised. The operating parameters of thorium fuel appear to be adequate, and containment of fission products tighter than in uranium oxide. Whether this advantage would be retained at longer

irradiation times, such as might be desirable in view of reprocessing difficulties, is less certain. If a closed cycle were adopted, safety issues in reprocessing would more probably arise in connection with potential for mishaps than in normal operation (section 3.4). The extent of such problems may not become apparent until relatively late in a development programme, when it passes from laboratory verification to full-scale engineering.

Minimizing production of minor actinides, ostensibly on grounds of safety, avoids only remote risks and slight additions to natural radioactivity. The improvement is by too small a factor to make a convincing case, since long-lived fission products are still produced in the thorium cycle, so this is more an issue in public relations or politics than in technical terms.

2.3. Proliferation resistance

An argument forcefully proposed for replacing uranium with thorium is that it would reduce the risks of weapon proliferation, and specifically of nuclear terrorism, associated with plutonium. Against this, in so far as that risk may lie in the possibility of theft from present stockpiles, it appears insensitive to additions or likely reductions in the inventory, and so to the choice of fuel cycle. Where there is a question of creating a new inventory in a state not already possessing nuclear technology, the case may however be rather different.

If such a state is willing to comply fully with IAEA safeguards procedures, and trusted to continue indefinitely meeting its obligations thereunder, the problem is to that extent remote. If the state were to refuse compliance, nuclear enterprises elsewhere could be expected to withhold co-operation, and their own technological course would then be largely irrelevant. Where the question of how far to export nuclear capability becomes difficult is with a hypothetical state that presents no convincing grounds for refusing the benefits of nuclear power, but in some respect raises doubts about its ability to restrict the materials to purely civil purposes or wishes to minimize its responsibility for doing so.

In one suggested solution, [5] a small, simple reactor fuelled for life with the intention that the core should be returned intact to the vendor at the end of that life, the property of thorium in conferring a low reactivity swing would be a particular advantage. Difficulty in reprocessing might be welcomed in case the host country should break its contract and attempt to recover fissile material from the fuel, although it would be a fragile barrier since difficulties tend to be more readily overcome for military than for civil ends.

2.4. Environmental conservation

The chief environmental burden generated by the nuclear industry is the waste from mining and milling operations. For thorium, residues from the rare-earth industry, of uncertain extent but perhaps capable of yielding a thousand tons, could initially be processed without further mining. Thereafter the high proportion of thorium in the best ores would afford some benefit in the scale of operations, although it would be diminished by the drastic chemical conditions needed and would decline as poorer sources had to be exploited. However, the claimed advantage over. uranium applies in any case only to virgin material; still greater benefits in this sense could be achieved more easily by using recovered or depleted uranium which is readily available in tens or hundreds of thousand tons and could be brought directly into the cycle.

In reactors, improved retention of fission products may be an advantage, increased production of krypton-85 a drawback. The net effect is probably slight.

Any clear environmental advantage of thorium is therefore limited to its low production of transuranic elements and consequent reduction in the risk to future generations due to assumed leakage of long-lived radionuclides from waste repositories. As considered above, this is essentially a political rather than a technical issue.

2.5. Public acceptability

The main anxieties exercising the public on the subject of nuclear power are about the risk of major accidents, the likelihood of nuclear proliferation, and the effects of artificial radionuclides in the environment. The first topic involves such esoteric questions of physics and engineering that whatever the technical arguments may be, they will make no favourable impression outside the industry. Changing from uranium to thorium would therefore have little impact in this respect, except perhaps to diminish whatever confidence remained in the older system.

On weapon proliferation, criticism of the industry is usually in the terms that a certain quantity of plutonium amounts to so many bombs' worth, and increasing the number attracts much more attention than reducing it. Inventory reductions that might be practicable over the next few decades would not eliminate the supposed potential for immense destruction, and could not disarm that criticism.

Health risks from nuclear sources arouse concern unrelated to their actual magnitude. If they are a thousandth of those due to natural causes, reducing them to a ten-thousandth - indeed, any step short of total elimination - is unlikely to make much impression.

The most determined opposition to nuclear power comes from zealots interested in problems, not solutions, with the declared objective of closing down the industry. For tactical reasons, they may favour a particular development so long as it remains a hypothetical measure that the industry can be accused of culpably neglecting, and replacing uranium by thorium may appear in that light; but presented with an actual proposal that could prolong the nuclear age, they would assuredly find grounds for objection.

Nevertheless, their stated views make a significant contribution to the political pressures on the industry. To that extent, and regardless of practical considerations, there could be advantage in being seen to investigate the possibilities and drawbacks. On the other hand, the very fact of contemplating alternatives to actual practice is liable to be taken as confirming that an intolerable risk exists, so presentation requires the utmost care.

2.6. Summary of drivers

At present there is no obvious commercial drive for thorium, a doubtful advantage in safety or public acceptability, and a real but slight benefit to be expected for the future environment. The argument for proliferation resistance appears sound only in relation to states holding no acknowledged plutonium stocks, but could become increasingly important if nuclear technology were exported to developing countries.

Such a case appears to be the only one in which there is a significant proliferation risk that thorium would substantially diminish. It might also be among the most favourable for using thorium to limit reactivity swing. However, even if the concept of a segregated market were acceptable, to expect that the customer state or group of states should bear the whole burden of adopting a new fuel system would be unrealistic; so is the idea that utilities in the rest of the world would subsidise it voluntarily without some compensating benefit to themselves. Unless prospects of such a benefit can be demonstrated, the development is therefore unlikely in the short to medium term without governmental intervention on non-commercial grounds.

Government action could also require recycling of minor actinides for essentially political reasons, and this may well become one of the main drivers for using thorium in the short to medium term. Beyond that, of course, the consumption of uranium reserves and a concomitant increase in price can be expected to provide a commercial incentive, the earlier if objections to fast reactors are not overcome.

3. DEVELOPMENT CONSIDERATIONS

Much of the work recently reported on thorium as a fuel relates to irradiation characteristics of the fuel itself or of candidate nuclides for incineration. This is important, but so is the rest of the cycle, which seems to have received less attention.

3.1 Fuel manufacture

Thorium fuels have been made in the past, and for oxide, one presumably suitable technology would be similar to that already established industrially for uranium oxide and MOX fuels formed from pellets in tubular cladding. Separate plant would be needed to avoid cross-contamination, and the optimum conditions could well be rather different, but no serious difficulties seem likely in the first cycle. If interest passes to nitride or other less familiar forms, then industrialisation is likely to need a more radical development programme.

To recycle thorium and ^{233}U, containing ^{228Th} and ^{232}U with their high-energy gamma- active daughters, operations would need heavy shielding and remote handling. The problem differs in degree rather than kind from that with highly-irradiated plutonium, and provided that the remote manipulative techniques can encompass all relevant maintenance requirements, its importance is likely to be chiefly economic rather than technical or operational. Otherwise radiation doses to personnel would become a serious consideration. The development priorities should therefore be to minimise the manipulation needed in the manufacturing process and particularly in maintenance operations, to maximise the use of robotics for such manipulation, and to improve its reliability.

Such considerations would favour manufacturing techniques more amenable than pellet formation to remote operation, such as vibro-packing with less intricate manipulative stages. Problems in fabrication could in principle be avoided almost completely by using a molten-salt fuel with no structure at all, but the corresponding reactors appear to be technologically a generation or more further away.

Recycle fuel once manufactured would still need shielding during transport and storage, but again the penalty lies in additional costs rather than difficulties of principle. Any development is likely to be evolutionary along existing lines.

Waste radionuclides added to a reactor load could in principle be either homogeneously or heterogeneously distributed. There could be advantage, in a complete separation of minor actinides, with neptunium in the core, americium at the periphery and curium (which cannot be effectively incinerated in the usual range of neutron spectra) allowed to decay to plutonium before incorporation; its radioactivity and heat generation would then present no immediate difficulties in fabrication. Recycling undifferentiated minor actinides would however raise issues of gamma and neutron irradiation. Requirements would be specific to thorium only in so far as it is the favoured matrix for such operations.

If the technical case for recycling minor actinides is to be taken seriously, the long-lived fission products must be treated likewise. Given a sufficient neutron flux, iodine-129 and technetium-99 can be incinerated satisfactorily, although the volatility of iodine will require a careful choice of target material. Caesium-135 cannot usefully be incinerated without a preliminary isotopic separation that is unlikely to be economically practicable; sooner or later serious thought must be given to the question of whether its effective exclusion from recycling undermines the whole rationale behind the process, and indeed much of the drive for thorium.

3.2. Reactors

An important consideration here is to avoid the mistake of the early nuclear industry in pursuing too many types at once. Recently, research specifically on reactors" suitable for thorium has been largely theoretical and relatively inexpensive, but the time for concentrating on those most promising may be approaching. This, and the choice itself, will of course be matters for judgement by specialists in the field.

The easiest way to introduce thorium fuels initially would probably be in existing reactors, so that forms designed as direct replacements for uranium, such as the Radkowsky pattern, [3] are of particular interest.

3.3. Managing irradiated fuel

Management policy will depend very much on the purpose of adopting thorium. If it is to aid the disposal of unwanted materials, whether plutonium or minor actinides, then the diminishing return with prolonged residence must be taken into account: no incinerable component can be entirely consumed in a single irradiation. The actual reduction may be considered sufficient for particular purposes if an acceptably small residue (or in the case of plutonium, a sufficiently low fissile content) is to remain in the matrix for direct disposal: however, a very wide spread of opinions can be expected on what may constitute a sufficient reduction in this context, and provision should be made for a requirement to recover and recycle the remaining portion.

If thorium is used chiefly as a fertile poison to limit reactivity swing, then reprocessing may perhaps, but not necessarily, again be required to recover the fissile product. It would certainly be needed for an indefinitely sustained generating programme.

Such a requirement, for whatever reason, would virtually rule out coated-particle fuel of the HTR type, since no means of breaking it down for dissolution or other treatment seems likely to be both technically and economically acceptable. On the other hand, given adequate assurance on long-term stability, such fuel might have considerable advantages in once-through applications. The chemical stability of thorium dioxide would also be an advantage,

provided that the structure was not unduly disturbed by fission products, but this proviso needs to be verified particularly at the high bum-up values claimed to be feasible. [6]

To the extent that irradiation targets are distinct from the fuel proper, their management can be optimised independently, regardless of whether reprocessing or direct disposal is chosen for the main cycle. Accordingly they are better discussed elsewhere.

3.4. Reprocessing

In process terms, thorium differs from uranium chiefly in (a) the inertness of thorium oxide to nitric acid, and (b) being less extractable by an order of magnitude into tributyl phosphate.

The classic Thorex process, needing fluoride to assist dissolution, introduces corrosion problems alleviated by complex formation with aluminium; this, however, provides no protection in the vapour spaces where a significant risk could arise. Possible approaches might be to look for resistant materials of construction (likely to be expensive, particularly if their use has to be extended to ventilation ducts etc.), find a less aggressive and involatile alternative to fluoride (with presumably slower dissolution), or change the chemical nature of the fuel. The most likely of such changes is from oxide to nitride, with problems due to formation of ^{14}C unless the nitrogen is highly enriched in ^{15}N and recovered in near-quantitative yield to avoid unacceptable expense. The relative extraction properties of thorium and uranium require low solvent loading and larger equipment than in the Purex process for a given throughput, with less ready backwashing of the fissile ^{233}U and the possibility of a significant residue to enter the solvent wash system. In this respect, the relationship between fissile and fertile components is the opposite to that in the uranium-plutonium cycle, so that the safety case cannot be a direct extension of that for a Purex plant. Unless equipment can be limited to ever-safe geometry, consequent difficulties may arise, if not with respect to normal operation then in convincing licensing authorities that no credible maloperation could lead to a nuclear excursion. Current developments in intensified solvent-extraction contactors can be expected to ease the problems, particularly if the throughput is limited to serving only special-purpose facilities, but may not eliminate them completely.

Such problems would also be eased if only uranium and perhaps plutonium were to be recovered in the main line, and the bulk of the thorium discarded as waste. However, any subsequent separation of other actinides from the raffinate would then be complicated by the presence of this thorium, and of course the amount of waste would be greatly increased. These considerations apply directly only to aqueous reprocessing. The pyrochemical alternatives warrant closer investigation. Such methods were applied in the early days of interest in thorium, but the preferred chemistry has since changed. Calculations, said to have shown that the techniques now favoured for uranium and plutonium could be applicable, would need to be experimentally verified. The process as demonstrated operates in batch mode, which as a rule means substantially higher costs than in continuous operation; conversion to continuous mode is just conceivable but doubtfully practical, while scaling up to an industrial throughput may also be difficult.

3.5. Waste management

No programme of waste minimisation can totally eliminate the need for deep geological disposal, and forms suitable for thorium fuel residues must be found; they may well differ from those best for the uranium cycle, if only because of the additives necessary to make

aqueous processing possible. If a dry reprocessing route is adopted, based on halide salts rather than nitrates, then simple calcination will no longer convert the residues to insoluble oxides and a very different conditioning process will be needed, probably to a different waste form [7]. If long-lived fission products and minor actinides are to be recycled, as is one likely rationale behind the whole scenario, then the necessary processes will be generally similar to those in the uranium cycle and except perhaps for technetium do not need special attention here.

4. SUGGESTED DEVELOPMENT TOPICS

Outside areas of uranium scarcity, the most likely motives for using thorium fuels in the near future are to help consume transuranic elements, or to limit the reactivity swing during the course of irradiation. Neither course necessarily implies a closed cycle, and indeed the reason for the latter could be simply to extend the utilisation of fuel in once-through mode. Nevertheless, abandoning the terminal fissile content would be wasteful, and may come to be considered unacceptably so. Moreover, given the propensity for requirements to become increasingly stringent during the course of development, it would be imprudent to assume that the residues of minor actinides or fissile material would be slight enough to cause no further concern. Provision should therefore be made for reprocessing and recycling.

Accordingly, BNFL would favour devoting a greater proportion of effort to solving problems in the fuel cycle area, and together with European partners is preparing a joint proposal for such work under the Fifth Framework for Co-operation. Topics include thorium, supply, fuel design and manufacture, reprocessing and waste management.

Once components with substantial gamma and neutron emission have to be incorporated into fuel, remote fabrication will become obligatory. This will require mechanically simplified methods of fabrication, an extension of the capabilities and sophistication of robotics, or both.

Since a likely purpose of using thorium is to incinerate wastes, provision for their disposition in a reactor should be considered at this stage, particularly where (as with neptunium) it is likely to be homogeneous. The manufacture of heterogeneous targets is a separate issue.

Recycling of course requires reprocessing. Since non-aqueous routes are suggested to avoid the problems of the classic Thorex process, but are relatively undeveloped and would raise different questions of waste management, they should perhaps be given priority until an informed choice can be made between the alternatives. Whichever emerges as preferable (or both, since each may conceivably serve a particular niche) will need substantial efforts in commercial implementation, but that is a later phase that need not be considered until details of the chemistry are established.

5. CONCLUSIONS

Returning to the criteria initially stated, the difficulties that thorium fuels might alleviate are pressing only in certain areas; otherwise the problems have yet to become insistent, there are alternative acceptable solutions, or thorium does not present decisive advantages, while some difficulties in using it have not always been adequately recognised. Any general adoption of thorium is therefore likely to be on essentially political rather than commercial grounds.

Nevertheless, such grounds may become persuasive. In any case, major nuclear developments need a time-scale of decades, and lack of immediate need is no reason to wait until it becomes pressing before taking at least precautionary steps. Together with European partners, BNFL is therefore supporting a programme of work to clarify the most doubtful areas.

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EXPERIENCE OF THORIUM FUEL DEVELOPMENT IN INDIA*

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Abstract. India has one of the largest resources of thorium in the beach sands of Southern India. Generation of nuclear power through utilization of thorium is the ultimate goal of India's three stage nuclear power strategy. Bhabha Atomic Research Centre (BARC) is actively pursuing research, development, fabrication, characterization and irradiating testing of ThO_2, ThO_2-PuO_2, ThO_2-UO_2 fuels in test and power reactors. Work related to developing the fuel fabrication technology including automation and remotization needed for ^{233}U based fuels is in progress. Use of ThO_2 fuel bundles for initial flux flattening in our PHWRs; recent criticality of KAMINI - a small neutron source reactor, with ^{233}U-Al alloy fuel; introduction of ThO_2 as axial and radial blankets in our Fast Breeder Test Reactor (FBTR) at Kalpakkam; proposed ThO_2-PuO_2 and ThO_2-$^{233}UO_2$ fuel for Advanced Heavy Water Reactor (AHWR) are some of the steps taken by us towards utilization of Thorium in India. The paper summarizes the present status of thorium fuel development in India.

1. INTRODUCTION

India has a relatively modest uranium resource (~50,000 tons) but is endowed with one of the largest deposits of thorium in the world (~ 360,000 tons) in the beach sands of Southern India. Any long term planning of the growth of nuclear power programme in India, therefore, has to be based on proper harnessing of energy potential of thorium. This was realized quite early by the founders of our nuclear energy programme who drew up a clear three stage power development profile with the generation of nuclear power through utilization of thorium as its ultimate goal. The first phase of the programme is based on Pressurized Heavy Water Reactors (PHWR) using natural uranium as fuel. The second phase is based on utilization of plutonium, generated as by-product from the first phase, in Fast Breeder Reactors (FBRs) for power generation and to enhance our fissile material inventory both in terms of ^{239}Pu and ^{233}U. The third phase is based on thorium fuelled thermal reactors. Several theoretical studies have been carried out [1] on thorium fuel cycles in Heavy Water Reactors (HWR).

As thorium plays such an important role in our nuclear power programme, it is natural that we have significant R&D Programmes devoted to thorium fuel cycle development. We are actively pursuing research & development programme in fabrication, characterization and irradiation testing of ThO_2, ThO_2-PuO_2 and ThO_2-UO_2 fuels in our test and power reactors. Fuel bundles containing high density ThO_2 fuel pellets are being used in all our new PHWRs for flux flattening in the initial Core. ThO_2 pins and sub-assemblies are also to be used as axial and radial blankets in our Fast Breeder Test Reactor (FBTR) operating at Kalpakkam. KAMINI, a neutron source reactor, is operating with ^{233}U -Al alloy fuel. ThO_2-PuO_2 and ThO_2-$^{233}UO_2$ are proposed as fuel for the Advanced Heavy Water Reactor (AHWR), the detailed design of which is being carried out in our Centre. Development of novel fuel fabrication processes and techniques related to automation and remotization needed for ^{233}U based fuel fabrication are under study.

* 1998 meeting.

2. FUEL FABRICATION

2.1. ThO_2 Fuel

Fabrication of high density sintered ThO_2 pellets for the ThO_2 bundles used for flux flattening of the initial Core of PHWRs and as blanket in FBTR, is carried out by the conventional Powder Metallurgy technique of cold compaction and high temperature sintering either in reducing or in oxidizing atmosphere as shown in Figure 1.

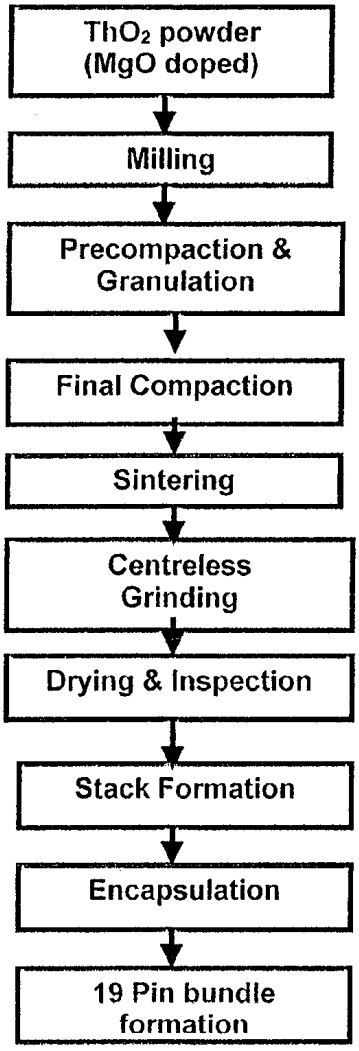

Figure 1. Flowsheet for ThO_2 fuel bundle fabrication.

The oxalate derived ThO_2 powder has a rectangular plate morphology and needs intensive milling to break the platelets and to increase the surface area and sinterability. To avoid caking of powder during milling, once-through dry nitrogen atmosphere is maintained in the enclosure around the pot mill/attritor. As the oxalate derived powder is not free- flowing, precompaction and granulation of powder is necessary to enhance flowability for ease of pneumatic powder/granule conveying and to obtain a uniform die fill during powder compaction.

ThO_2, being a perfectly stoichiometric compound with a high melting point (~3400°C), needs a sintering temperature of over 1800°C for obtaining high sintered density pellets (96% theoretical density). However, with the addition of 500-600 ppm of MgO dopant as sintering aid, high

density pellets are presently being fabricated on an industrial scale using a sintering temperature of about 1650°C-1680°C under reducing atmosphere. The improved sintering characteristics of MgO doped ThO_2 pellets is due to enhanced volume diffusion of thorium brought about by formation of oxygen ion vacancies as Mg^{+2} is substituted for Th^{+4}. MgO doping is done in the form of $MgSO_4$ in the thorium nitrate solution before oxalate precipitation.

A method of low temperature sintering (~1250°C) of ThO_2 pellets in air, using 0.5w% Nb_2O_5 as additive, has also been developed [2] and test irradiation of low temperature sintered ThO_2 pellets are being planned shortly.

Tonnage quantity of ThO_2 pellets are now a days fabricated using glove box trains with local shielding around equipment and interim storage facilities for powder/granule/pellets to reduce dose to operating personnel, aerosol generation and air borne activity.

2.2. ThO_2-PuO_2 and ThO_2-$^{233}UO_2$ Fuels

Thorium does not have any naturally occurring fissile isotope. However it can be converted to highly fissile ^{233}U by using it in reactors in combination with either ^{235}U or ^{239}Pu. Once sufficient quantity of ^{233}U is accumulated, ThO_2-^{233}U O_2 fuel cycle can be adopted in the PHWRs with near breeder characteristics.

It is in this context that knowledge of fabrication technology of (ThO_2-PuO_2) and (ThO_2-^{233}U O_2) fuels for advanced fuel cycles of PHWRs assumes great significance.

ThO_2 has similar crystal structure as that of UO_2 and PuO_2; forms complete solid solution at all percentages with UO_2 and PuO_2 and has similar physical properties. Hence, fabrication procedure of mixed oxide fuels of ThO_2 with PuO_2 or UO_2 are similar to that of (UO_2-PuO_2) mixed oxide fuel for which experience exists in India. However, fabrication of ^{233}U bearing fuels in standard glove-boxes is normally not feasible due to the presence of high gamma radiation field associated with the daughter products of ^{232}U, which is always present along with ^{233}U as a minor constituent. Hence, a high degree of automation and remotization and thickly shielded hot cell facility is needed for fabrication of ^{233}U based fuels.

In general, the following techniques have been tried for ThO_2 based fuel fabrication for PHWR:
(a) Cold pressing of powder mixture of ThO_2-PuO_2 or ThO_2-UO_2 followed by high temperature sintering.
(b) Vacuum impregnation of partially sintered low density (~70-80%T.D.) ThO_2 pellets with uranyl nitrate or plutonium nitrate solution followed by drying and final sintering.
(c) Sol-gel derived microsphere pelletization (SGMP) followed by sintering.

2.2.1. Cold Pressing and Sintering Route

This process is essentially the same as the process followed for the fabrication of ThO_2 fuel. The process consists of co-milling the ThO_2, PuO_2 or ThO_2 and UO_2 powders, cold pressing of the powder mixture into green pellets and sintering of the green pellets to get high density. This method needs handling of very fine and non-free- flowing powder and it generates highly radioactive dust which settles on the equipment and glove box/ hot cell surfaces necessitating their frequent decontamination in order to keep personnel exposures to a minimum. Moreover, this method of fuel fabrication is least amenable to automation and remotization. However, we have some experience of fuel fabrication by this method.

2.2.2. Impregnation Method

In this process (Figure 2), partially sintered, porous (density in the range of 70-80% T.D.) ThO_2 pellets, fabricated in a conventional ceramic fuel fabrication plant, are transferred to a shielded facility where the pellets are vacuum impregnated with Uranyl nitrate solution, dried and then sintered. The most attractive feature of this process is its amenability to automation and remotization and possibility of separating most of the equipment for pellet production from shielded facility where ^{233}U solution is handled.

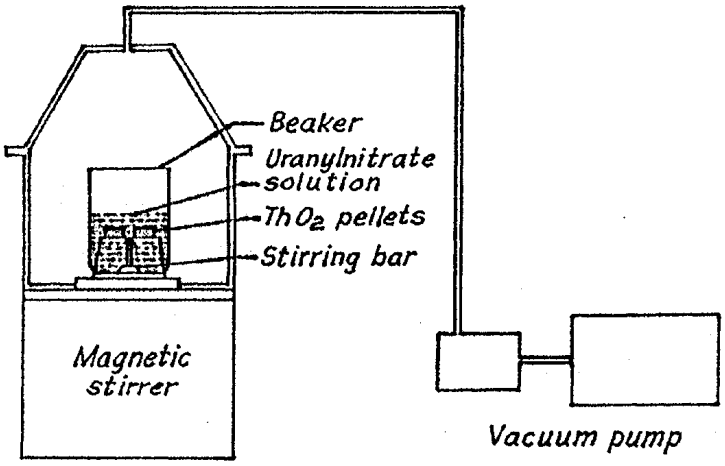

Figure 2. Schematic diagram of ThO_2 pellet impregnation set-up.

The limitations of this processes are:

(a) by this technique only 2-3w% ^{233}U can be introduced in ThO_2;
(b) multiple cycle of impregnation and drying are needed even to introduce 2-3w% uranium in ThO_2 as needed for PHWR pellets;
(c) it is very difficult to get a uniform distribution of UO_2 in ThO_2 over the whole cross section of the PHWR pellet.

In BARC, ThO_2-2w% nat.UO_2 pellets have been fabricated by this technique with homogeneous and uniform uranium distribution over the whole cross section of the pellet [3].

.2.3. Sol-Gel Microsphere Pelletization (SGMP) Process

Sol-gel microsphere pelletization process, popularly known as SGMP technique, utilizes sol-gel derived dust-free and free-flowing soft microspheres of (Th-U)O_2 [either by internal gelation process or by external gelation process], in the size range of 100-600 microns in diameter, which are cold compacted and sintered to high density pellets the same way powder pellets are fabricated. The general flow-sheet of fuel fabrication by SGMP is shown in Figure 3. Because of the free flowing and dust-free nature of the microspheres, the fuel fabrication process is amenable for automation and remotization needed for hands-off plant operation philosophy. SGMP process is being vigorously pursued in BARC for the fabrication of UO_2, UO2-PuO_2 and ThO_2-UO_2 fuel pellets.

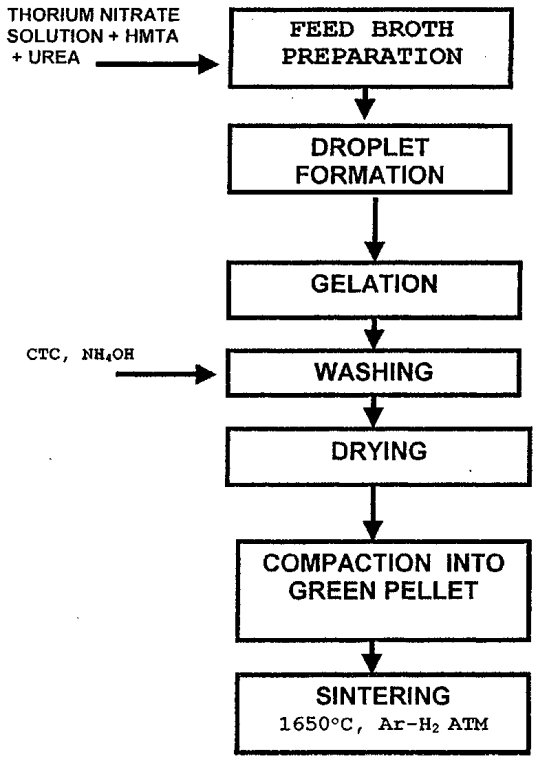

Figure 3. Flowsheet for fabrication of ThO_2 pellets by SGMP technique.

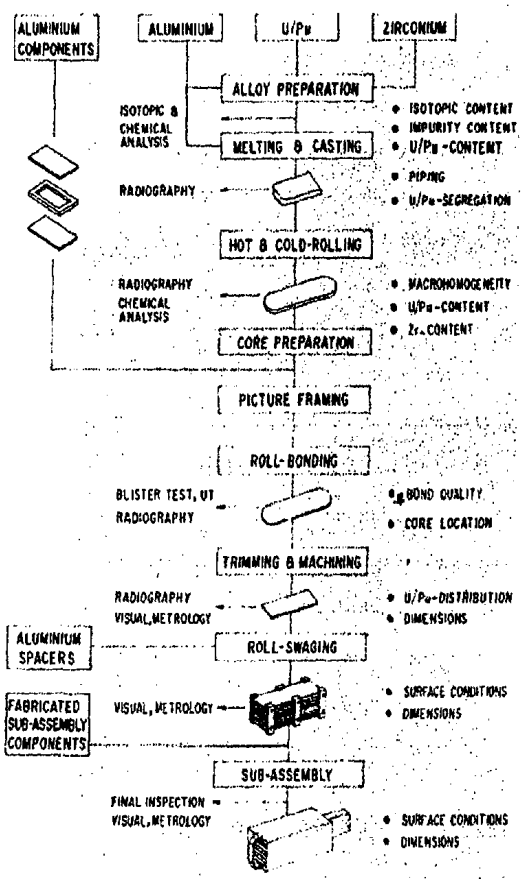

Figure 4. Flowsheet for KAMINI fuel fabrication.

3. FABRICATION OF Al-^{233}U PLATE FUEL

KAMINI, a 30 kW (thermal) light-water cooled and moderated, compact research reactor, is operating at our Indira Gandhi Centre for Atomic Research (IGCAR). BARC has gained substantial experience in fabrication of ^{233}U based fuel by supplying the Al-clad Al-20wt% ^{233}U plate type fuel assemblies for the reactor. The fabrication flow-sheet (Figure 4) consisted of preparing the master alloy using aluminium & uranium as feed materials, remelting and casting of the fuel alloy ingots, rolling, picture framing and sandwiching the fuel between thin aluminium sheets, roll-bonding, core location by radiography, trimming & machining to final dimensions. The detailed procedure of fuel fabrication has been described elsewhere [4].

4. THERMOPHYSICAL PROPERTY EVALUATION

A data base of thermal conductivity and hot hardness of ThO_2 fuel with temperature and PuO_2 and/or UO_2 content as variable is being generated to theoretically predict and model prediction of in-pile central temperature of these fuels and their performance. The general methods followed and description of the instruments used have been reported elsewhere [5, 6]. The results of thermal conductivity and hot hardness with temperature for ThO_2-2wt% UO_2 and ThO_2-4% PuO_2 pellets are reproduced here in Figures 5 & 6.

5. IRRADIATION TESTING OF (ThO_2 - PuO_2) ASSEMBLIES IN REACTORS

A six pin cluster consisting of (ThO_2-4%PuO_2) fuel pellets produced by powder pellet route has undergone irradiation testing in the pressurized water loop (PWL) of our research reactor CIRUS upto a burn up of 18.4 MWd/kg and is awaiting post irradiation examination. The loop test conditions, pellet details and irradiation data are shown in Tables I, II & III respectively. Two additional six pin clusters, containing high density ThO_2 and ThO_2-6.75% PuO_2 pellets clad in collapsible Zircaloy-2 tubes, (similar to our PHWR) are presently undergoing irradiation in the PWL - CIRUS and have accumulated a burn up of about 13 MWd/kg to-date. ThO_2 based fuel pellets fabricated by SGMP technique and low temperature sintering technique are being planned for future irradiation in our test loops.

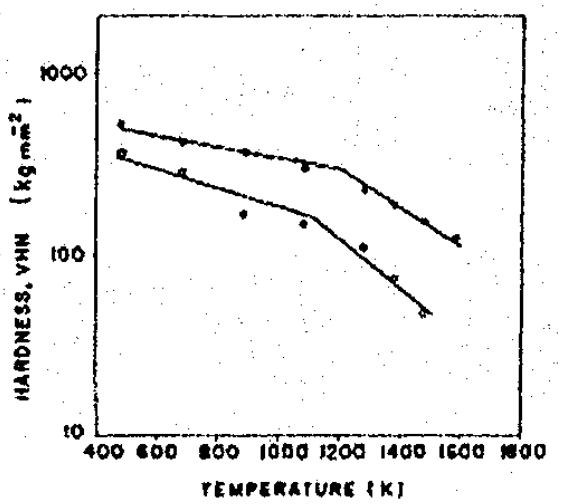

Figure 5. Hardness vs temperature plots for (•) ThO_2 - 2% UO_2 and (o) ThO_2 - 4% PuO_2 sintered pellets.

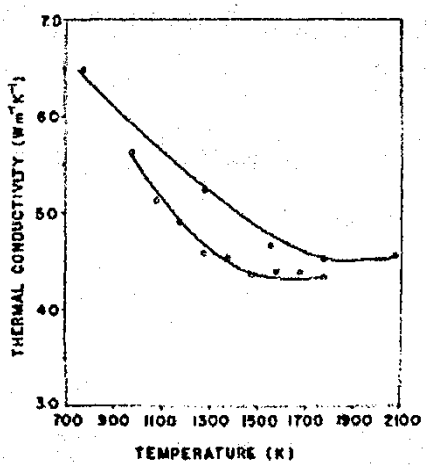

Figure 6. Thermal conductivity vs temperature for (•) ThO_2 - 2% UO_2 and (o) ThO_2 - 4% PuO_2 sintered pellet (corrected to 100% T.D.).

Table I. Loop Test Conditions

Test Section ID	57.4mm
Neutron Flux (Thermal)	5 x 10^{13} n/cm^2/sec
Coolant/pH	Demineralized Water/9.5-10.5
Coolant Flow Rate	16899 kg/h
Pressure	105 kg/cm^2
Temperature	204°C

Table II: Details of pellets for tests 1 & 2.

	Test 1	Test 2
Enrichment (PuO_2%)	4%	6.75%
Diameter	12.22 mm	14.4 mm
Density	92-94% T.D.	> 96% T.D.
Stack Length	435 mm	471.5 mm
Cladding Outer Diameter	14.3 mm	15.23 mm
Cladding Wall Thickness	0.8 mm	0.38 mm
Cold Plenum Length	20 mm	NIL

Table III: Irradiation data

	Test 1	Test 2
Peak Linear Power	385 w/cm	435 W/cm
Peak Burn-Up	18.4 MWd/kg	13.00 MWd/kg
Number of Power Cycles (> 30% Full Power)	100	177
Fuel Surface & Centre Temp.	462°C/1980°C	--

6. CONCLUSION

We have made a modest beginning in utilizing thorium for power generation and are planning for large scale utilization of thorium based fuels in future. Development of fabrication technologies, generation of data base for thermophysical properties and irradiation testing of thorium based fuels are being actively pursued and will be further intensified in future so that thorium can play an important role in the growth of nuclear power in India.

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SOME RESEARCH AND DEVELOPMENT OF THORIUM FUEL CYCLE IN RUSSIA*

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Abstract. There has recently been increasing interest in implementation of thorium into nuclear power. Among the new areas of that interest research in the field of nuclear fuel resources, safety improvement of nuclear reactors and their ecological acceptability should be mentioned. Many specialists consider the proliferation resistance to be the main advantage of thorium fuel cycle. In particular, this advantage leads to some proposals to use thorium-plutonium-based reactor fuel for plutonium utilization. This paper presents an overview of investigation, which are under way at the State Scientific Center - Institute of Physics and Power Engineering in Obninsk, Russia.

1. INTRODUCTION

Discussed in this paper are some researches on thorium fuel cycle which are always supported in IPPE at least a little. The authors do not intend to make a comprehensive overview of the problem but present their personal vision of the problem as well as the way how to solve it.

2. ^{232}U ISSUE

First experiments on ^{233}U accumulation in thorium irradiated in graphite-moderated reactor revealed some problems with ^{233}U handling. The decay of ^{232}U accumulated together with ^{233}U during irradiation leads to formation of some daughter isotopes, which are the source of intensive γ-radiation. ^{233}U can be worked with for a rather long time in glove boxes only if the presence of ^{233}U in thorium is less than 1 g/kg thorium, the nuclear content of ^{232}U being about 5 ppm in this case. If ^{232}U content is more than 10 ppm, either frequent extractions of decay products (approximately within 4 months) or installation of a remote control equipment is required for safe handling with irradiated materials.

At the initial stage of ^{233}U-Th fuel cycle implementation pure ^{233}U would be better to use for manufacturing samples and targets, experimental fuel pins and subassemblies. However huge amounts of ^{233}U "contaminated" by ^{232}U would appear. For example, ^{233}U with ^{232}U content of 2000-3000 ppm would be produced in a core of a conventional PWR with thorium for typical fuel burnups.

The production of pure ^{233}U can be started in fast reactors with rather favorable economics characteristics of nuclear power plant (NPP). Thus, in the thorium blanket of fast reactor the hard component of neutron spectrum is cut at the distance of 15-20 cm from the core, so ^{233}U accumulation in thorium of ~ 2.5 g/kg would correspond to the content of ^{232}U a few ppm. Compared to thermal reactors, utilization of thorium in fast reactors does not require a short irradiation time and special reloading schemes of thorium subassemblies.

* 1998 meeting.

For example, in the BN-800-type fast reactor with a thorium radial blanket it is possible to extract the following quantities of ^{233}U per year from different rows (kg): 82 (42 ppm of ^{232}U), 48 (11 ppm), 30 (3 ppm), 16 (0.7 ppm).

There have been some experiments on BN-350 fast reactor where the thorium samples were irradiated in the radial blanket up to ^{233}U accumulation in thorium of 1.3 g/kg. The ^{232}U content was in the range of 2-11 ppm depending on the position. It is worth noting here that the content of ^{232}U in thorium samples placed in the whole uranium blanket is higher than in the whole thorium blanket.

Among the experiments on ^{232}U problem we should mention here laboratory works on ^{233}Pa extraction from irradiated thorium. ^{233}Pa decay leads to formation of pure ^{233}U. These experiments require a quick delivery of irradiated thorium to the radio-chemical laboratory. IPPE possesses approximately 100 mg of ^{233}U extracted from irradiated thorium samples obtained as a result.

Some specialists think it's possible to make the isotope separation of ^{232}U from ^{233}U using a centrifuge process. Obviously, in case of a significant progress of works on thorium fuel cycle trade-off evaluations between the two fuel cycle technologies would be required: either to work with highly radioactive fuel with a special remote control equipment or to use rather simple equipment but with preliminary separated ^{232}U from ^{233}U.

3. THORIUM FUEL CYCLE TECHNOLOGIES

No special problems are expected in manufacturing technology of mixed oxide thorium-uranium (or plutonium) pelletelized fuel. High quality experimental fuel pellets of mixed (U-Th)O_2 were manufactured at both IPPE and other research laboratories.

Highly thermal-conductive dispersion fuel compositions characterized by lower temperature in fuel pins and lower accumulated internal heat are very attractive from the point of view of reactor safety.

Two types of fuel compositions for WWER-type reactors: UO_2 (60%)-Zr (40%) alloy and UO_2-Al alloy are under investigation and have successfully stood complex tests [1]. The authors suppose oxide UO_2 can be easily substituted by a mixture of UO_2-ThO_2. An other unconventional dispersion composition with pirolytic expanded graphite is under consideration [2]. Technological aspects of these compositions with uranium oxide have already been tested but more complex experiments including reprocessing are required.

The principal technology of ^{233}U extraction from irradiated in thermal reactor samples was tested in IPPE. The cooling period was approximately 3 years. Uranium water extraction method used resulted in 99.4% uranium extraction ratio. The fission product (FP) purification coefficient per one extraction cycle was more than 1000. In a separate reprocessing cycle thorium was extracted from the mixture of Th–FP with the extraction ratio of more than 99% and purification coefficient of 100.

Samples of ThO_2 irradiated in the radial blanket of BN-350 were used to investigate the dissolution process. The technologically accepted rate of ThO_2 dissolution was achieved when the fluoric acid was added to the nitric acid (0.1 mole/liter).

All these experimental results together with some reference data show the principal feasibility of the closed thorium fuel cycle based on water extraction.

Some proponents of dry reprocessing methods advocate the complex technology combining electrochemical reprocessing in salts resulted in oxide grains with subsequent vibropacked technology for fuel manufacturing.

4. SOME REACTOR CONCEPTS UTILIZING THORIUM FUEL CYCLE

4.1. Lightwater reactors with thorium.

As the initial stage of thorium implementation, IPPE proposed utilization of highly enriched uranium (HEU) resulted from weapon disarmament [3], which could help to avoid a special uranium enrichment required for thorium cycle initiation.

This proposition was considered for WWER-1000-type reactor with the dispersion fuel based on the metal thorium matrix.

The annual ^{235}U consumption due to ^{233}U breeding is lower for the considered reactor type compared to the conventional WWER-1000 with UO_2 fuel (735 and 940 kg per year, respectively). The number of consumed subassemblies is 1.8 times lower in the former case. ^{233}U is effectively bred (350 kg/year) instead of plutonium production (250 kg/year).

Safety characteristics of the thorium fuelled reactor such as feedback reactivities, burn-up reactivity swing and reactivity required for compensation during the heating are better. Also the value of energy accumulated in the fuel is lower in this kind of a reactor.

Reactors WWER with (Th-Pu)O_2 fuel have been under study due to the problem of weapon grade plutonium (WG-Pu) utilization [3,4]. As an example, WWER-1000 reactor with mixed oxide thorium and weapon grade plutonium subassemblies in 1/3 of the core was considered. The main reactor hardware was unchanged compared to the conventional WWER.

The annual consumption of WG-Pu in WWER-1000 with the above mentioned core is 355 kg while of the same reactor with 1/3 of the core MOX-fuelled VVER-1000 is only 270 kg. After burn-up of 40 MWt×days/kg, the amount of plutonium in subassemblies is 1/3 compared to the initial content. The content of ^{239}Pu in discharged subassemblies with (Th-Pu)O_2 is less than 30%. Thus, only one irradiation cycle in such a reactor is required for all loaded WG-Pu to lose its weapon grade quality completely.

Safety characteristics of the considered reactor are practically the same as for WWER with 1/3 MOX-fuelled subassemblies. The control rod system is unchanged.

Since reactors with (^{233}U-Th) fuel will be implemented only in the future, the core layout of this reactor is not necessarily be the same as in existing WWERs. For (^{233}U-Th) fuel cycle a balance should be found for ^{233}U breeding, safety specific power rating and fuel burn-up. Compared to the existing generation of PWRs, the inherent safety of the new generation of reactors should be higher.

The results of calculations showed the advantage of tighter fuel pin lattices with lower water volume ratio and at least two types of subassemblies in the core. These changes significantly

influence thermal-hydraulics parameters of a reactor, its reactivity characteristics and will require intensive efforts for design developments.

4.2. Mixed fuel cycle with ^{233}U and plutonium breeding in fast reactors

The idea of this cycle was proposed and developed in IPPE [5]. ^{233}U, plutonium and ^{238}U are used in the core of a fast reactor. Thorium is considered only as fertile material of a radial blanket. ^{233}U with ^{238}U is loaded into the inner core and plutonium with ^{238}U – into the outer core. As a result ^{233}U is bred only in blankets and plutonium - only in the core. The material for axial blankets should be chosen for any particular case.

Separated loading of ^{233}U and plutonium into the core gives some advantages:

- Having the same value of Doppler reactivity coefficient, sodium void effect has a significant negative component and as whole can be negative.
- The β_{eff} in the case of fertile ^{238}U is larger than that in the case of fertile thorium.
- The protactinium reactivity effect can be eliminated by thorium loading in blankets.
- The content of ^{232}U in ^{233}U is the lowest as compared with any other reactors.

Authors would like to attract the attention to this problem for further discussion.

4.3. U-233 for space reactors

The HEU-fuelled “TOPAZ” reactors with thermoionic direct conversion of energy have successfully been tested in space [6]. The thermoionic elements fuel inventory can be reduced in case of ^{233}U employment. This advantage can be used for increasing reliability of fuel elements with higher porosity and thicker cladding. As a result, the reactor lifetime can be significantly prolonged, which is a very important figure-of-merit for space reactors. Some preliminary results showed that 7-10 years of reactor lifetime could be achieved utilizing ^{233}U fuel.

However, the acceptable radiation condition at sites of fuel elements manufacturing, reactor assembling and launch is a provision for successful utilizing ^{233}U in space reactors.

Thermoionic elements manufacturing is a very complicated technology, absolutely excluding high radiation background. According to some evaluations, small contents of ^{232}U in ^{233}U (4-5 ppm) and a short technological cycle provide acceptable radiation conditions for the personnel. As is mentioned above, the production of ^{233}U with this content of ^{232}U is possible.

5. NUCLEAR DATA FOR THORIUM FUEL CYCLE

Increased interest to thorium fuel cycle has recently become the motivation for comprehensive nuclear data provision evaluations, similar to those existing for uranium-plutonium fuel cycle. These evaluations has been made at IPPE [7], where the detailed requirements for various cross sections measurement accuracy and comparisons with the obtained ones are presented. The fulfillment of the requirements will probably take a long time and obviously, the work activity will depend on the development of thorium cycle itself.

It should be mentioned that the present day works on thorium fuel cycle are supported either on the concept level or on the level of a particular problem and the existing nuclear data are

quite sufficient. Two examples show it. First, the uncertainty in evaluation of K_{eff} for systems with ^{233}U is higher (1.0%) than for plutonium-systems (0.5%) but is quite enough. Second, ^{232}U generation is evaluated nowadays with not less than 20% uncertainty. 10% uncertainty would be desirable but the values of thorium inelastic scattering and (γ, n) cross section on ^{231}Pa are to be studied better.

At present integral experiments support the studies of thorium cycle [8,9].

Thus, multiplication properties of media containing thorium, HEU and hydrogen were studied on COBRA critical installation at IPPE. Four assemblies with different composition central regions had K_∞ of these regions close to unity. The value of average absorption cross section of thorium was determined from the neutron balance conditions. The correction on heterogeneity is the main interpretation problem.

Two other assemblies of the same facility were used for the evaluation of critical parameters of conventional cores. The materials of the one were thorium and enriched uranium, the other one contained the same material and hydrogen. Preliminary results of this set of critical experiments showed 3% uncertainty in thorium absorption cross section.

The ratios of average cross sections to the fission cross section of ^{235}U were determined and the value of thorium absorption cross section was confirmed.

Various samples were irradiated in the core of BN-350 fast reactor. Many samples have not been investigated yet, but some of the results related to the thorium fuel cycle have already been obtained.

- The average uncertainty of 3% thorium absorption cross section is confirmed;
- The sum of (n, 2n) and (γ, n) cross sections for thorium with 5% experimental uncertainty corresponds to data usually taken;
- The data of ENDF/B-V library for (γ, n) cross section on ^{231}Pa are higher (50 ± 5%) compared to experimental results.

All this work is continued and the efforts will be increased depending on the financial support.

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A COMPETITIVE THORIUM FUEL CYCLE FOR PRESSURIZED WATER REACTORS OF CURRENT TECHNOLOGY*

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Abstract. Two important issues may influence the development and public acceptance of the nuclear power worldwide: a reduction of proliferation potential and spent fuel disposal requirements of the nuclear fuel cycle. Both problems may be addressed effectively by replacement of uranium by thorium fertile part of the fuel. A practical and competitive fuel design to satisfy the described design objectives and constraints may be achieved by seed-blanket core, proposed by A. Radkowsky and implemented in Shippingport reactors. The main idea is to separate spatially the uranium part of the core (seed) from the thorium part of the core (blanket), and thus allow two separate fuel management routes for uranium and thorium parts of the fuel. The uranium part (seed) is optimized to supply neutrons to the subcritical thorium blanket. The blanket is designed to generate and bum in-situ ^{233}U.

1. Introduction

It was noted at an early stage of the nuclear technology development that ^{233}U presents a superior fissile nuclide producing more neutrons per neutron absorbed than all other fissile isotopes. This feature, and the fact that thorium is much more abundant as a natural ore than uranium, prompted numerous attempts to design and implement a nuclear reactor based on thorium fuel. The most notable examples are the Light Water Breeder Reactor (LWBR) and early High Temperature Gas-Cooled Reactor (HTGR).

The main challenge encountered in the design of a thorium based system is the necessity to supplement natural thorium with a pre-generated fissile component. Several design solutions were proposed and investigated, such as: initial start-up of the thorium cycle by enriched uranium, continuous addition of uranium as a fissile component to supplement self-generated ^{233}U, reprocessing and recycling ^{233}U, and addition of plutonium to supplement self-generated ^{233}U.

The improvement in natural uranium utilization by using thorium could be achieved only if the self-generated ^{233}U fissile material was separated and recycled into a closed fuel cycle. This approach, adopted by the LWBR, violated the non-proliferation requirement.

An efficient utilization of thorium in a once-through cycle encounters a "neutron economy" problem: the ^{233}U build-up process is quite slow (compared with the plutonium build-up of the uranium chain), reaching saturation at a burnup of about 40 GW·d/t. Under irradiation thorium undergoes a rapid increase in ^{233}U concentration (and K_∞) and retains a value higher than that of plutonium in uranium lattice. Because of the very large fission cross section of plutonium, uranium builds up plutonium early in life, but this quantity saturates and declines

* 1998 meeting.

much sooner than in the case of ^{233}U in thorium. This difference results from the fact that the equilibrium concentration of plutonium is low, by virtue of its high cross section and because of the high capture to fission ratio, α. For thermal neutrons, $\alpha(^{233}U) = 0102$ and $\alpha(^{239}U) = 0.339$.

During the long build-up the subcritical thorium part of the fuel requires a continuous "investment" of neutrons created by fissioning ^{235}U, i.e. a large initial resource investment in uranium. In order to "recover" this investment in terms of fuel utilization gains by an taking advantage of superior ^{233}U properties, the thorium based fuel should be burned further, up to a burnup of at least 70-80 GW·d/t, corresponding to 8-9 full power years. Thus, the main challenge of an efficient utilization of thorium in LWR's is reduced to a problem of achieving a very large accumulated burnup of the thorium in a once-through fuel cycle.

It should be noted that, similarly to plutonium created by the transmutation of ^{238}U (fertile) isotope, another fissile isotope (^{233}U) is created by transmutation of ^{232}Th. While, pure ^{233}U is by itself an efficient fissile material and therefore a diversion risk, it may be easily denatured (neutralized) by an addition of a relatively small amount of natural uranium.

The SBU geometry provides the necessary flexibility to satisfy a major design constraint – full compatibility with existing pressurized water reactor (PWR) power plants. In addition, the heterogeneity of the SBU design allows the necessary (and separate) optimization of seed and blanket lattices. Additional U isotopes created within the thorium transmutation chain, such as ^{232}U, ^{234}U, and ^{236}U present major natural barriers to a diversion of ^{233}U isotope.

Physical properties of thorium and thorium-chain isotopes may be instrumental in solving the problem of incineration of excess fissile material originated from dismantled nuclear weapons. Both, plutonium and highly enriched uranium (HEU) may be used as a fissile component of a thorium -based fuel.

Advantageous properties and design challenges of the thorium -based fuel cycle may be addressed by introducing a heterogeneous fuel assembly design. This, seed-blanket (SBU), fuel assembly geometry allows separation of the uranium (or plutonium) part of the fuel (seed) from the thorium part of the fuel (blanket). This paper presents two fuel cycle options for a thorium-based heterogeneous fuel assembly design: the non-proliferative thorium fuel cycle and the plutonium-incineration thorium fuel cycle.

2. Heterogeneous thorium reactor core

The Seed-Blanket concept proposed by Professor A. Radkowsky offers a solution to the thorium utilization problem. The fuel assembly/fuel cycle design (1) based on this concept is designated as Radkowsky Thorium Fuel (RTF). The basic idea is to use the heterogeneous, seed/blanket (SBU), fuel assembly. The thorium part of the fuel assembly is separated from the uranium part of the assembly. This separation allows separate fuel management schemes for the thorium part of the fuel (a subcritical "blanket") and the "driving" part of the core (a supercritical "seed"). The design objective of the blanket is an efficient generation and an in-situ fissioning of the ^{233}U isotope, while the design objective of the seed is to supply neutrons to the blanket in a most economic way, i.e. with a minimal investment of natural uranium.

For the plutonium incineration option of the RTF design, enriched uranium is replaced by plutonium. The seed fuel is composed of Zr/Pu alloy, where the plutonium content is selected

to provide a given inter-refuelling interval. The blanket fuel is composed mainly of the thorium oxide with an addition of the plutonium oxide introduced to assure an acceptable power share (and a correspondingly power density) of the blanket during the initial period of ^{233}U buildup. During the first 300-400 full power days, the initial plutonium loaded into the blanket fuel is burned out, simultaneously with a buildup of an amount of ^{233}U, sufficient to sustain a reasonable criticality and an acceptable power density.

A similar approach is adopted for the non-proliferative design option: the blanket fuel contains a small amount of enriched uranium, which sustains an adequate blanket criticality during the initial period of ^{233}U buildup. It should be noted that natural uranium may be added to a thorium plutonium oxide mixture in the plutonium-incinerator option, in order to assure that the fissile content of the uranium part of the discharged blanket fuel will not exceed 15%. Thus, the discharged blanket fuel may be considered as non-proliferative in accordance with accepted international standards.

3. Fuel management approach

The fuel utilization efficiency of the once-through thorium-based fuel cycles may be achieved by taking an advantage of the superior properties of ^{233}U as a fissile component of the fuel. The thorium part of the fuel (blanket) should accumulate a burnup of at least 80 MW·d/kg, as explained in the previous section. This high value may be achieved due to a better irradiation resistance of the ThO_2-based fuel in comparison with the UO_2-based fuel. Correspondingly, the in-core residence time of the seed and blanket parts should be different, as well as the in-core fuel management schemes.

The seed-blanket fuel assembly is designed to allow a separate fuel management handling of the blanket and seed sub-assemblies. A blanket sub-assembly is loaded at the core beginning of life (BOL) and is kept in the core for 8-10 annual cycles, while a seed sub-assembly is kept in the core for 3 annual cycles, and is reshuffled in a manner similar to the light water reactor approach. At each cycle a third of all seeds in the core are replaced by fresh seeds, while the remaining two-thirds of the partially depleted seeds are reshuffled to maintain an acceptable power density distribution.

The blanket loaded at the beginning of the first cycle (BOC1) remains in the core for about 10 seed cycles, accumulating a burnup high enough to assure fuel utilization efficiency. In general, the blanket residence time depends on the blanket criticality value, and should be optimized in accordance with the chosen design objective. It should be noted, that for the non-proliferative design option (enriched uranium in seed), the fuel cycle economics is optimized, while for the plutonium incinerator option, the rate of plutonium destruction is optimized under a constraint of an economic feasibility. A detailed optimization study is beyond the scope of the present paper and is neglected at this stage.

The in-core fuel management scheme described above is a key feature of the concept presented in this paper. Design objectives of the fuel options considered are:

- a high rate of excess plutonium disposition for the plutonium -incinerator cycle, and
- a high proliferation resistance and reduction in the waste disposal requirements for the non-proliferative cycle.

These objectives were achieved with an efficient thorium utilization and an acceptable fuel cycle economics.

4. Design Description

A typical PWR core of Westinghouse design (4 loop, 3,400 MW(th) output, 193 assemblies) was chosen as a reference design basis. The heterogeneous fuel assembly approach is implemented by dividing the fuel assembly into two spatial regions: an inner (central) region - seed, and an outer (annular) region - blanket. The seed region includes a super-critical fuel part, and is composed of an enriched uranium or plutonium in a metal alloy with a zirconium matrix. The blanket region includes a sub-critical fuel part and is composed of the ThO_2 ceramic fuel, spiked by a small amount of an enriched uranium or plutonium.

The mechanical design of the assembly is adjusted to accommodate a separate handling of the seed and blanket sub-assemblies as described in the previous section. The grid plates, as well the support plates of the core structure, are modified to allow removal and insertion of a seed subassembly into a corresponding blanket sub-assembly. A schematic outlay of the seed-blanket unit (SBU) is shown in Figure 1.

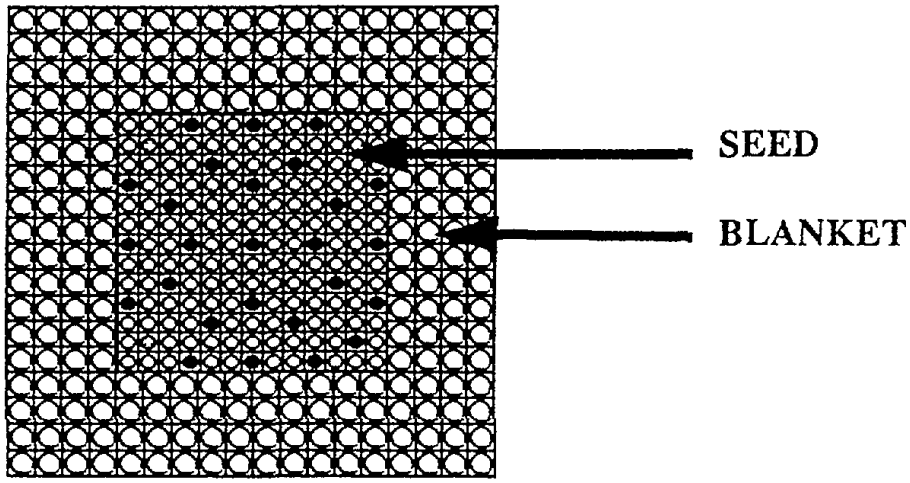


Figure 1. Fuel assembly (SBU) geometry.

Several comments are due to clarify the assembly outlay shown above:
- fuel rods lattices of the seed and blanket regions are optimized separately, and are therefore different,
- the spatial division of the SBU into two regions (i.e. relative seed and blanket volumes) is optimized according to a given design objective and is constrained by a maximum allowable power density in the seed region,
- the guide tubes (for control rods and burnable poison rods) are concentrated in the seed region (seed solid circles in Figure 1).

5. The Non-proliferative cycle design option

The design objectives and constraints applied to the Non-proliferative thorium fuel design are summarized below:

1. The RTR concept should be realized as a new fuel design, and thus, be completely compatible with existing power plants. Only minor plant hardware modifications, directly related to a modified fuel assembly internal arrangement are allowed.

2. All safety and operational parameters of existing power plants will be preserved.
3. The fuel design will be based mainly on an existing (not necessarily commercial) fuel technology. The maximum allowable fuel enrichment will be kept below 20% of the ^{235}U content.

A reference core and assembly design parameters are summarized in Table I.

The heterogeneous neutronic design of the core results in a highly supercritical seed and a subcritical blanket, which in turn leads to a relatively high power density in the seed. In order to satisfy thermal-hydraulic constraints, the seed fuel material in the present design is a metallic U/Zr alloy. The heat transfer coefficients of such an alloy are about an order of magnitude higher than those of an oxide fuel. Therefore, the maximum seed fuel temperature achieved is about 500°C, which is consistent with acceptable thermal margins for a metal fuel.

Table I. Core and fuel assembly parameters (Th - U cycle)

parameter	value
Total Power, MW(th)	3,400
Number of assemblies (SBU's)	193
Seed/Blanket Volume Fractions	0.4/0.6
Seed V_m/V_f	3.0
Blanket V_m/V_f	1.8
Seed Fuel	U/Zr alloy, U volume content ~ 20%, U enrichment=20
Blanket Fuel	$(Th+U)O_2$, U volume content ~ 10%, U enrichment=20
Seed Fuel Weight, kg H.M.	10,000
Blanket Fuel Weight, kg H.M.	40,000
In-core fuel management	3-batch seed scheme, inter-refueling interval = 300 FPD

The inter-refueling interval of 300 full power days was obtained by adjusting the enriched uranium content of each seed reload to sustain a required criticality during this period. The blanket reactivity (negative) is changing with burnup at each seed cycle, which in turn leads to a variation in the seed fissile load. The most significant phenomenon effecting blanket criticality is the ^{233}U buildup. Total weight of ^{233}U accumulated in the blanket at end of each cycle is shown in Table II.

Table II. ^{233}U accumulated weight (blanket)

cycle #	1	2	3	4	5	6	7	8	9	10
^{233}U weight (kg)	408	602	756	805	837	850	860	872	875	875

The ^{233}U buildup is demonstrated: an equilibrium density is reached near EOC5 and remains almost constant for the following 5 cycles. A subcritical blanket is sustained by neutrons generated in a supercritical seed. A mass balance summary is shown in Table III.

The total amount of uranium charged into a core during 10 seed cycles is 40,350 kg. Following the first 10 cycles all blanket sub-assemblies are replaced by a fresh reload, and seed replacement cycles are continued at the same refueling rate. An averaged uranium reload per cycle is estimated as 2,950 kg of the 20% enriched per year. The discharged plutonium (an annual discharge rate of approximately 33 kg) contains a plutonium composition vector, which is quite different from the typical LWR discharged plutonium, as shown in Table IV.

The RTF discharged plutonium is of a "low quality", namely isotopes, which are non-fissile in a thermal lattice, constitute almost 40% of the total. Thus, the discharged plutonium (especially from the blanket part of the fuel) is not reusable as a reactor fuel, removing the incentive of reprocessing.

Table III. Fuel mass flow summary (weight in kg H.M)

cycle #	SEED			BLANKE			
	Charged (enriched U)	Discharged (total H.M)	Discharged Pu	Charged Th	Charged (enriched U)	Discharged (total H.M)	Discharged Pu
1	8,300	2,950	13	45,000	5,500	-	-
2	3.900	2,950	20	-	-	-	-
3-9	3,900	2,950	37	-	-	-	-
10	3,900	2,950	37	-	-	50,500	152

Table IV. Plutonium isotopic vector

Pu isotope	RTFdischarged Pu		PWR discharged Pu
	Seed	Blanket	
Pu-238	0.054	0.116	0.010
Pu-239	0.486	0.400	0.590
Pu-240	0.243	0.150	0.210
Pu-241	0.135	0.150	0.140
Pu-242	0.082	0.184	0.050

The balance of fissile isotopes production (^{233}U in blanket) and destruction (plutonium in seed) determine corresponding multiplication properties of seed and blanket sub-assemblies and their respective relative powers. The blanket multiplication factor dependence on an accumulated burnup is demonstrated in Fig. 3, and the corresponding power share in Fig. 4. The apparent discontinuity of the criticality curve may be attributed to the fact that each blanket sub-assembly is depleted in a "piece-wise" manner, i.e. within a single blanket a seed sub-assembly is replaced/reshuffled each cycle. The spectral shift caused by replacement of a depleted seed by a fresh seed results in a corresponding blanket criticality shift.

5.1. Reactivity control

The feasibility of the reactivity control of the proposed fuel design is evaluated by calculating the reactivity worth values for different control methods, such as control rods and soluble boron. These values are compared for a standard uranium based PWR (PWR) and the SBU fuel design, designated as RTF-U. All values summarized in Table V below were obtained for a lattice (assembly) calculational level. Thus, these values do not represent the result of a detailed core analysis of the final design and are considered only for a comparison. An additional parameter of interest presented in Table V is the moderator temperature coefficient (MTC). All data presented below was calculated for BOC, full power conditions.

Table V. Reactivity worth[a] and MTC summary

Fuel Lattice Type	$\Delta\rho$ per ppm	$\Delta\rho$[b] CR	MTC $\Delta\rho$/°C
PWR	0.86×10^{-4}	0.576	0.22×10^{-4}
RTF-U	0.91×10^{-4}	0.412	0.21×10^{-4}

a. - all reactivity worth values are negative.
b. - standard PWR control rod (Ag-In-Cd), $\Delta\rho = \rho$ (CR in) - ρ (CR out).

A comparison of the reactivity worth values for the PWR lattice with those for the RTF fuel lattice demonstrate a reduction in the total reactivity worth of the control rods and a slight increase in the reactivity worth of the soluble boron. A combination of these reactivity control methods with an extensive utilization of Burnable Poisons seems to be adequate to satisfy reactivity control requirements of the RTF-based PWR core.

5.2. Spent fuel storage

Spent fuel storage and disposal requirements are derived from the following characteristics of the spent fuel stockpile: 1) mass and volume, 2) radioactivity level, 3) thermal power level, and 4) toxicity. A detailed evaluation of the latter three parameters is beyond the scope of this paper. Nevertheless, the mass and volume of annual discharge of the RTF fuel cycle may be evaluated on the basis of the data presented in Table III, and compared in Table VI with the corresponding values for a standard PWR cycle.

The comparison presented in Table VI indicates a large reduction in the discharged fuel mass (about 70 percent) and volume (about 50 percent). This reduction indicates a potential of a significant reduction in the overall spent fuel storage requirements and associated costs.

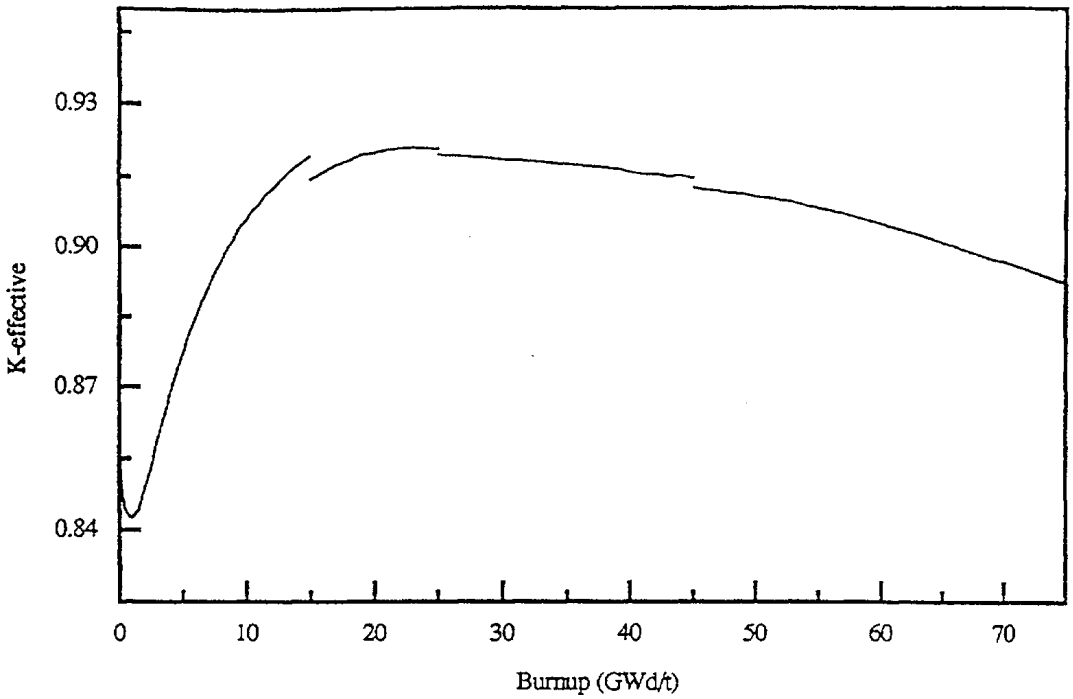


Figure 2. Blanket multiplication factor as a function of burnup for 10 cycles

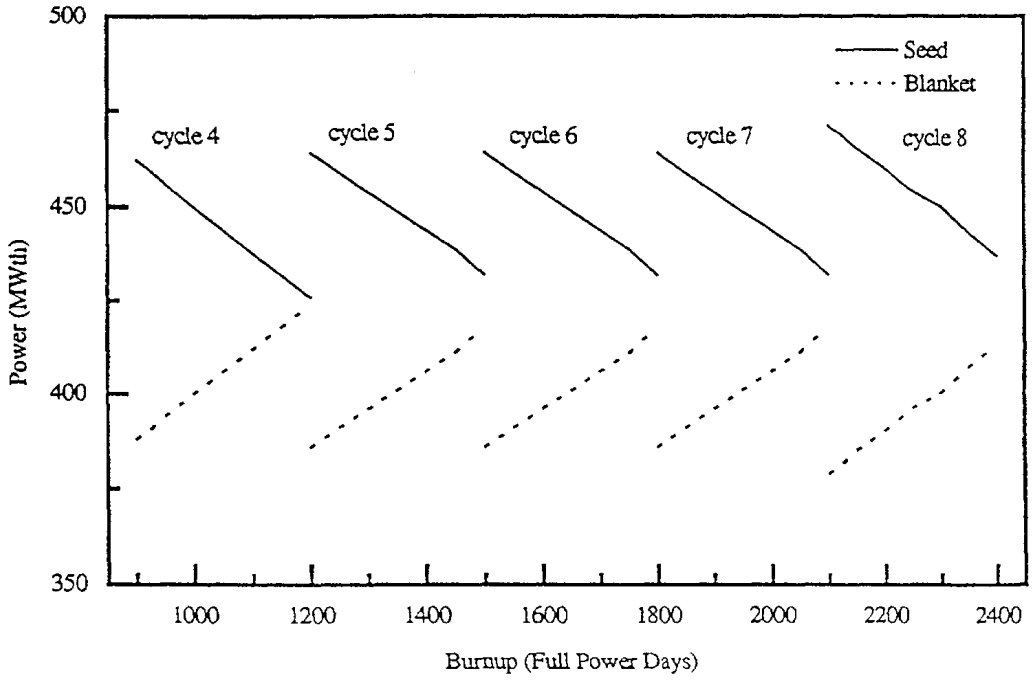


Figure 3. Seed and blanket power share as a function of burnup for selected cycles.

5.3. Proliferation resistance of the RTF cycle

One of the main concerns related to nuclear power industry is the potential of diverting the fissile component of the discharged fuel for production of weapons. To assemble a nuclear explosive device, one needs a certain amount of fissile material. The materials of interest are enriched ^{235}U, plutonium and ^{233}U [1]. The quality of the material is very important for the construction of an explosive device.

Table VI. Annual spent fuel discharge

parameter	PWR[b]	RTF[a]		
		seed	blanket[c]	total
total H.M. weight (MT)	28'	3.9	5.0	8.9
total fuel volume (m^3)	10	4.0	0.70	4.70

a. - RTF parameters evaluated for a preliminary core design.
b. - PWR parameters for a typical 3-batch fuel management scheme.
c. - an equivalent annual value for a blanket discharged every 10 seed cycles.

The lowest fissile content for the construction of a nuclear weapon is the (somewhat arbitrary) value of 20%. This value is adopted by international organizations as a threshold. Similarly, the quality of plutonium affects the ease of construction and the efficiency of a plutonium bomb. Consideration of the plutonium composition of the RTF spent fuel stockpile and its comparison with the weapon grade and PWR reactor grade material indicate a significantly increased proliferation resistance:

- The total amount of plutonium produced annually is reduced by a factor of 6 to 7,
  The isotopic composition of the seed plutonium, and especially blanket plutonium, requires a significant increase of the critical mass,
- An increased content of the ^{240}Pu and ^{242}Pu increase the spontaneous fission rate of the RTR plutonium mixture and cause significant yield degradation of the weapon device based on plutonium diverted from the RTR fuel cycle,
- A higher content of ^{238}Pu increase thermal power production of the plutonium mixture, which presents a serious obstacle to building a stable and reliable explosion device.

The ^{233}U created in blanket was denatured by the ^{238}U, which was added to thorium. The amount of uranium added for dilution of fissile components was carefully chosen to reduce the overall content of fissile uranium isotopes below 20%. In principle, all uranium isotopes may be chemically separated from the blanket spent fuel and be further enriched by standard industrial methods. However, there are two major barriers to this diversion path provided by the RTR fuel cycle design:

- The contamination of the recycle material by a hard gamma-emitter (^{208}Tl), originating in the ^{233}U chain, will require that the reprocessing facility be remotely operated.
- The necessity of the additional enrichment of the mixture of uranium isotopes will be extremely inefficient due to its isotopic composition. An attempt to separate ^{233}U from ^{238}U, ^{236}U and ^{234}U isotopes will also remove the fissile ^{235}U from the resulting enriched stream. In addition, the separation process involved in enriching the mixture of all uranium isotopes will also require a remote operation.
- In conclusion, the RTF cycle provides an inherently elevated proliferation resistance in comparison with a standard LWR cycle of current technology. The comparative analysis shows that RTF spent fuel stockpile will produce significantly reduced amounts of fissile material, the produced material will be more resistant to separation and diversion and of a significantly lower weapon grade quality.

6. The plutonium incinerator design option

The design objective of the SBU plutonium-incinerator option is to achieve an efficient destruction of the excess plutonium in light water reactors of current technology through an economically competitive fuel cycle. The seed fuel of the Non-proliferative option is replaced by Pu/Zr metal alloy, and the blanket fuel is composed of thorium oxide spiked by plutonium oxide to provide blanket criticality and power density during the initial period of ^{233}U buildup.

The core, assembly and cycle designs described in this section are not optimized for the maximum plutonium destruction rate, but rather to demonstrate the capability of the thorium cycle, based on a heterogeneous fuel assembly, to reduce the plutonium inventory in an economically competitive manner. The blanket in-core residence time was chosen to achieve an accumulated burnup of 80 - 100 MW·d/kg, i.e. a value compatible with that of the Non-proliferative version. Clearly, economic advantages of using thorium based fuel for plutonium incineration are predicated by a high burnup of the thorium part of the fuel. Such an approach allows a consistent comparison of the proposed design with alternative reactors/fuel cycles. A reference core and assembly design parameters are summarized in Table VII.

Table VII. Core and fuel assembly parameters (Th - Pu cycle)

parameter	value
Total Power. MW(th)	3,400
No. of assemblies (SBU's)	193
Seed/Blanket Volume Fractions	-0.5/0.5
Seed V_m/Vf	-3.0
Blanket Vm/Vf	-1.8
Seed Fuel	Pu/Zr alloy, U volume content ~ 20%, U
Blanket Fuel	(Th+Pu)02, U volume content ~ 10%, U
Seed Fuel Weight, kg H.M.	-2,600
Blanket Fuel Weight, kg H.M.	-48,000
In-core fuel management	3-batch seed scheme, inter-refueling interval = 300 FPD

The inter-refueling interval of 300 full power days was obtained by adjusting the plutonium content of each seed reload to sustain a required criticality during this period. The blanket reactivity (negative) is changing with burnup at each seed cycle, which in turn leads to a variation in the seed fissile load. The most significant phenomenon effecting blanket criticality is the ^{233}U buildup. Total weight of ^{233}U accumulated in blanket at end of each cycle is shown in Table VIII. The ^{233}U buildup is demonstrated: an equilibrium density is reached near EOC5 and remains almost constant for the following 5 cycles. A subcritical blanket is

sustained by neutrons generated in a supercritical seed. A schematic mass balance summary of the plutonium flow is shown in Fig 4. The amounts of plutonium charged and discharged from the core are presented. The plutonium core inventory at the BOC and EOC time points are also shown and allow an estimate of the plutonium destruction rate. The blanket multiplication factor dependence on an accumulated burnup is demonstrated in Fig. 5 and seed/blanket power distribution in Fig. 6.

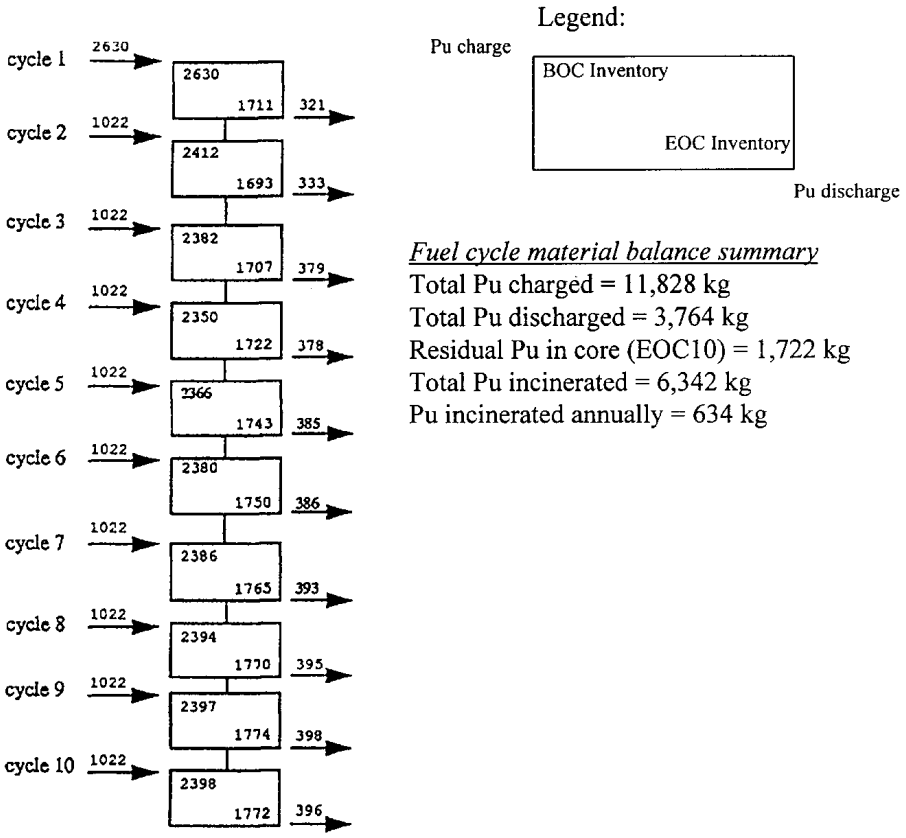


Figure 4. Plutonium mass flow summary for 10 cycles.

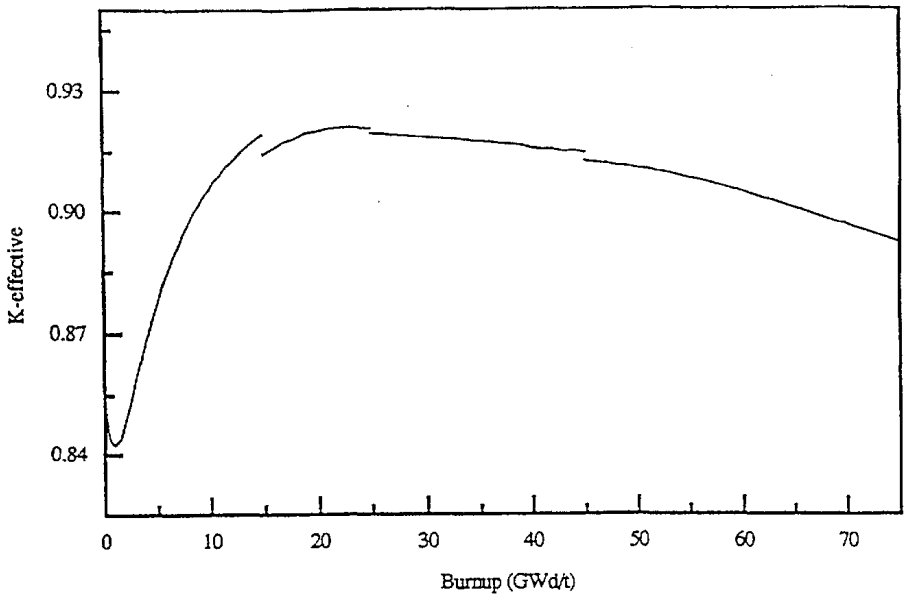
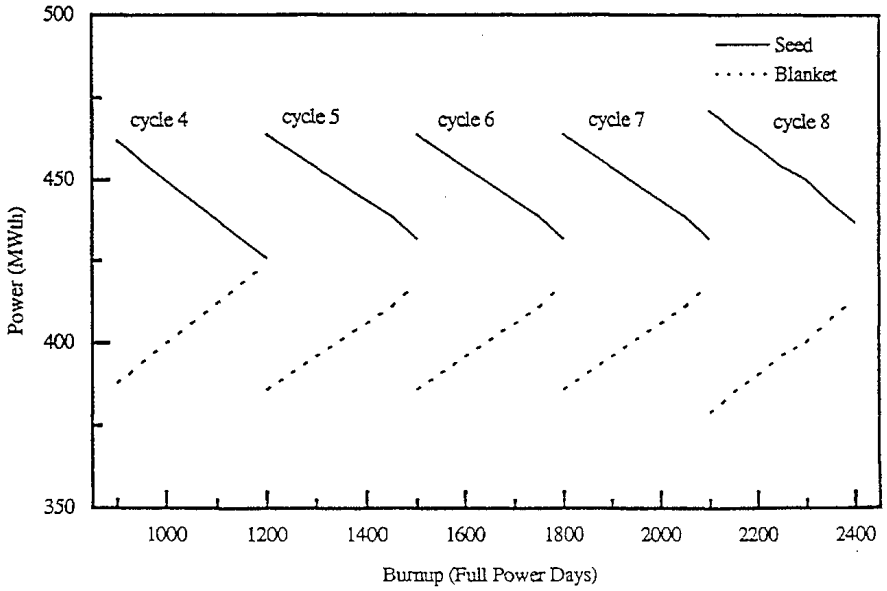


Fig. 5 Blanket multiplication factor as a function of burnup for 10 cycles



Fig, 6 Seed and blanket power share as a function of burnup for selected cycles

Table VIII. ^{233}U accumulated weight (blanket)

cycle #-	1	2	3	4	5	6	7	8	9	10
^{233}U weight (kg)	348	541	651	687	705	713	716	715	714	712

Table IX. Plutonium isotopic vector

Pu isotope	Charged	Discharged
Pu-238	0.0005	0.0025
Pu-239	0.9360	0.3542
Pu-240	0.0590	0.4207
Pu-241	. 0.040	0.1643
Pu-242	0.0005	0.0583

The total amount of plutonium charged into a core during 10 seed cycles is 11,828 kg. Following the first 10 cycles all blanket sub-assemblies are replaced by a fresh fuel, and seed replacement cycles are continued ar the same refueling rate. An averaged plutonium incineration rate is estimated as 634 kg of the weapon grade plutonium per year. The discharged plutonium (annual discharge rate of approximately 376 kg) contains a plutonium composition vector completely different from the initial one, as shown in Table IX. The total weapon grade plutonium incineration rate is estimated as 1,183 kg/ year.

The present cycle design may be easily modified by reducing the blanket in-core residence time. Such a modification would results in an increased plutonium incineration rate and a reduced thorium fuel utilization efficiency. The cycle design and fuel management scheme may be finalized by adopting specific design objectives and constraints.

The discharged plutonium is of a "low quality", namely isotopes, which are non-fissile in a thermal lattice, constitute almost 50% of the total. Thus, the discharged plutonium is not reusable as a reactor fuel, removing the incentive of reprocessing.

6.1. Reactivity control

The feasibility of a reactivity control of the proposed fuel design is evaluated by calculating the reactivity worth values for different control methods, such as control rods and soluble boron. These values are compared with corresponding values for different fuel concepts [2]: a standard uranium based PWR (designated PWR), a full core mixed oxide, namely natural uranium with weapon grade plutonium (designated MOX), and a homogeneous assembly of thorium oxide mixed with weapon grade plutonium (designated TMOX). The SBU fuel design is designated as RTF-Pu.

It should be emphasized that all values summarized in Table X below were obtained for a lattice (assembly) calculational level or for a preliminary design of a core. Thus, these values do not represent the result of a detailed analysis of the final design and are relevant for a comparison only. An additional parameter of interest presented in Table X is the moderator temperature coefficient (MTC). All the data presented below was calculated for BOC, full power conditions.

Table X. Reactivity worth[a] and MTC summary.

Fuel Lattice Type	$\Delta\rho$ per ppm	$\Delta\rho$[b] per CR	MTC $\Delta\rho$/°C
PWR	0.86×10^{-4}	0.576	0.22×10^{-4}
MOX	0.35×10^{-4}	0.262	0.32×10^{-4}
TMOX	0.44×10^{-4}	0.303	0.22×10^{-4}

Fuel Lattice Type	$\Delta\rho$ per ppm	$\Delta\rho$[b] per CR	MTC $\Delta\rho$/°C
RTF-Pu	0.60×10^{-4}	0.330	0.21×10^{-4}

a. - all reactivity worth values are negative

b. - standard PWR control rod - (Ag-In-Cd), $\Delta\rho = \rho$ (CR in) - ρ (CR out

Table X data demonstrates the well known effect of a reduction of the reactivity worth of control absorbers due to the presence of plutonium. It is also shown that the heterogeneous design improves the control absorbers performance and alleviates, to a certain extent, the reactivity control problem of the plutonium-based lattices.

7. Summary

The heterogeneous fuel assembly design (seed-blanket) offers a solution to a problem of an efficient thorium utilization in LWR's of current technology. The SBU design allows a

separate lattice optimization for the fissile part of the fuel (seed) and the fertile part of the fuel (blanket), as well as an implementation of separate in-core fuel management schemes. This flexibility is shown to be a major advantage in achieving the design objectives and satisfying the design constraints.

Two fuel cycle opftions are considered in this paper: the Non-proliferative thorium-based cycle, and the plutonium-incinerator thorium-based cycle. Both cycle options were applied to a typical PWR core, were analysed and demonstrated a potential for an efficient and a competitive thorium-based fuel, aimed to improve an overall proliferation resistance of the fuel cycle and to reduce the spent fuel storage requirements.

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NUCLEAR ENERGY RESEARCH INITIATIVE: THORIUM FUEL CYCLE PROJECTS*

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Abstract. The United States (U.S.) Department of Energy (DOE) is conducting four projects involving the use of the thorium fuel cycle. All four projects are based on an once-through, proliferation resistant, high burnup, long refueling cycle use of thorium in a light water reactor. Three of these projects are part of the Nuclear Energy Research Initiative (NERI) program. These are: "Advanced Proliferation Resistant, Lower cost Uranium-Thorium Dioxide Fuels for Light Water Reactor," with Idaho Nuclear Engineering and Environmental Laboratory as the lead organization; "Fuel for a Once-Through Cycle (Th,U)O2 in a Metal Matrix," with Argonne National Laboratory as the lead; and "A Proliferation Resistant Hexagonal Tight Lattice BWR Fuel Core Design for Increased Burnup and Reduced Fuel Storage Requirements," with Brookhaven National Laboratory (BNL) as the lead. The fourth project is "The Radkowsky Thorium Fuel project," also under BNL lead. This paper describes the three NERI thorium fuel cycle projects.

INTRODUCTION

A new approach to the Department of Energy's (DOE) conduct of nuclear energy research and development (R&D) was recommended by the President's Committee of Advisors on Science and Technology (PCAST) "Panel on Energy Research and Development," in November 1997. As a result, DOE is making a fundamental change in the management of it's nuclear energy research activities. DOE's new approach was initiated with the Nuclear Energy Research Initiative (NERI), which was started in 1999. NERI features a competitive, peer-reviewed, R&D selection process to fund researcher initiated R&D proposals from the universities, national laboratories, and industry. NERI receives guidance from the Nuclear Energy Research Advisory Committee (NERAC). NERAC's primary function is to assist DOE in effectively carrying out its role in nuclear energy research. The advisory committee consists of expert members from a wide variety of research backgrounds and perspectives.

The objective of the NERI program is to address and help overcome the principal technical and scientific obstacles to the future use of nuclear energy in the United States. These obstacles include issues involving proliferation, economics, nuclear waste, and safety. Technologies addressed by NERI include, but are not be limited to, work on proliferation-resistant reactors or fuel cycles; new reactor designs with higher efficiency, reduced cost, and enhanced safety to compete in the global market; lower output power reactors for applications where larger reactors may not be advantageous; and new techniques for on-site and surface storage and for permanent disposal of nuclear waste. NERI is also expected to help preserve the nuclear science and engineering infrastructure within the universities, laboratories, and industry to advance the state of nuclear energy technology and to maintain a competitive position worldwide. DOE believes that by funding creative research ideas under NERI, solutions to important nuclear issues will be realized, and a new potential for nuclear energy in the United States will emerge.

* 1999 meeting.

The budget requests and Congressional appropriated funding for the NERI program to date are the following:

In Fiscal Year 1999, the Administration requested $24 million for NERI and Congress appropriated $19 million.

In Fiscal Year 2000, the request was $25 million and Congress appropriated $22.5 million.

In the first year of the NERI program, DOE made awards for 46 projects. This involved issuance of 53 grants and 38 Interoffice Work Orders (IWOs). For information on the awards and the recipients see the NERI web site at "http://neri.ne.doe.gov."

Three of these NERI projects involve the thorium fuel cycle. These are:

- The Advanced Proliferation Resistant, Lower Cost, Uranium-Thorium Dioxide Fuels for Light Water Reactors Project. The lead organization for this project is the Idaho National Engineering and Environmental Laboratory (INEEL). The Principle Investigator is Phillip E. MacDonald.

- The Fuel for a Once-Through Cycle $(Th,U)O_2$ in a Metal Matrix Project. The lead organization for this project is Argonne National Laboratory (ANL). The Principle Investigator is Sean McDeavitt.

- A Proliferation Resistant Hexagonal Tight Lattice BWR Fuel Core Design for Increased Burnup and Reduced Fuel Storage Requirements. The lead organization for this project is Brookhaven National Laboratory (BNL). The Principle Investigator is Hiroshi Takahashi.

BACKGROUND

These thorium fuel cycle projects differ from earlier thorium fuel cycle work conducted in the U.S. to develop thorium cycle converter-reactor systems. Several prototypes, including the HTGR (high-temperature gas-cooled reactor) and MSRE (molten salt converter reactor experiment), have operated. A uranium-thorium seed blanket fuel arrangement was also used to demonstrate the light water breeder concept at the Shippingport Atomic Power Station. This reactor operated for five years from August 1977 to October 1982. At the end of this period, the core contained approximately 1.3 percent more fissile material after producing heat for five years than it did before initial operation. The only U.S. commercial thorium/uranium fueled HTGR was the Fort St. Vrain reactor near Platteville, Colorado. The reactor, with a capacity of 330 MW(e), began full operation in early 1979. The operation of this full-scale commercial HTGR was marked by intermittent operations resulting in low capacity factors.

While uranium technology in light water reactors has been demonstrated to be very dependable, the use of thorium technology has lagged ever since the closure of the Fort St. Vrain commercial HTGR in 1989. All currently operating commercial nuclear power plants in the United States use uranium

ADVANCED PROLIFERATION RESISTANT, LOWER COST, URANIUM-THORIUM DIOXIDE FUELS FOR LIGHT WATER REACTOR PROJECT

In addition to the lead organization, INEEL, with Phillip E. Macdonald as the Principle Investigator, the following organizations and investigators are participating in this project:

ABB Combustion Engineering Inc., George P. Smith, Jr.
Argonne National Laboratory, Dr. James C. Cunnane
Framatome Technologies, Steward W. Spetz
Massachusetts Institute of Technology (MIT), Prof. Mujid S. Kazimi and Prof. Michael J. Driscoll
Purdue University, Prof. Alvin Solomon
Seimens Power Corporation, Dr. Leo F. P. Van Swam
University of Florida, Prof. James S. Tulenko
Westinghouse Electric Corporation, Dr. E. J. Lahoda

The goal of this project is to develop a ThO_2-UO_2 fuel that is assembly-for-assembly compatible with existing light water reactors (LWRs). The fuel will be developed for a once-through fuel cycle in which in-reactor conversion of ^{232}Th to ^{233}U is maximized and plutonium production is minimized. The fuel will be taken to higher burnup than planned in previous work. No chemical processing of the fuel is considered. The durability of the fuel as a wasteform is important.

The objective of this project is to develop a fuel for the existing LWRs that is less expensive to fabricate than the UO_2 fuel, allows longer refueling cycles and higher sustainable plant capacity factors, is very resistant to nuclear weapons-material proliferation, results in a more stable and insoluble waste form, and generates less high level waste.

The fuel cycle economics of the fuel being investigated is influenced by a number of factors. Extended burnup reactor cores using conventional UO_2 fuel require high ^{235}U enrichments and significant quantities of burnable poisons for reactivity control, which significantly increases costs. However, the reactivity in a ThO_2-UO_2-fueled reactor remains more constant during long irradiations than in a UO_2 core because of the high conversion ratio of the thorium. Calculations using the MOCUP code system indicate that the mixed ThO_2-UO_2 fuel, with about 5.8 wt% of the total heavy metal ^{235}U, could be burned to 72 MW·d/kg using 30 wt% UO_2 and the balance ThO_2. The ThO_2-UO_2 cores can also be burned to about 86 MW·d/kg using 35 wt% UO_2 and 65 wt% ThO_2 with an initial enrichment of about 6.8 wt% of the total heavy metal fissile material.

Longer refueling cycles and higher plant capacity factors can be achieved with this fuel. ThO_2-UO_2 fuel has a significantly higher thermal conductivity at LWR operating temperatures and a lower rate of fission gas release. Therefore, ThO_2-UO_2 fuel can be operated to higher burnup with less difficulty than UO_2 fuel. With improved fuel, many of the U.S. plants could move to 24- to 36-month refueling cycles. An improvement to 24-month cycles is worth about 2.5 percent in plant capacity and an improvement to 36-month cycles would increase plant factors by about 5 percent. Having the same plants generate 5 percent more electricity would save U.S. utilities and thus taxpayers about $1 billion per year.

This thorium fuel cycle also offers a high level of nuclear weapons-material proliferation resistance. The uranium is calculated to remained below 20 wt% total fissile fraction throughout the cycle, making it unusable for weapons. Total plutonium production per MWd was a factor of 3.2 less in the ThO_2-UO_2 fuel than in the conventional fuel. ^{239}Pu production per MWd was a factor of 4.2 less in the ThO_2-UO_2 fuel than in the conventional fuel. The plutonium produced

was high in ^{238}Pu, leading to a decay heat rate 3.7 times greater than that from plutonium derived from conventional fuel and 29 times greater than that from weapons grade plutonium. The decay heat in spent ThO_2-UO_2 fuel is high enough to melt and render ineffective the explosives commonly used in nuclear weapons, unless the weapon is actively cooled. Spontaneous neutron production for plutonium from ThO_2-UO_2 fuel was 1.75 times greater than that from conventional fuel and 12 times greater than that from weapons grade plutonium. High spontaneous neutron production drastically limits the probable yield of a crude weapon.

The fuel investigated in this project has improved waste form stability. Spent UO_2 fuel fragments react and disintegrate relatively rapidly (about 1 percent per year) with water containing Yucca Mt. contaminants. ThO_2 is the highest oxide of thorium and does not depart significantly from its stoichiometric composition when exposed to air or water at temperatures up to 2000 K. Heavily oxidized high thoria solid ThO_2-UO_2 solutions contain urania structures only between UO_2 and U_4O_9 and, therefore, retain their mechanical integrity. The thoria stabilizes the UO_2 and prevents oxidation beyond U_4O_9.

The fuel investigated also has a high level of waste minimization. Use of higher burnup fuel will result in proportionally fewer spent fuel bundles to handle, store, ship, and permanently dispose. The facility operating portion of the planned system to dispose of the nation's spent nuclear fuel and high-level waste has been estimated to be about $13.6 billion over about 40 years, or $32,000 per Boiling Water Reactor (BWR) fuel bundle and $60,500 per Pressurized Water Reactor (PWR) fuel bundle. Approximately 4,000 BWR and 3.400 PWR fuel assemblies are discharged each year in the United States. If the equilibrium cycle discharge burnups in the United States could be increased to 75 MWd/kg, for example, the government could save more than $100 million per year.

This project includes four tasks.

Task 1: Fuel-Cycle Analysis will evaluate the economic viability of a ThO_2-UO_2 fuel cycle in commercial reactors operating in the U. S. Framatome Technologies will add cross-sections for thorium and related isotopes to its SCIENCE nuclear code package and then perform two- and three-dimensional fuel-lattice calculations and calculate power distributions in a typical PWR 17×17 core. Finally, costs for ThO_2-UO_2 and conventional uranium cycles will be compared. MIT will try to further optimize the core design by investigating such things as fuel rod geometry, metal-water ratio, and ThO_2-UO_2 ratios using the CASMO-4 and SIMULATE lattice codes. Both MIT and the INEEL will perform benchmark quality calculations at the rod, cell, and assembly levels using the Monte Carlo code MOCUP, which combines MCNP and ORIGEN.

Task 2. Fuel Manufacturing Costs will determine if the current nuclear fuel fabricators in the U.S. have the capability to manufacture ThO_2-UO_2 fuel economically. Westinghouse will generate process flow sheets; identify equipment, process, safety, and licensing issues and the required plant modifications to current uranium based manufacturing facilities; and determine the projected capital and operating costs. Criticality and radiological safety are particularly important issues that must be addressed for this type of fuel. Purdue will evaluate fabrication issues associated with co-precipitation of the powder and with pressing, sintering and grinding ThO_2-UO_2 fuel pellets and investigate manufacturing techniques to produce low cost fuel.

Task 3. Fuel Performance will evaluate the thermal, mechanical, and chemical aspects of the behavior of ThO_2-UO_2 fuel rods during normal, off normal, and design basis accident conditions. ThO_2-UO_2 fuel has different properties than UO_2 fuel:

· slightly higher decay heat,
· higher thermal conductivity at normal operating temperatures and lower thermal conductivity at high temperatures,
· higher fission gas production per fission, but a lower rate of release of fission gases, and
· higher melting temperature.

Three organizations will be involved in the evaluation of the performance of ThO_2-UO_2 fuel: INEEL, MIT, and Purdue. Purdue will make additional material property measurements including thermal conductivity, creep, and gas induced swelling. MIT will develop a fission gas release model for ThO_2-UO_2 fuel and evaluate innovative ThO_2-UO_2 fuel designs. All three collaborators will do the property correlation work and some steady state analysis; the final transient analysis will be done at the INEEL.

Task 4. Long-Term Stability of ThO_2-UO_2 Waste will determine weather thoria-urania fuel is superior to urania as a fuel waste form. The objective in Year 1 of this task is to determine the oxidation rates in air and in oxygen saturated water of ThO_2-UO_2 fuels with various ratios of thorium and uranium. The objective in Year 2 is to determine the corrosion and dissolution release rates of ThO_2-UO_2 fuel in synthetic ground water. These experiments will be continued in Year 3, along with experiments in a hot cell with 50 MW/kg Shippingport fuel to benchmark the out-of-pile work. The cold laboratory work will be done at the University of Florida, and Argonne will do the hot cell work in collaboration with the University of Florida.

FUEL FOR A ONCE-THROUGH CYCLE–$(TH,U)O_2$ IN A METAL MATRIX PROJECT

The lead organization for this project is Argonne National Laboratory, and the Principle Investigator is S. M. McDeavitt. He is assisted by M. C. Hash. In addition to ANL, Purdue University is participating in this project. The Purdue investigators are A. A. Solomon, T. J. Downar, & S. T. Revankar.

The concept for this fuel is a dispersion of $(Th,U)O_2$ particles that are 50 to 100_μm diameter. The fuel would have a density of 80 to 90 percent and is expected to have low swelling. The fuel particles would be dispersed in a zirconium matrix that has high density, high thermal conductivity, and provides fission product containment. The fuel matrix would be enclosed in a tubular Zircaloy shell that would serve as the powder packing form. The shell would be drawn for the proper density and shape and would be compatible with current Light Water Reactors. The concept is shown in the following Figure 1.

There are two tasks for this 3 year NERI project:
Task 1. Proof-of-Principle Activities will include ceramic microsphere fabrication, dispersion fuel rod fabrication, and fuel modeling of neutronic and thermal properties.

Task 2. Fuel Performance Estimates will be based on past data from dispersion fuels. It will include performance modeling, bounding calculations, and preparation for irradiation experiments.

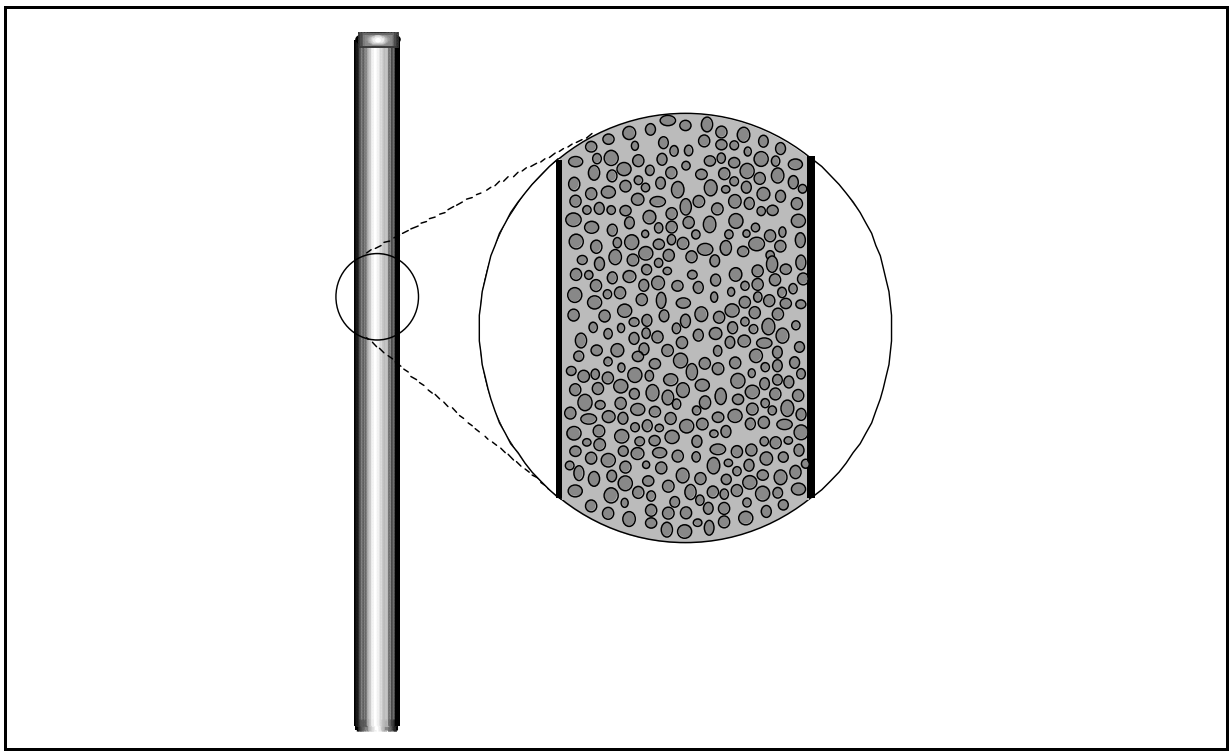

Figure 1. (Th, U)O_2 Metal Matrix Concept

The potential benefits of the metal matrix fuel include:

· High Actinide Burnup. The ^{232}Th to ^{233}U conversion extends the fuel life.
· Proliferation Resistance. Mixed oxides prevent direct chemical separation of ^{233}U and ^{239}Pu.
· Improved Irradiation Stability. Reduced centerline temperature results in stronger physical properties.
· Minimal Waste Treatment. The concept uses direct disposal of spent fuel from the once-through cycle.
· Low Fabrication Cost. Low temperature and simple industrial methods can be used.

PROLIFERATION RESISTANT HEXAGONAL TIGHT LATTICE BOILING WATER REACTOR (BWR) FUEL CORE DESIGN FOR INCREASED BURNUP AND REDUCED FUEL STORAGE REQUIREMENTS PROJECT

The lead organization for this project is Brookhaven National Laboratory, and the Principle Investigator is Hiroshi Takahashi. Upendra S. Rohatgi is also a BNL investigator. Other participating organizations are Purdue University and Hitachi Ltd. The investigator for Purdue University is Thomas J. Downar.

The design objectives of the High Conversion, Boiling Water Reactor (HCBWR) concept are to achieve a high conversion of Th to ^{233}U, reduce accumulated inventory of plutonium while producing useful energy, develop very high burnup BWR fuel using a high concentration of plutonium and a large rate of ^{233}U production, minimize potential for proliferation of weapons grade fissionable materials, maximize inherent safety features of reactor, maximize plant capacity factor, and minimize cost of electricity generation.

The HCBWR is a proliferation resistant, economically competitive concept. It has a very tight lattice with relative small water volume fraction and will operate with a fast reactor neutron spectrum. It has a radially and axially segmented core design. A thin annulus of neutron moderating and absorbing materials separating core and blanket segments provides negative reactivity feedback for high core voiding. Preliminary design study parameters for the HCBWR Reactor are given in the following Table I.

Table I. HCBWR Reactor. Preliminary Design Study Parameters

Parameter	**Value**
Reactor Type	Boiling water cooled Pu oxide Th-233 U oxide high burn up fast reactor
Core Layout	Segmented design (radial and axial) tight hexagonal lattice
Power Level	Range: 600 MW(e) to 1350 MW(e)
Primary System Pressure	~ 8 MPa
Fuel Material	Pu oxide fuel and ^{233}U plus thorium fertile (^{233}U) oxide
Blanket Material	Thorium oxide
Coolant	Boiling water
Blanket Design	(1) Radial and axial blankets (2) Internal blankets
Working Fluid	Water and superheated steam

The HCBWR is expected to have a very high proliferation resistance. The design is constrained so that no natural uranium is incorporated into the fuel feedstock at any time in the fuel cycle. Uranium-233 produced from conversion will not be separated from other isotopic products. The Uranium-232 will be retained with the Uranium-233 to provide handling difficulty resulting emitted radiation and internal heat generation.

RECENT ADVANCES IN THORIUM FUEL CYCLES FOR CANDU REACTORS*

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Abstract. The once-through thorium fuel cycle in CANDU reactors provides an evolutionary approach to exploiting the energy potential of thorium. In the "mixed bundle" strategy, the central 8 elements in a CANFLEX[1] fuel bundle contain thoria, while the outermost 35 elements contain slightly enriched uranium (SEU). Detailed full-core fuel-management simulations have shown that this approach can be successfully implemented in existing CANDU reactors. Uranium requirements are lower than for the natural uranium fuel cycle. Further energy can be derived from the thorium by recycling the irradiated thoria fuel elements, containing ^{233}U, as-is without any processing, into the center of a new mixed bundle. There are several examples of such "demountable" bundles. Recycle of the central 8 thoria elements results in an additional burnup of 20 MW·d/kgHE from the thoria elements, for each recycle. The reactivity of these thoria elements remains remarkably constant over irradiation for each recycle. The natural uranium requirements for the mixed bundle (which includes the natural uranium feed required for the outer SEU fuel elements), without recycle, is about 10% lower than for the natural uranium fuel cycle. After the first recycle, the uranium requirements are ~35% lower than for the natural uranium cycle, and remain fairly constant with further recycling (the total uranium requirement averaged over a number of cycles is 30% lower than a natural uranium fuelled CANDU reactor). This thorium cycle strategy is a cost-effective means of reducing uranium requirements, while producing a stockpile of valuable ^{233}U, safeguarded in the spent fuel, that can be recovered in the future when predicated by economic or resource considerations.

1. INTRODUCTION

High neutron economy, a simple fuel bundle design, and on-power refuelling result in unsurpassed fuel cycle flexibility that is a hallmark of the CANDU[1] reactor. High neutron economy enables maximum energy to be derived from the thorium, minimizing uranium requirements. High neutron economy also opens the door to a variety of fuel cycle strategies that would not otherwise be possible. While the simple fuel bundle design contributes to the high neutron economy of the reactor (by minimizing the amount of structural material associated with the fuel), the simplicity of the fuel design also increases the fuel cycle flexibility.

In fact, the direct-recycle concepts that are the subject of this paper are feasible only because of the simple fuel design. The bundle design also lends itself to optimizing the composition from ring-to-ring, again, a feature that is exploited in the concepts discussed in this paper.

Finally, on-power refuelling of pairs of fuel bundles, with adjacent channels refuelled in the opposite direction (bi-directional fuelling) provides the ability of shaping both the axial power

* 1997 meeting.
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distribution along the channel, and the radial power distribution across the core. The amount of reactivity added to the core during refuelling, and the resultant perturbations in power, can be controlled by the number of bundles added. The axial power distribution can be shaped by judicious arrangement of bundles in the channel.

The fuel cycle flexibility of the CANDU reactor makes it particularly attractive for utilizing thorium as a fuel. The abundance of thorium in the earth's crust is about three times that of uranium. For countries having abundant thorium reserves, the use of the thorium fuel cycle in CANDU reactors would enhance both the sustainability of nuclear power, and the degree of energy independence, using a single reactor type. The physical, chemical, and neutronic properties of thorium make it an attractive nuclear fuel [1].

However, since thorium has no fissile isotope, neutrons must be initially provided by adding a fissile component, either directly to the ThO_2 itself, or outside as separate "driver" fuel, to transmute the ^{232}Th to valuable fissile ^{233}U. The manner in which this is done defines a variety of thorium fuel-cycle options in CANDU reactors.

Since thorium fuel cycles are not commercially employed, there is the opportunity to build into the design of these cycles a very high degree of proliferation resistance, right from the start. This would apply to all parts of the cycle, from the supply of the fissile material to initiate the cycle, to the design of recycle technology, to the supply of any fissile component required as "topping" for the recycled material. Of course, thorium fuel cycles could also be employed to effectively disposition surplus weapons-material (plutonium or HEU), while at the same time creating a valuable source of fissile material for future generations, safeguarded in the spent fuel [2].

The near-term challenge for the use of thorium as a fuel, is conceiving a means of benefiting from its use that does not depend on reprocessing to recycle the ^{233}U produced. Such technology is not available commercially for thorium fuel, and would be extremely expensive. The once-through thorium (OTT) fuel cycle in the CANDU is an elegant solution.

The OTT cycle produces a mine of valuable $^{23}3U$ in the spent fuel, an little or no extra cost, available for future recovery as predicated by economic or resource considerations. Two general OTT options have been developed for CANDU reactors. The first is a "mixed channel" approach, where some channels are fuelled with enriched fuel, which supply the neutrons required to "drive" the ThO_2, contained in a smaller number of separate channels.

This approach allows different dwell times, or burnups, for the two fuel types (a higher dwell time being desirable for the ThO_2 fuel). Because of the disparity in reactivity and power output between driver channels and thoria channels, sophisticated fuel-management schemes would be required to shape the channels and bundle power distributions in this core.

An alternative approach is the "mixed bundle", where the driver fuel and the ThO_2 would be in the same bundle. This is a practical means of utilizing thorium in existing CANDU reactors, while keeping the fuel and the reactor within the current operating and safety envelopes. In the mixed bundle, the central 8 elements in a CANFLEX bundle (e.g. the central element and the next ring of 7 elements) would contain ThO_2. The outermost 35 elements (the ring of 14 elements, and the outer ring of 21 elements) would contain SEU (Figure 1). In this approach, the natural uranium requirements are lower than for a natural uranium fuelled CANDU reactor, but higher than for an SEU-fuelled core of optimal enrichment (around 1.2%).

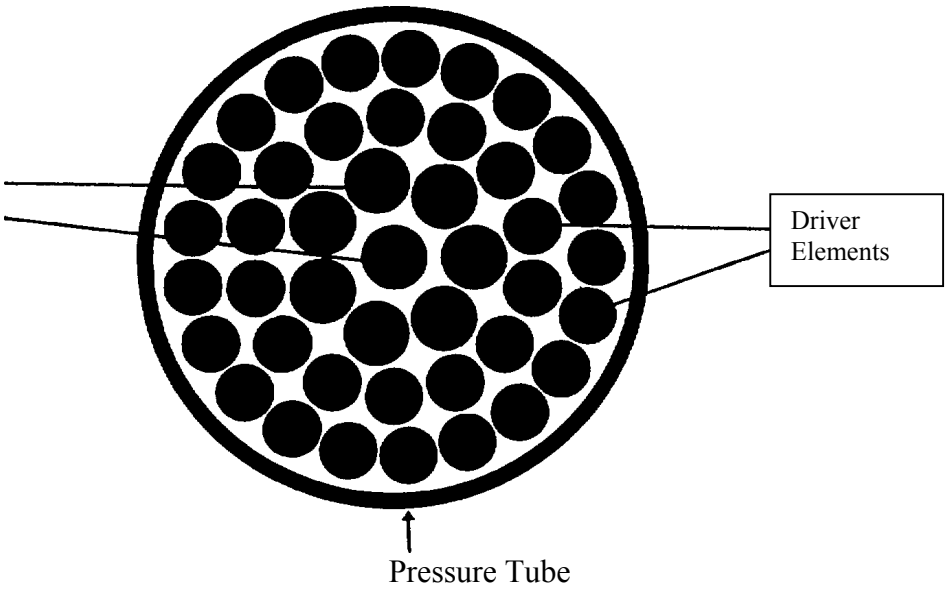


Figure 1. Configuration of Fuel Elements in a CANFLEX Bundle

A previous paper [3] described fuel management simulations for several mixed bundle OTT cycles. It was shown than not only are these concepts technically feasible, but they have several advantages over the current natural uranium cycle. Figure 2 shows the typical axial power distribution in a high powered central channel for a core containing mixed bundles, in which the SEU enrichment is 1.8%. The adjuster rods have been removed from the core, and a 2-bundle shift, bi-directional fuelling scheme is used throughout. For comparison, the axial power distribution for the same channel in a natural uranium-fuelled CANDU reactor, with adjuster rods present, is also illustrated. While adjuster rods flatten the axial power distribution with natural uranium fuel, reducing peak bundle powers, they are not needed for this purpose with enriched fuel. Even without adjuster rods, the axial power distribution in the mixed bundle core is flatter than in the natural uranium core. The inletskewed axial power distribution results in higher thermalhydraulic margin (higher critical channel power), and helps to ensure good fuel performance, since power increases during refueling only occur for relatively fresh fuel, which is resilient to power-ramps. This earlier study shows the flexibility that exists through bundle design and fuel management, in accommodating a variety of mixed bundle OTT fuel cycle options in existing CANDU reactors.

2. DIRECT SELF-RECYCLE IN CANDU

This paper extends the previous work by examining the effect of reusing the central 8 thoria elements after irradiation, into the center of a new mixed bundle containing fresh SEU in the outer 2 rings. Hence, it is an extension of the once-through cycle, to a recycle option that does not involve reprocessing. It is called "direct self-recycle", because the irradiated fuel elements would be directly transferred into a new fuel bundle without any modification to the elements. In a way, it is analogous to the DUPIC cycle [4], involving instead of recycle from a PWR into a CANDU, direct self-recycle into CANDU. This recycle option would have the highest degree of proliferation resistance, with no chemistry involved, and no access to the fuel pellets, and no altering of the fuel element. It would also be immensely cheaper than reprocessing technology.

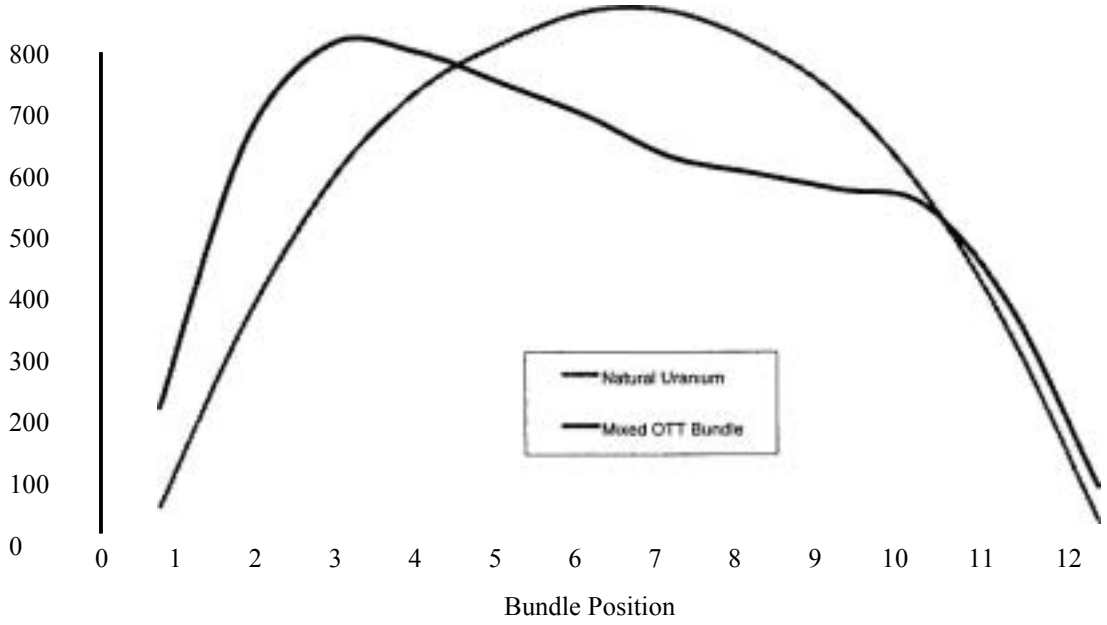


Figure 2. Time-Average Bundle Power Distributions in a High-Power Channel.

3. METHODOLOGY

The on-power fuelling of CANDU reactors means that the core carries a continuous distribution of fuel burnup values from fresh to discharge. This burnup distribution simplifies some aspects of the mathematical modelling of the core. For example, the integral k-infinity ($\int k_\infty$), defined as

$$\frac{\int k_\infty \, dt}{\int dt}$$

of a lattice-cell calculation run from fresh to discharge burnup provides a good indication of the reactivity state of the core as a whole. Conversely, knowing the total buckling from the lattice-cell (leakage from the reactor plus absorption by non-lattice-cell components such as control absorbers and adjuster rods) allows the calculation of the average discharge burnup of the fuel. With this technique, lattice-cell calculations can be used to scope many properties of new fuel cycles without the need for full-core fuel management simulations.

For a typical CANDU reactor, the lattice-cell buckling is around 45 mk, giving a minimum $\int k_\infty$ of 1.045. Many advanced fuel cycle concepts provide sufficient axial power shaping that the adjuster rods could be removed from the reactor. This reduces the lattice-cell buckling to around 35 mk ($\int k_\infty = 1.035$), increasing the discharge burnup of the fuel.

The standard lattice-cell code used at AECL is WIMS-AECL [5]. It has been used extensively for many years to perform these sorts of calculations as well as to provide lattice-cell and kinetics parameters as input for other physics and thermalhydraulic codes. Simulations of the standard, 37-element CANDU fuel give $\int k_\infty = 1.045$ at a fuel burnup of 7.1 MW·d/kg, slightly underestimating the 7.5 MW·d/kg typically achieved by operating CANDU reactors. Calculations performed on natural uranium CANFLEX fuel for this study yield the same discharge burnup. Decreasing $\int k_\infty$ to 1.035, to simulate the removal of the adjuster rods, increases the discharge burnup to 8.44 MW·d/kg. The removal of adjuster rods may not be

practical with natural uranium fuel, but it provides a consistent comparison to the properties of advanced fuel cycles which do permit their removal.

A key parameter in the evaluation of a fuel cycle is the "uranium consumption." This is the amount of natural uranium required to produce a quantity of electrical energy. The uranium consumption is determined by the total energy produced by the fuel, the amount of natural uranium required to fabricate the fuel and by the efficiency of the plant at converting thermal energy to electrical energy. To calculate the amount of natural uranium required in the production of enriched uranium fuel, this study assumes that the enrichment results in tails containing 0.2% ^{235}U. The thermal efficiency of the generating station is assumed to be 31 %. Given these assumptions, the two natural uranium examples above (with and without adjuster rods) give a uranium consumption of 166 MgU/GW(e)·a and 140 MgU/GW(e)·a respectively.

To examine the thorium fuel cycle options described in Section 2, a WIMS-AECL model of the CANFLEX bundle was developed in which the inner eight elements were pure ThO_2 and the outer thirty-five elements were SEU. A calculation was performed to determine the fuel properties with increasing burnup until the target $\int k_\infty$ of 1.035 was reached. Then the fuel was "cooled" to allow the decay of short-lived nuclides, particularly ^{233}Pa, in the ThO_2 elements; the SEU elements were replaced with fresh SEU and the calculation repeated. Five such cycles were performed at which time the ThO_2 fuel was deemed to have reached the end of its life expectancy.

Three enrichment strategies were used for the SEU fuel elements:
1. the SEU elements were enriched to 1.3 wt% ^{235}U for all cycles;
2. the SEU elements were enriched to 1.6 wt% ^{235}U for all cycles; and
3. the SEU elements were enriched to 1.6 wt% ^{235}U for the first cycle and 1.3 wt% ^{235}U for the remaining cycles.

The results of all calculations are given in the following section.

4. RESULTS

4.1. Uranium Fuel Cycles

To serve as a measure of the value of the thorium fuel cycles, four uranium cycles were examined. In addition to the two natural uranium examples described above (with and without adjuster rods) calculations were performed for "optimally enriched" (1.2 wt%) SEU. Thus, the four reference, uranium cases examined are:

1. natural uranium with a target $\int k_\infty$ of 1.045 (a CANDU reactor with adjuster rods)
2. natural uranium with a target $\int k_\infty$ of 1.035 (a CANDU reactor without adjuster rods)
3. enriched uranium with a target $\int k_\infty$ of 1.045 (a CANDU reactor with adjuster rods)
4. enriched uranium with a target $\int k_\infty$ of 1.035 (a CANDU reactor without adjuster rods)

The two natural uranium cases achieve exit burnup values of 7.1 and 8.4 MW·d/kg; and uranium consumption values of 166 and 140 Mg U/GW(e) a respectively. The two SEU cases achieve exit burnup values of 21.6 and 22.8 MW·d/kg and uranium consumption values of 107 and 101 Mg U/GW(e)·a respectively. It can be seen that converting to slightly enriched uranium can provide a substantial improvement in uranium consumption. These results are summarized in Table I.

Table I. Burnup and Uranium Consumption for the all-Uranium Reference Cases

Enrichment	Target $\int k_\infty$	Burnup MW·d/kg	Consumption Mg/GW(e)·a
natural	1.045	7.1	165.8
natural	1.035	8.4	139.5
1.2%	1.045	21.6	106.7
1.2%	1.035	22.8	101.1

4.2. 1.3% SEU/Th bundle

The first thorium fuel cycle examined is one based on a CANFLEX fuel bundle in which the outer thirty-five elements are SEU with an enrichment of 1.3%. The central eight ThO_2 elements were repeatedly burned and reused as described in Section 2.1. The analysis assumes that the bundle is run at a constant, high power. The actual power history would be determined by the fuel management scheme (see, for example, the axial power shape in Figure 2).

Figure 3 shows the lattice cell k_∞ as a function of burnup for each of the five cycles. All calculations are cut off when the integral k_∞ ($\int k_\infty$) has reached 1.035. As can be seen from this figure, the fresh fuel bundle runs out of reactivity at a comparatively low burnup value (12.6 MW·d/kg). After the first recycling of the ThO_2 elements, however, the build in of ^{233}U provides a substantial increase in initial reactivity of the fuel. Thereafter, the reactivity stays remarkably constant and the remaining cycles all achieve in burnup values in excess of 20 MW·d/kg.

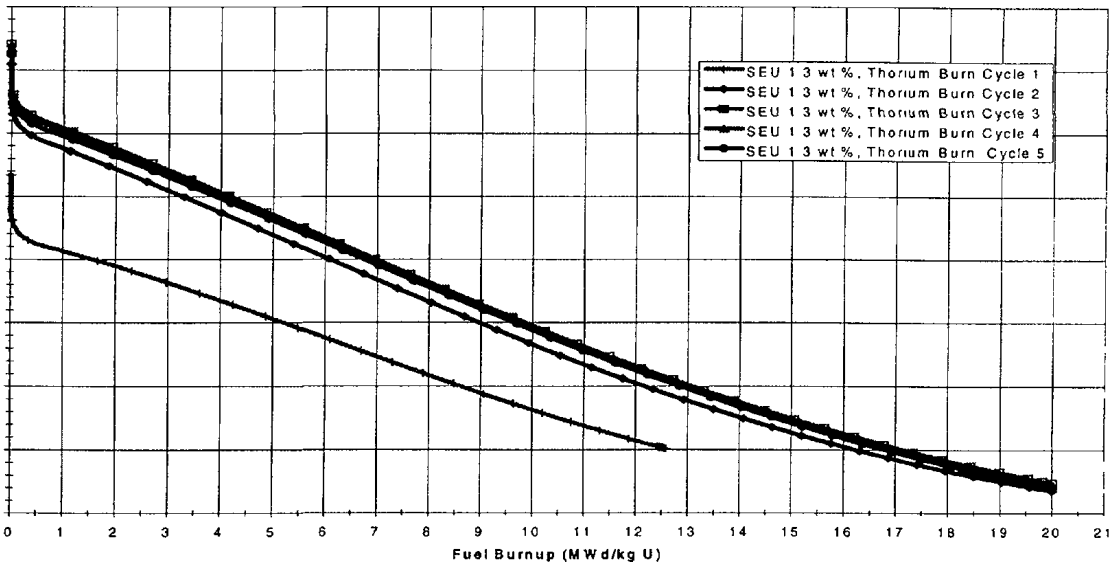
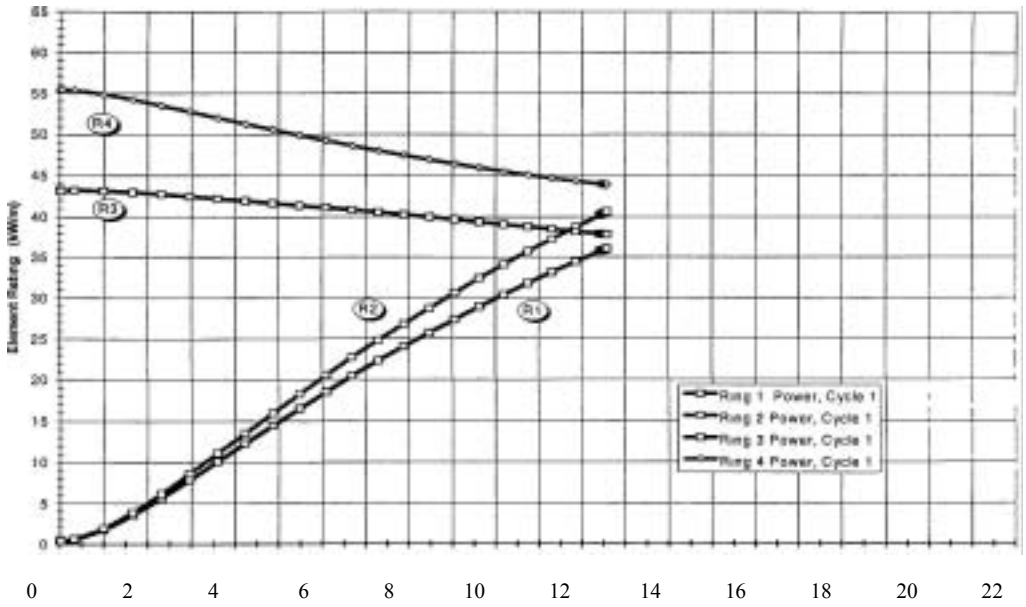


Figure 3. Lattice K-infinity for Mixed OTT Bundle with 1.3% SEU Driver Pins.

Table II gives the bundle-average burnup, uranium consumption, and the cumulative (i.e. including all previous cycles) consumption after each cycle. Because of the low burnup, the first cycle has a relatively high consumption of 156.4 Mg U/GW(e)·a. This value is worse than the comparable natural uranium case (for a CANDU reactor with no adjuster rods) but somewhat better than the present natural uranium cycle. Successive cycles produce a consumption of around 90 Mg U/GW(e)·a, which results in a steady improvement in the total consumption.

Table II. Burnup and Uranium Consumption for the Mixed OTT Bundle with 1.3% SEU Driver Pins

Cycle	Burnup MW·d/kg	U Consumption Mg/GW(e)·a	Cumulative U Consumption
1	12.6	156.4	156.4
2	20.5	96.0	126.2
3	22.4	87.8	113.4
4	22.1	889	107.3
5	21.6	91.1	104.0



Bundle average burnup (MW·d/kg U)

Figure 4. Element Ratings of 1.3% OTT Bundle in Cycle 1.

With no initial fissile material, the fresh ThO_2 elements produce no power. The power produced by the ThO_2 elements increases as irradiation progresses because of the conversion of the fertile ^{232}Th to fissile ^{235}U. The linear ratings of thorium elements have increased to the level of the SEU elements by the time when the whole bundle reaches the discharge burnup. The linear ratings of all four rings of fuel are presented in Figure 4. The ThO_2 elements have a substantial fissile component starting from the second cycle. The fresh bundle produces a smooth power profile, with the ratings highest on the outside of the bundle and lowest in the centre. As the bundle accumulates burnup, the power in the SEU elements decreases (due to the depletion of ^{235}U and the build up of parasitic fission products), while the power generated by the ThO_2 elements increases (due to the increasing concentration of ^{233}U). This trend is shown in Figure 5.

For successive cycles, the initial ^{233}U concentration in the ThO_2 elements is sufficient to generate ratings similar to the outer SEU elements. Again, as the bundle accumulates burnup, the power in the SEU elements decreases and the power generated by the ThO_2 elements increases. At discharge burnup, the ThO_2 elements have substantially higher liner ratings than the SEU elements. Figure 6 shows the power profile for the fifth (final) cycle of thoria fuel elements.

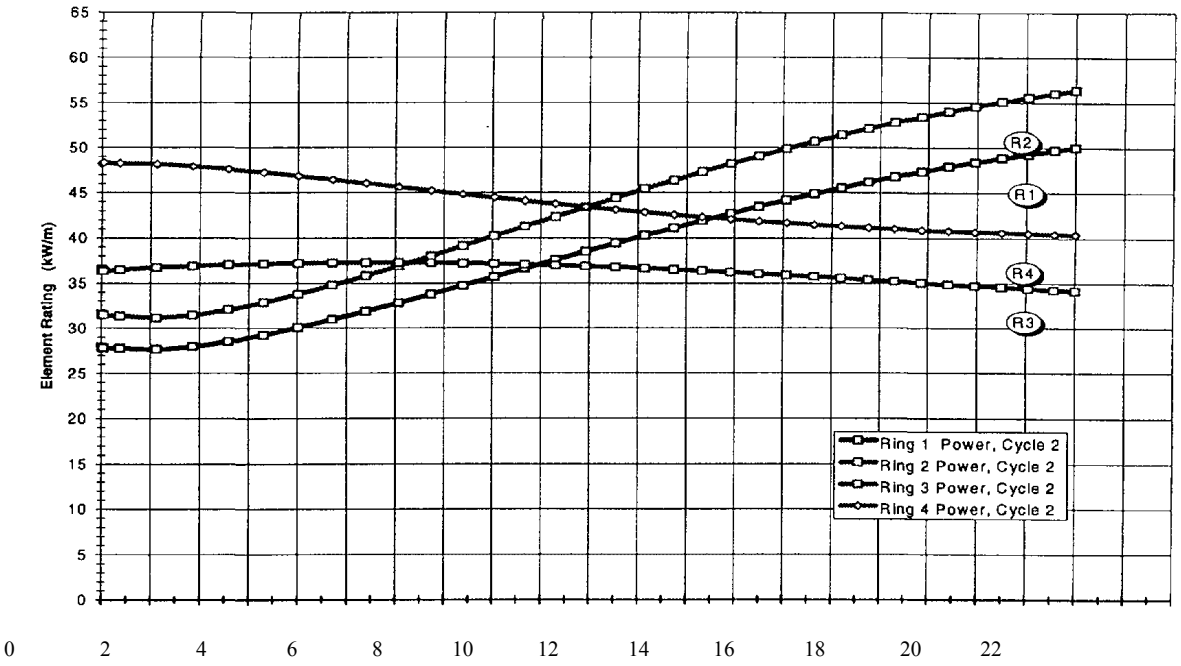


Figure 5. Element Ratings of 1.3% OTT Bundle in Cycle 2.

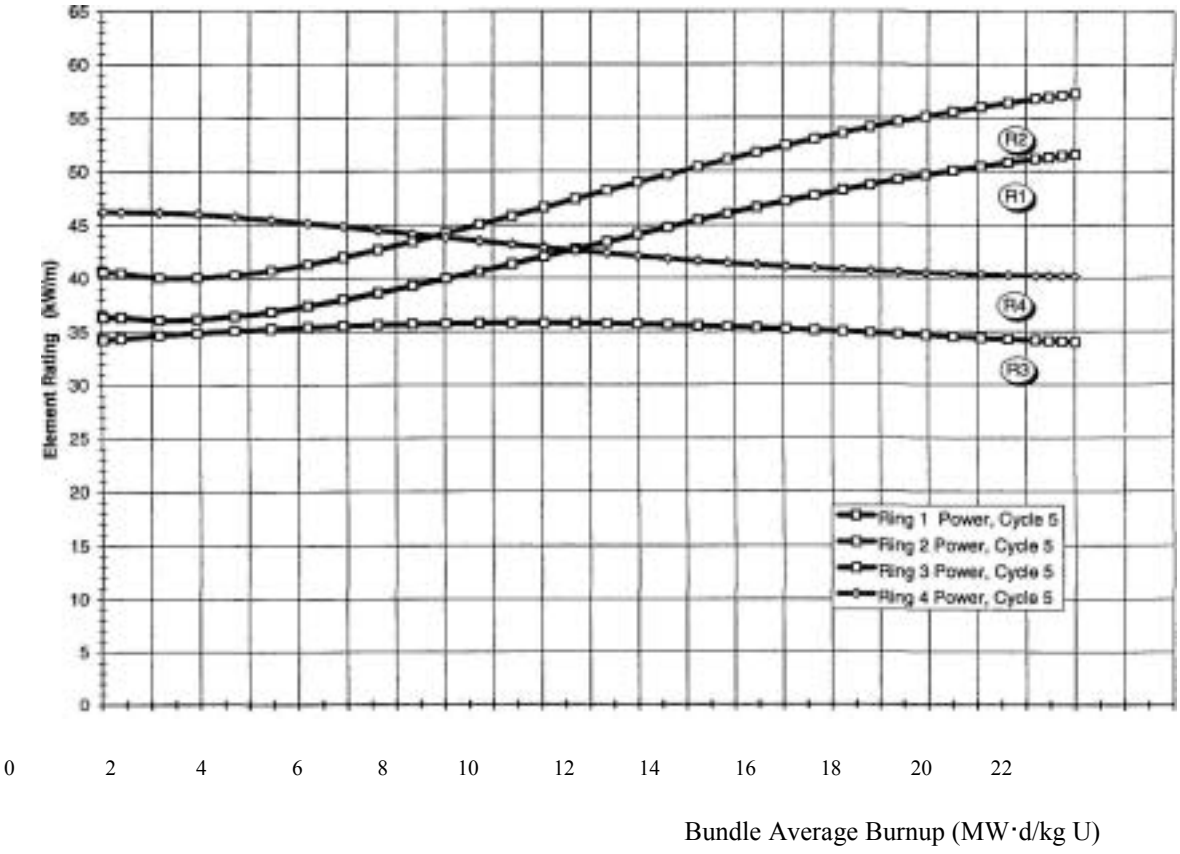


Figure 6. Element Ratings of 1.3% OTT Bundle in Cycle 5.

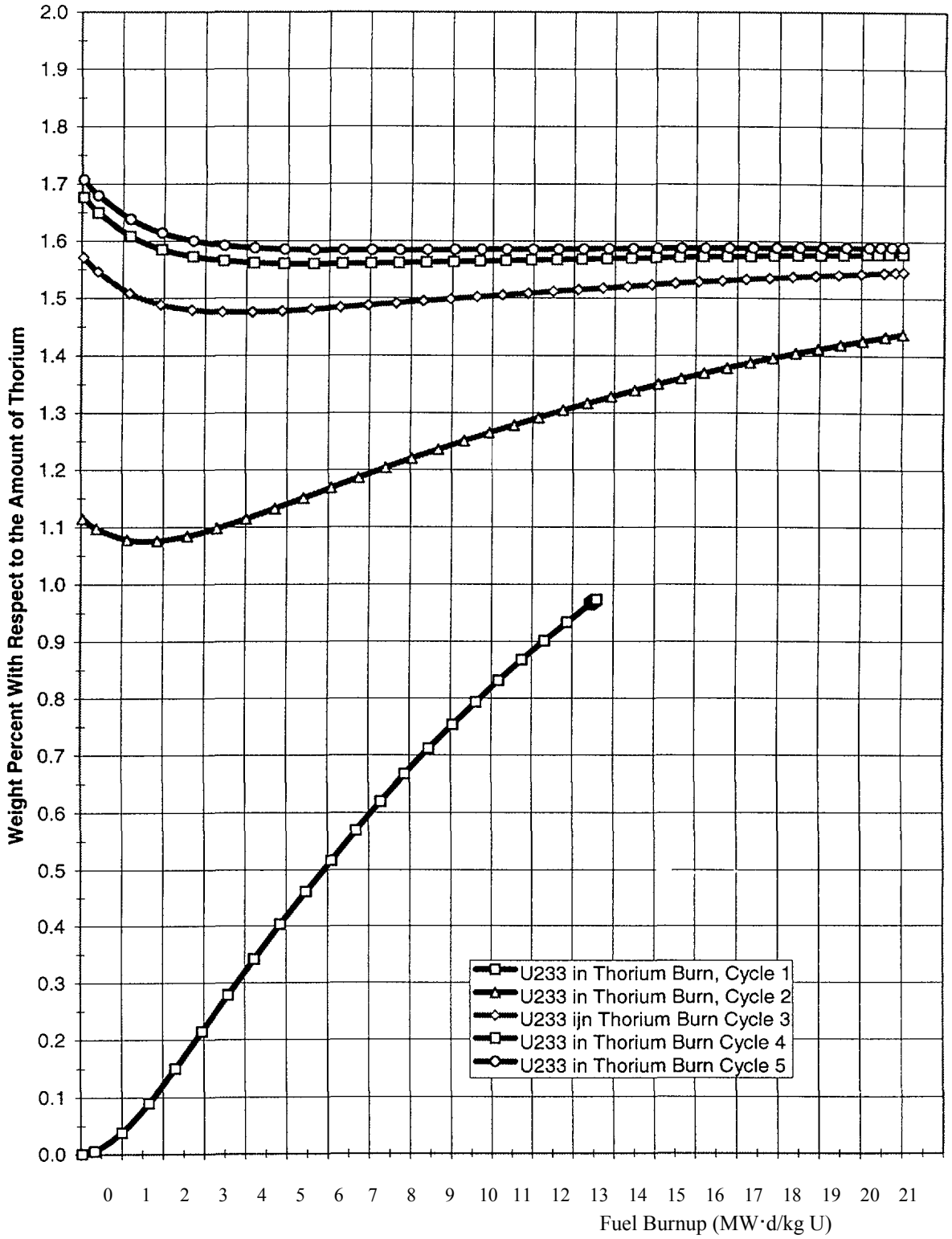


Figure 7. U-233 Content in Ring 2 Vs Bundle-Average Fuel Burnup in 1.3% OTT Bundle.

The characteristic shape of the power profiles are driven by the accumulation of ^{233}U in the ThO_2 elements. Figure 7 shows the ^{233}U content of the second ring of ThO_2 elements for each irradiation cycle. The ^{233}U content increases steadily for the first two cycles, after which it has reached an equilibrium value and remains remarkably constant with increasing burn up. The figure clearly shows the transient increase in concentration at the beginning of each cycle from the decayed ^{233}Pa.

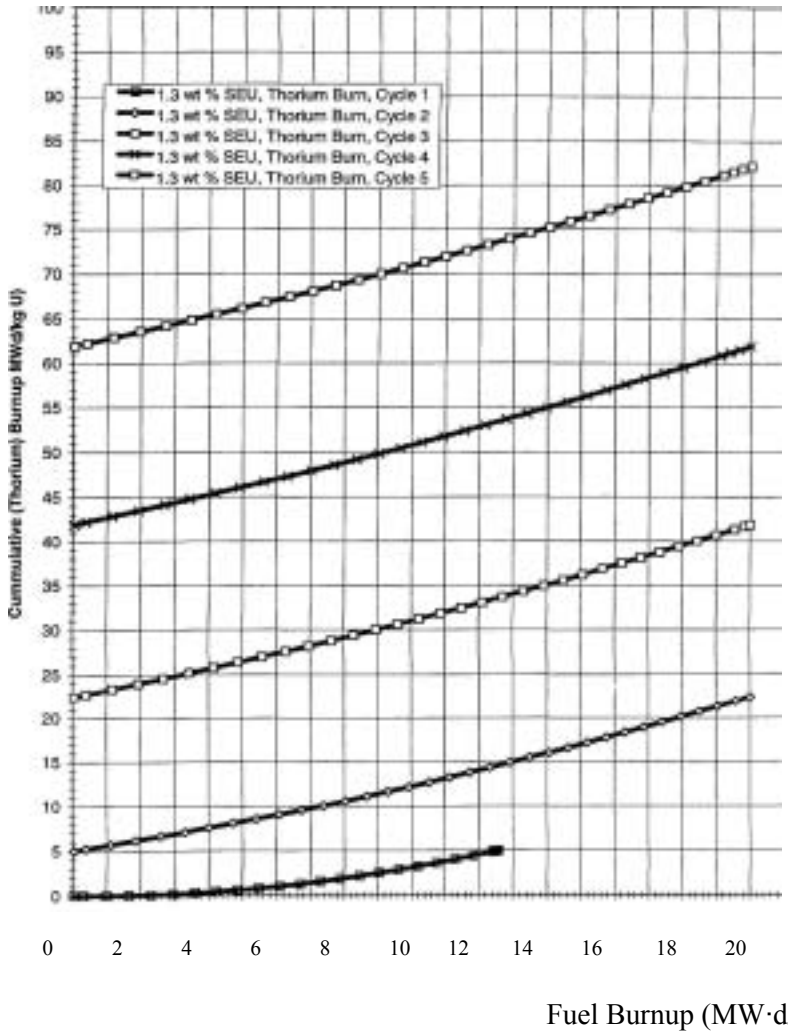


Figure 8. Accumulation of Burnup in Ring 2 Thoria Element.

Figure 8 shows the burnup accumulated on the second ring of ThO_2 elements. Initially, the thoria burnup increases very slowly, due to the low fissile content, acquiring only 5 MW·d/kg of burnup after the first cycle. After reaching an equilibrium fissile content by the end of the second cycle, the accumulation of burnup is nearly constant, with each cycle producing approximately 20 MW·d/kg of burnup on the thoria fuel elements. This is to be expected because the initial fissile concentration is almost constant at the beginning of each cycle.

After five burnup irradiation cycles on the thoria fuel elements, the net uranium consumption is around 104 Mg U/GW(e)·a. This value represents a 25% improvement over natural uranium and is comparable to the optimum SEU fuel cycle.

4.3. 1.6% SEU/Th bundle

The overall improvement in uranium consumption in the previous case is limited by the low burnup of the first cycle. Driven by 1.3% SEU, the resulting burnup at the end of the first cycle is insufficient to recover much of the initial investment in fissile material required to produce a ^{233}U inventory in the thoria fuel elements. High burnup in the thoria elements is a prerequisite for a successful once-through thorium cycle in order to recapture this initial investment.

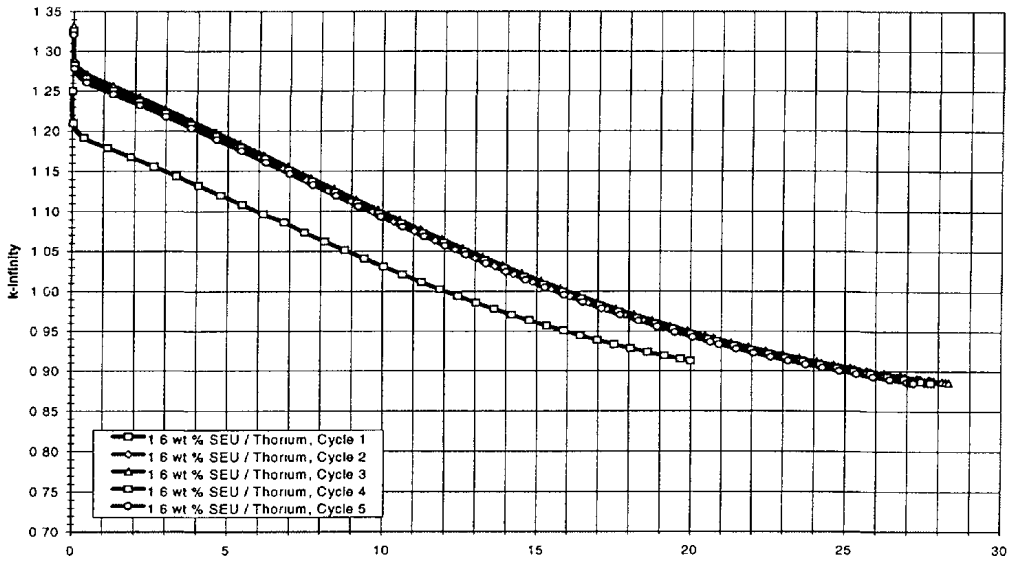


Figure 9. k-infinity Vs Fuel Burnup for 1.6% OTT Bundles.

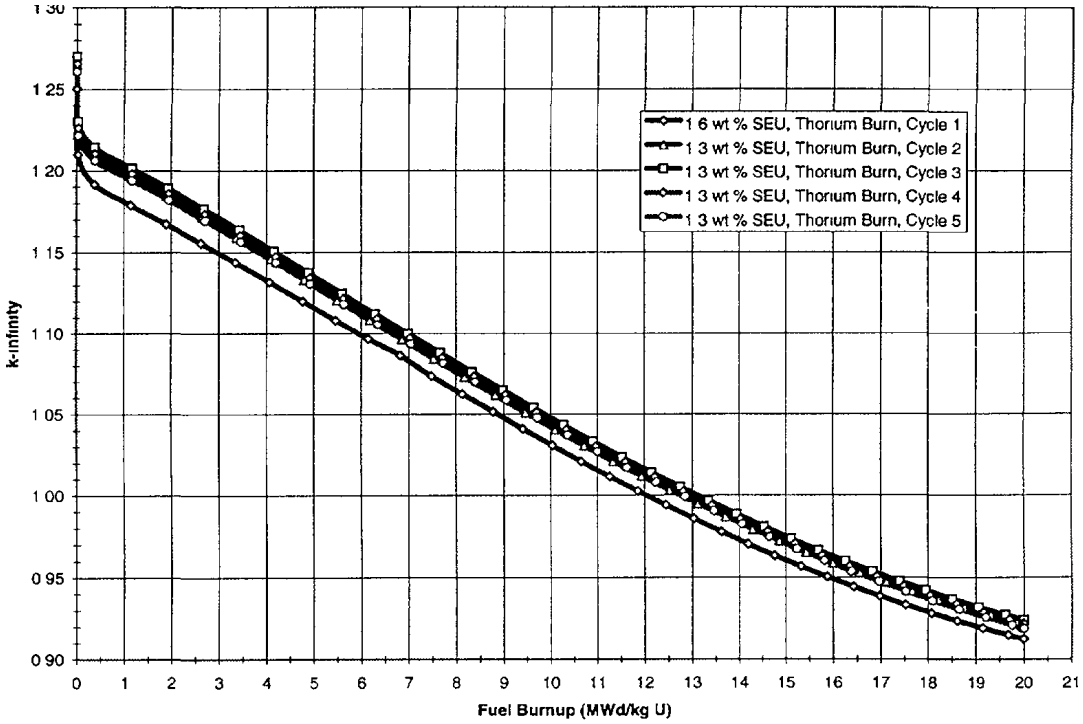


Figure 10. k-infinity Vs Fuel Burnup for Combined 1.6% and 1.3% OTT Bundles.

One method of increasing the burnup achievable by the fuel bundle is to increase the initial fissile content. To explore this effect, WIMS-AECL calculations have been conducted for a mixed OTT bundle with 1.6% SEU driver pins.

For the first cycle, the bundle reaches an average burnup of 20 MW·d/kg and achieves uranium consumption of 125.2 Mg U/GW(e)·a. This uranium consumption is substantially better than the first cycle of the case with 1.3% SEU driver pins and the natural uranium

reference case. As a result of the higher burnup in the thoria fuel elements, the uranium consumption of the second cycle, with a burnup of 27.7 MW·d/kg, is already at the equilibrium value of 90 Mg U/GW(e)·a. After five irradiation cycles the cumulative uranium consumption is around 97.0 Mg U/GW(e)·a. This is 37% better than the corresponding value for the 1.3% SEU/Th case, mainly because of the much higher burnup in the thoria fuel elements. It is 30% better than the natural uranium reference case and is slightly better than the optimally enriched SEU fuel cycles.

Table III. Burnup and Uranium Consumption for the Mixed OTT Bundle with 1.6% SEU Driver Pins.

Cycle	Burnup MW·d/kg	U Consumption Mg/GW(e)·a	Cumulative U Consumption
1	20.0	125.2	125.2
2	27.7	90.4	107.8
3	28.3	88.5	101.4
4	27.7	90.4	98.6
5	27.2	92.1	97.3

Table IV. Burnup and Uranium Consumption for the Mixed OTT Bundle with 1.6% SEU Driver Pins in the first cycle and with 1.3% SEU Driver Pins in Successive Cycles.

Cycle	Burnup MW·d/kg	U Consumption Mg/GW(e)·a	Cumulative U Consumption
1	20.0	125.2	125.2
2	21.7	90.6	107.9
3	22.4	87.9	101.2
4	21.9	89.7	98.3
5	21.5	91.7	97.0

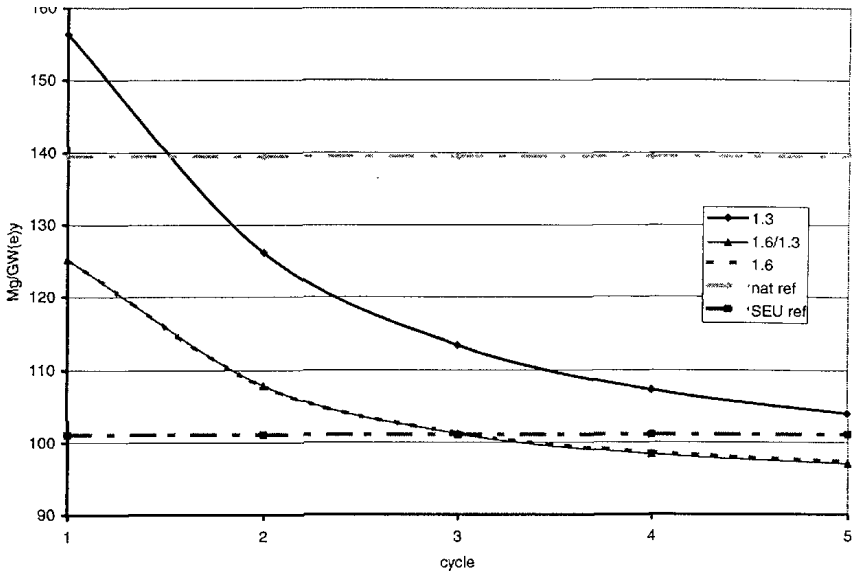


Figure 11: Comparison of Uranium Consumption of All Fuel Cycles.

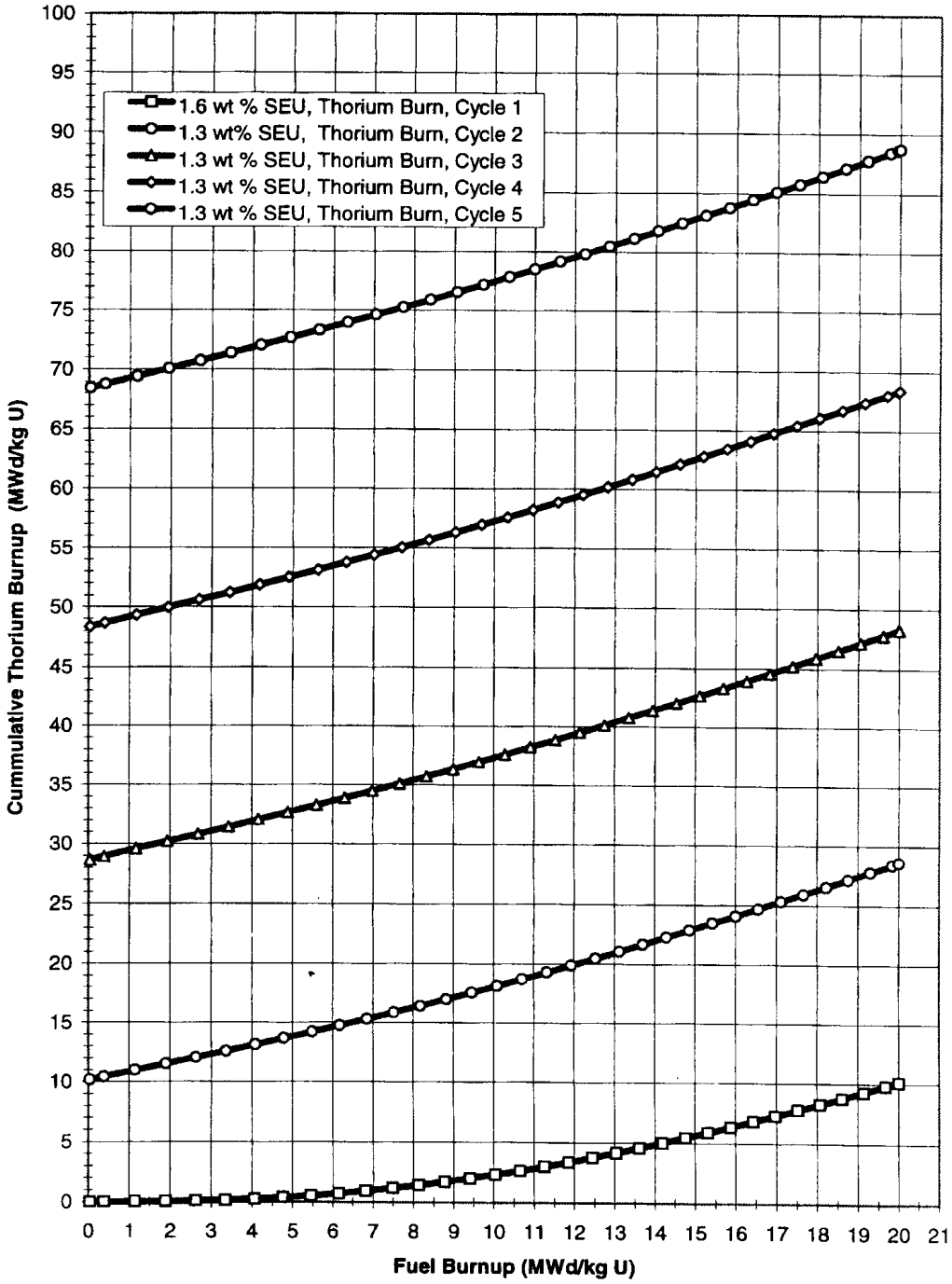


Figure 12. Cumulative burnup for ring 2 thoria pins in combined 1.6% and 1.3% OTT cycles

Figure 9 shows the lattice-cell k_∞ as a function of burnup for each of the five cycles. Table III gives the bundle-average burnup, uranium consumption, and the cumulative consumption after each cycle.

The results show that there a benefit to achieving a higher burnup value in the first cycle. For subsequent cycles, however, the increased enrichment of the driver pins has little effect on the uranium consumption because larger amount of natural uranium is required to produce fuel of higher enrichment.

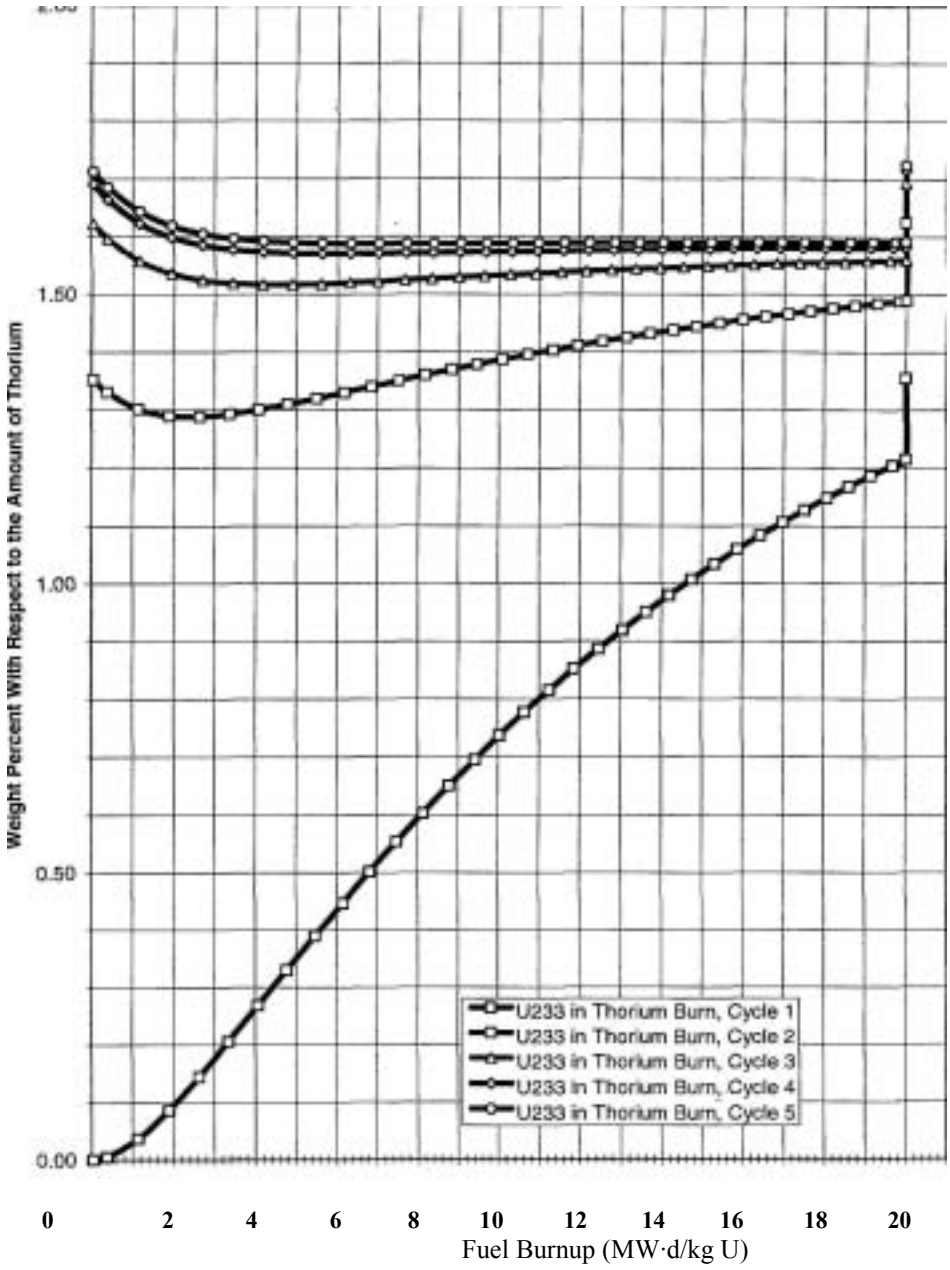


Figure 13. U-233 Content in Ring 2 Thoria Pins of Combined 1.6% and 1.3% OTT Cycles

4.4. 1.6% SEU/Th bundle in first cycle, 1.3% SEU/Th bundle in subsequent cycles

Comparisons of the results obtained for the two OTT designs lead to the third design option where 1.6% enriched SEU driver elements were used for the first irradiation cycle and 1.3% enriched SEU driver elements were used for the subsequent cycles.

Figure 10 shows the lattice-cell k_∞ as a function of burnup for the combined 1.6% /1.3% driver element option. For this case, all cycles achieved similar bundle-average burnup values of between 20 and 22 MW·d/kg. The similarity of the five curves would significantly simplify the implementation of the fuel management strategies required to accommodate the various irradiation cycles in the reactor.

Table IV summarizes the bundle-average burnup, uranium consumption, and the cumulative consumption after each cycle. Figure 11 compares the uranium consumption of all three thorium-fuelled cases and the two uranium-fuelled cases without adjuster rods. The uranium consumption of the combined 1.6%/1.3% SEU driver element case is better than that for optimally enriched SEU fuel cycles.

Some of the improvement of this cycle over the cycle with all 1.3% SEU driver elements can be seen by examining the burnup on the thoria fuel elements, as shown in Figure 12. The first irradiation cycle of the thoria elements produces a burnup of 10 MW·d/kg on the second ring of thoria elements in comparison to only 5 MW·d/kg for the case with all 1.3% SEU driver elements. In both studies, subsequent cycles produce an additional 20 MW·d/kg of burnup on the thoria fuel elements.

Figure 13 shows the ^{233}U content of the second ring of thoria fuel elements. The curves show a similar behavior as for the case with all 1.3% SEU driver elements. The most obvious difference is that the extended burnup on the first cycle of the 1.6% SEU case results in a much higher fissile content at the beginning of the second cycle (1.35% ^{233}U after ^{233}Pa decay as opposed to only 1.1%). In both cases, the equilibrium ^{233}U concentration of about 1.6% is reached at the end of the second cycle.

5. OTHER CONSIDERATIONS

These studies illustrate the tremendous flexibility that is inherent in the CANDU reactor for exploiting the use of thorium fuel cycles. At this stage, these are conceptual studies, although previous work has demonstrated the practicality of the OTT cycle, and the ease and feasibility of fuel management. The same fuel management scheme could be used for the recycled fuel, or for a core containing a mixture of fresh and recycled mixed-bundles. No attempt has been made to optimize either the bundle or fuel cycle strategy. Some additional considerations that might be taken in further studies are as follows.

The ^{233}U content of the thoria elements remains approximately constant with recycle; e.g., self- recycle does not increase the amount of ^{233}U available for future use. Moreover, by recycling the thoria elements, the number of those elements also does not increase. If the overall objective is to maximize the amount of ^{233}U stockpiled" for future recycle, then recycle might not be done, or the number of recycles might be limited. For instance, if the thoria elements in the discharged 1.6% SEU/Th mixed bundle are recycled into a mixed bundle containing 1.3% SEU for the first recycle, the overall uranium requirements (averaged over the first case and the first recycle) are about the same as for the best SEU case. Hence, ^{233}U has been produced at no net expense in terms of uranium requirements (and 25% lower uranium requirements than for the natural uranium cycle). That spent fuel could then be stored in the spent fuel bays, and more ^{233}U generated in fresh SEU/Th mixed bundles.

Also, there has been no attempt in these studies to optimize the uranium-to-thorium content of the bundle. This could be done to minimize the overall uranium requirements, to maximize the amount of ^{233}U produced, or to maximize the usage of thorium. For instance, the improvement in uranium consumption (and the ^{233}U produced) in this study is limited by the amount of thoria in the bundle (confined to the central 8 elements, or 25% of the fuel volume). One could increase the amount of thoria in the mixed bundle (and in the core) by using thoria in the ring of 14 elements. The flexibility of the CANDU fuel bundle design offers many optimization options to meet specific fuel cycle objectives.

6. DEMOUNTABLE BUNDLES

A key enabling technology in this direct, self-recycle option is the so-called "demountable bundle". The intent of this study is to explore the reactor physics aspects of this concept without providing detailed bundle design specifications. Nonetheless, it is very likely that such a bundle is technically feasible, and in fact, there are several embodiments of this concept.

For many years, fuel researchers at AECL have made use of a demountable 37-element bundle for irradiation testing of advanced fuels in the NRU research reactor. The bundle is

designed to enable any of the 18 elements in the outer ring of fuel to be removed, remotely, with the fuel bundle under water in the reactor bays. Thus, elements can be removed at different bumps and new elements added to the bundle. Elements that develop defects can be removed, and the irradiation continued with the rest of the elements. The design has proven to be durable and practical.

A demountable CANFLEX bundle has been recently designed, and is undergoing qualification testing before use in NRU. This design allows even more flexibility than the demountable 37-element design, allowing access to both the smaller elements in the outer ring, and the 7 larger elements in the inner ring.

A final example of such technology is the "advanced carrier bundle" [6]. This bundle was designed for irradiating specimens of pressure tube or calandria tube material in a commercial CANDU power reactor. In this bundle, two of the elements in the ring of 6 elements in a 37-element bundle are replaced by a tube occupying the space of those two elements. The tube is perforated to allow access to the coolant, and the specimens are mounted inside the tube. To achieve high fluence, the bundle is designed to allow the tube to be removed, once the bundle has been discharged into the spent fuel bays. The tube would be mounted into a fresh carrier bundle, underwater in the bays, and this bundle would then be "back-fuelled" into the reactor to continue the irradiation of the specimens. This demonstrates the demountable bundle concept, its handling under water in the spent fuel bays, and its reintroduction into the fuelling machines and the reactor via "back-fuelling".

7. SUMMARY

The CANDU reactor is an ideal vehicle for exploiting the energy potential of thorium-based fuels. Its high neutron economy, simple fuel bundle design, and on-power refuelling make many different methods possible. The direct self-recycle is one such method which meets the requirements of a viable fuel cycle. Further, it offers reduced uranium consumption, compared to optimal SEU cycles, while creating a valuable inventory of ^{233}U.

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II. THORIUM UTILIZATION, INCLUDING HYBRID SYSTEMS (ADS, FUSION ETC.)

THORIUM CYCLE IMPLEMENTATION THROUGH PLUTONIUM INCINERATION BY THORIUM MOLTEN-SALT NUCLEAR ENERGY SYNERGETICS*

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Abstract. Considering the increasing world energy demand and the urgent necessity of replacement of fossil-fuel by nuclear energy for survival of the global environmental crisis, we urgently need to prepare a more rational and a huge nuclear industry. As an improved alternative of present technology, the utilization of U is strongly recommended. ORNL proposed an idealistic MSBR since 1970. We modified it to the world-wide applicable system: THORIMS-NES [Thorium Molten-Salt Nuclear Energy Synergetic System], which is composed of simple thermal fission power stations (FUJI) and fissile-producing Accelerator Molten-Salt Breeder (AMSB). FUJI is a size-flexible NEAR BREEDER even not using continuous chemical processing and core-graphite exchange, and AMSB is based on a single-fluid molten-salt target/blanket concept, the technological development of which is easy and simple except for the high-current proton accelerator. THORIMS-NES has many advantages, and here the issues of safety, nuclear-proliferation and social/philosophical acceptance is mostly explained. In practice, the shift to THORIMS-NES from the present U-Pu cycle era will be smoothly implemented by converting Pu and TRU in weapons and spent-fuels into molten fluoride salt by a drying process (such as the Russian FREGATE project) which was established by the French, Russians and Czechs. Pilot plant "mini FUJI", 7MW(e) might be commissioned after 7 years depending on the result of successful 4 years operation of MSRE in ORNL, and Small Demonstration Reactor "FUJI-Pu", 150MW(e) can probably be in operation 12 years from now utilizing the world ability of Na-Reactor Technology. Depending on such MSR-technology development, AMSB-Pu might be able to industrialize 20 years from now.

1. INTRODUCTION

The world is facing several serious crises not only from nuclear weapon-material but also from poverty, population explosion and environmental problems. To solve such issues in the next century the world needs huge energy supplies and it seems that nuclear fission energy is the most promising solution if the following issues are to be solved:

(a) safety
(b) radio-waste
(c) anti-nuclear proliferation and terrorism, and
(d) public/institutional acceptance and economy in the global application.

* 1997 meeting.

Such an aim will not be achieved by minor modification of past technologies but should be expected to depend only on the principally new and ambitious fuel concepts. And it will be a semi-final attempt in the nuclear energy industry because the major energy technology at the end of the next century will be required to be non-heat-emission types such as solar energy.

2. GLOBAL ENERGY STRATEGY IN THE NEXT CENTURY

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

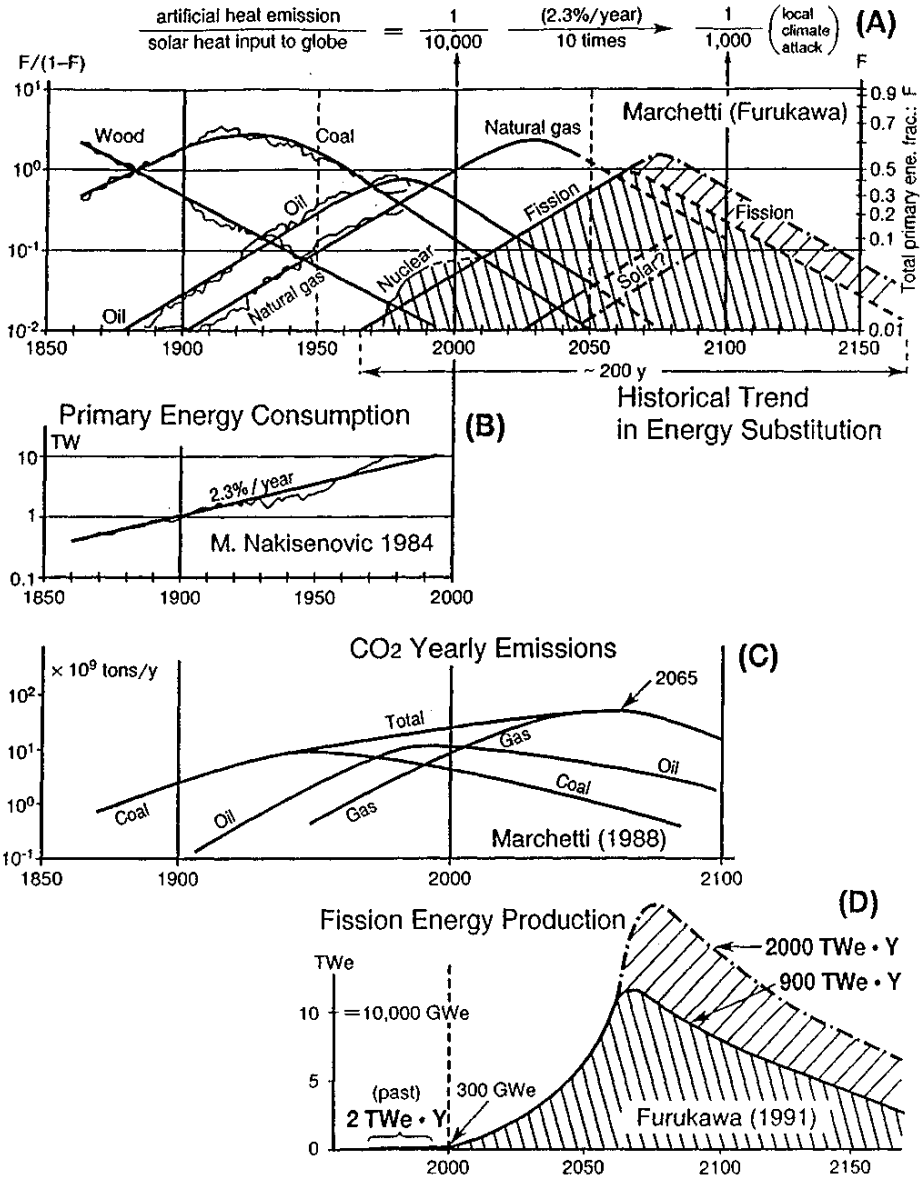

Figure 1. Global Future Energy Prediction. (A) is an extension of Marchett's estimate of historical trend in energy substitution; (B) growth-rate of world primary energy consumption. The predictions of © CO_2 yearly emission from fossil fuels, and (D) nuclear fission-energy production base on (A) and 2.3% annual growth-rate of world energy.

Such advanced prediction will be understood from the illustrations in Figure 1, basically depending on and extending the Marchetti's prediction on the future energy [1,2,3]. If we tentatively accept the global energy growth rate of 2.3% as in the past, the necessary fission energy will be "1000-2000" TW(e) per year in the next century. This is "500-1000" times larger than the past (peaceful) fission energy production of only "2" TW(e) per year (Figure 1(D)). (In here we have to recognize that even such huge nuclear energy will not be enough to solve the CO_2 Greenhouse effect as shown in Figure 1(C)[1]).

It will not be achieved by the present U-Pu solid-fuel cycle system such as lWR and LNFBR due to several difficulties connected with (a) safety [including severe accidents], (b) radio-wastes [including production of trans-uranium [TRU] elements, (c) nuclear-proliferation and -terrorism [including the plutonium-elimination issue], and (d) public and institutional acceptance related with the technological simplicity, flexibility and economy in the global applications.

A "nuclear energy system" should be a "NUCLEAR CHEMICAL REACTION ENGINEERING FACILITY" and essentially a "CHEMICAL PLANT'. Following that, a more rational nuclear energy system should be developed fully, re-examining all scientific/engineering efforts devoted in this century.

First of all, the "FISSION BREEDING POWER STATION" concept such as LMFBR and even MSBR [Molten-Salt Breeder Reactor] proposed by ORNL will not be practical due to (1) the complexity in structure and operation/maintenance (2) the weak breeding performance, and (3) non flexibility in power size [2]. As a new measure a simple rational thorium-molten salt breeding fuel-cycle system, named "Thorium Molten-Salt Nuclear Energy Synergetics [THORIMS-NES]" has been proposed [4,2], which might realize a rational New Nuclear Energy Era in 20-30 years.

3. NEW PHILOSOPHY: 'THORIUM MOLTEN SALT NUCLEAR ENERGY SYNERGETICS" (THORIMS-NES)

Our proposal, THORIMS-NES, depends on the following three principles [4,2]:

(I) Thorium utilization: Natural thorium has only one isotope, ^{232}Th, which can be converted into the fissile ^{233}U in a similar manner as ^{239}Pu converted from ^{238}U. ^{233}U is suitable for thermal reactors and produces only negligible TRU, but 233U fuel is accompanied with strong gamma activity requiring a fluid type fuel.

(II) Application of molten-fluoride fuel technology: The molten salt ^{7}LiF-BeF_2 (Flibe-named by OPRNL) is the significantly low thermal-neutron cross-section material and the best solvent of fissile and fertile materials. This liquid is multi-functional not only as nuclear reaction medium useful for fuel, target or blanket, but also as heat-transfer and chemical processing mediums, which was verified by ORNL [5].

(III) Separation of fissile-producing breeders (process plants-AMSB: Accelerator Molten-Salt Breeder) and power generating fission-reactors (utility facilities-MSR: Molten-Salt reactor): It will be essential for the global establishment of breeding-cycle all over the world. It should be recognized that the doubling time of fission industry growth needs 10 years as shown in Figure 1(D)[2].

This system is practically composed of simple power stations MSR named FUJI-series, fissile producers AMSB, and batch-type process-plants establishing a Symbiotic Thorium Breeding Fuel Cycle System [THORIMS-NES], which successfully presented a high public acceptance [Chap.7].

MSR: FUJI (as example): 155 MW(e) small fuel self-sustaining MSR: "FUJI-II", 7MW(e)

Pilot plant MSR: "mini FUJI-II', 1Gwe fuel-self-sustaining MSR: "super FUJI" [4,6]. FUJI-II will have fuel self-sustaining (near-breeder) characteristics even in small size, without core-graphite exchange and continuous fuel processing, which needs a huge R&D effort and investment, except the removal of Kr, Xe and T. The reactor is filled only with fuel-salt (ca. 10% vol) and graphite (ca.90% vol), which does not need to be exchanged during reactor life.

AMSB: The basic idea of AMSB was invented in 1980 depending on the "single-fluid type Molten-Salt target/blanket concept"[7], which is significantly simple and practical in structure. The target/blanket vessel is a simple pot of 4.5m in diameter and 7m in depth. A 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 solved by this design concept, except the proton-beam injection-port engineering which might be solved by the real beam test increasing intensity step by step and applying the gas curtain technology for example.

Technological rationality of THORIMS-NES: THORIMS-NES is a huge industry producing 1000-2000 TW(e) per year. In the development of this system, the following simple and rational nature of MSR technology should be recognized [2]:

> [A] no radiation damage in molten salt fuel and target/blanket, - chemical enert and stable glass forming
> [B] simple chemistry - highly predictable physico-chemical behaviors of molten salts - low R & D cost
> [C] simplicity in reactor design principle/configuration - commercialize from "small power stations"
> [D] widespread applicability of Na-FBR Technology results, hugely invested in the past, with the advantages of MSR Technology on the chemical enert, and low thermal shock.

These facts will guarantee the realization of THORIMS-NES in less than 20 years by very low R&D cost. Already ORNL has demonstrated an excellent 4 years operation of the experimental MSR named "MSRE" IN 1965-69[5]. The establishment of FUJI-II will b easily achieved in 12 years. AMSB will be developed in 15-20 years delaying a little, but it is enough because some difficulty to "initial ^{233}U fuel" can be solved by the following approach:
[F] easier commercialization by utilizing/eliminating commercial and weapon-head Pu [Chap.4].

4. PRACTICAL STRATEGY TO REALIZE TH-ENERGY ERA BY THORIMS-NES

Now the smooth and practical shift to Th-cycle: THORIMS-NES ERA from U-Pu Cycle ERA is the most important issue. After the termination of the Cold World War, this might be implemented easier that before including the effective incineration/elimination of weapon

materials and Pu, although such work should be performed inside a safeguarded area. Some detailed examination of this strategy has been reported in IAEA Tech.Comm.Meeting, Vienna, 1995[8] and others [9]. Here it will be briefly explained.

ORNL already demonstrated by means of their experimental reactor. MSRE that MSR can use any kind of fissile materials [10]. The only problem will be the solubility limit of Pu and TRU fluorides.

The complete elimination of Pu at present and in the future will be really established economically if we use the following strategy:

(1) D-plan: Pu (and trans-U elements [TRU] separation straightway in the form of molten fluorides by Dry-process from the spend solid fuels accumulating in the world. The technological basis has been examined by France, Russia and the Czech Republic and realized as the Russian FREGATE-project [11]. Here we need not reproduce any solid fuels.

(2) F-plan: Pu-burning and ^{233}U production by Fission MSR [FUJI-Pu], as explained already in Sec.4.1.

(3) A-plan: The same as the above (2) by AMSB-Pu, with F-plan, even delaying about 5-10 years.

Plutonium and TRU can effectively be transmuted by AMSB-Pu, producing ^{233}U in parallel in which the production ratio of ^{233}U to transmuted plutonium is much higher than the case of FUJIO-Pu [8].

Table I. The standard performance of FUJI-Pu [per 1 GE(e)] and AMSB-Pu [per 1 GeV 300 mA]

	Pu inventory	^{233}U inventory	Pu burnup/a	^{233}U production	Electr. output
FUJI-Pu	3t		0.86t	0.7t	1 GW(e)
FUJI-II		2t		self-sustain	1 GW(e)
AMSB-Pu lg	5t	0t	0.35	0.7t	-0.15 GW(e)
AMSB-PU hg	5t	5t	0.52	0.9t	1 GW(e)

However, the development of AMSB-Pu will be delayed than FUJI-Pu due to the large-current accelerator development and proton injection port engineering, although ASMB has significant technological advantages in the issues of radiation-damage, heat removal and reactor-chemistry.

U-Pu cycle system could not realize the energy production predicted in Figure 1(D) owing to the huge amount and steep increase. However, THORIMS-NES will be able to realize the following several scenario applying the above D-, F- and A-plans. Here, one of the simplest examples has been shown in Figure 2.

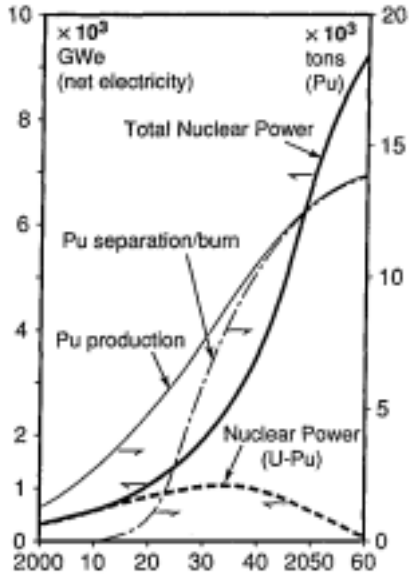

Figure 2. A scenario for THORIMS-NES deployment using plutonium incineration in the next century.

Tentatively the system size of U-Pu cycle power stations will be assumed as 4 times larger in maximum than the present. Even so low this will still produce more than 10^4 t plutonium (assuming 300KgPuk/GW(e) Y net) until 2050, which will be separated by Purex or D-plan process accompanying TRU in all the more proliferation-resistant mode, because a simple storage of spent-fuels will be a non real solution. Plutonium (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 2600 ton plutonium (TRU) or more. The latter will become 800 facilities in peak about 2040, burning about 10,000 ton plutonium (TRU) or more. The duty of FUJI-Pu will be finished until 2040. Now it can openly operate as proper $TH^{233}U$ power stations till the end of reactor life.

The technical development of AMSB-Pu will be significant in 2020 and 2040. The initial AMSB-Pu will be in lower grade (1g) not producing any outer electricity. Afterward the next high grade (hg) version will produce electricity improving in performance by near critical condition till the production of 1 or 2 GW(e)/facility [3].

After the middle of 2040 decade in which plutonium would be almost eliminated AMSB-Pu should be gradually dismantled, recovering ^{233} U fissile, which is useful to initiate FUJI power stations more. Therefore the main leading role of AMSB will be in the period of 30-40 years although afterward it will be continuously useful for radio-waste incineration as a flexible nuclear reaction facility [Figure1].

5. SIGNIFICANT ADVANTAGE IN SAFETY ISSUE

5.1 Basic Characteristics of MSR Safety

MSR, FUJI (and AMSB in general) is a significantly safe reactor, and has essentially "NO SEVERE ACCIDENTS". The most important safety performances are coming from the following factors:

(1) The primary and secondary systems are lower pressure than 5 bars, and do not have the danger of accidents due to high pressure such a system destruction or salt leakage.

(2) The fuel and coolant salts are chemically inert, and no firing or explosive with air or water.

(3) The boiling point of fuel salt is about 1670K or more, much higher than the operation temperature 973K. Therefore the pressure of primary system cannot increase.
(4) The fuel salt will be able to become just critical when it coexists with the graphite moderator. Therefore, leaked fuel salt will not induce any criticality accident. {EPI-thermal-type MSR is not the same.]
(5) MSR has a large prompt negative temperature-coefficient of fuel-salt. The temperature-coefficient of graphite is slightly positive, but controllable due to the slow temperature-increase depending on its high heat capacity.
(6) The delayed-neutron fraction in ^{233}U fission is smaller than that in ^{235}U, and half of the delayed-neutrons is generated outside the core. However, it is controllable owing to the longer neutron-life, and large negative prompt temperature-coefficient of fuel salt.
(7) As the fuel composition can be made up anytime if necessary, the excess reactivity and required control rod reactivity are sufficiently small, and the reactivity shift by control-rods is small.
(8) Gaseous fission such as Kr.Xe and T are continuously removed from fuel-salt, minimizing their leakage in accidents and in the chemical processing.

5.2. Basic Concept Securing the MSR Safety

For the confinement of radioactivity all reactor should have the following three safety functions:

[a] Reactor Shutdown Function: to stop (shut-down) the fission and to terminate the energy generation.
[b] Cooling Function of the Reactor: to keep the integrity of the fuel by providing enough cooling, and to prevent the release of radioactivity.
[c] Confinement Function of Radioactive Materials at Accident: to limit the release to the environment of radioactivity in the case of big accidents.

Besides the above, the concept of "Multiple Defence (Defence in Depth)" is adopted to assure the higher safety of the facility, taking in the following three different levels:

Level 1: Prevention of the abnormal situation when the reactor is operating: the reliability of equipment is raised sufficiently in design, manufacturing and maintenance.
Level 2: Prevention of the expansion of the abnormal situation: by the detection of abnormality in an early stage, by the plant inherent safety and by the reactor shut-down equipment.
Level 3: Prevention of the large release of radioactive materials: by setting up containment and ECCS. The multiple defence concept in MSR should be the same as LWR, and will be not touched more.

The above three safety functions [a], [b] and [c] in MSR will be explained as follows [12].

[a] Reactor Shutdown Function (Table II]:

All reactors should have inherent safety, which is achieved by suppression of power change by designing the reactor with a negative power coefficient. Because the temperature coefficient of fuel-salt is prompt negative and large, this condition is satisfied in MSR.

Control rods are also used for a rapid shutdown, and the fuel-salt drain system is also able to be used as another reactor shutdown function. Because the excess reactivity is small, the

number of control-rods is few and the diameter is large. The reliability will be high. The drain system is always necessary and effective on the pipe rupture accident. Since the fuel-salt falls to the drain tank by gravity through the freeze valve with a simple mechanism, its reliability is high. Although the freeze-valve operation may be slow, rapid response needs not due to no re-criticality.

Table II. Comparison of Reactor Shutdown Functions

demand function	LWR	MSR	merit on MSR
High Speed Shutdown System (Scram)	Control Rod	Control Rod	enough with small numbers
Second Shutdown System	Boric Acid Injection System	Fuel-Salt Drain System	no re-crificali1y in Drain Tank
Third Shutdown System		Fuel-Salt Composition Adjusting System	also used for makeup of Thorium Component

As a third measure, the adjustment of fuel composition using fuel-salt controlling system is possible to shutdown the reactor. One approach will be the Th addition, which is necessary to make up fuel-salt in any MSR, and again a slow action of this system does not cause any problem.

[b] Cooling Function of the Reactor [Table III].

In MSR, the possibility of piping rupture is very low due to the low pressure, and the ECCS will not need the same as FBR (Monju). It is possible to deal with the drain system, even if a piping rupture causes the fuel salt loss. Of course the decay heat removal system is necessary for the drain system.

Table III. Comparison of cooling functions of core in emergency

demand function	LWR	MSR	remark on MSR
Cooling Water make-up	ECCS	unnecessary	unnecessary (Drain System can be used as backup)
heat removal	Decay Heat Removal System	Decay Heat Removal System	for severe accident countermeasure

Table IV. Comparison of radioactive materials confinement functions

wall number	LWR	MSR	remark on MSR
1	Pellet	none (Liquid Fuel)	no LOCA, Gaseous Fission Products are removed always
2	Cladding	none (Liquid Fuel)	same as above
3	Pressure Vessel, Pipes	Reactor Vessel, Pipes	very low pressure
4	Containment	High Temperature Confinement	no Steam generation, no Flammable Gas generation
5	Reactor Building	Reactor Building	same as LWR

The MSR may have a capability of natural circulation when all pumps stop, because the pressure loss in the core is small. Detailed evaluation is necessary in the future. When natural circulation cannot be expected, or when a turbine system is isolated and the cooling by the secondary loop is impossible, the decay-heat removal system is necessary. As a final heat sink, the decay heat removal system by a static air cooler as in FBR is preferable to endure a long term severe accident, such as all AC power supply loos ("station black out") accident.

(c) Confinement Function of Radioactive Materials at Accident (Table IV]:

For this purpose, five barriers are applied in LWR. The first two barriers do not exist in MSR because MSR uses fluid fuel. The chance of radiation exposure by gaseous fission products (FP) is smaller due to their continuous removal from fuel-salt, and the danger of piping rupture is also very low. Therefore it is thought that the MSR safety is better than LWR.

The primary system of MSR is enclosed in a "high temperature confinement" and the entire reactor system is covered in the "containment" which is a reactor building itself. These arrangements are basically equal to the LWR. Since there is no water and no flammable gas generation, the MSR safety is excellent due to very few events which can threaten the integrity of containment.

5.3 Design Basis Accidents (DBAs)

Regarding the safety of MSR, accidents are categorized into two areas. The first is the so called DBAs (Design Basis Accidents) and the second is the severe accident which exceeds DBA.

DBAs are categorized into two events (A) initiated by dynamic equipment and (B) by static equipment. (A) is divided into two typical accidents (A1) and (A2), and (B) is divided into five (B1)~(B5) as shown in the following (2):

(A1) Fuel salt Flow Decrease Accident: In MSR, there is judgement that "the reactor is safe for the stop of all primary pumps, if an appropriate scram system is designed" [13]. One example of a scram system is a control rod drive located at the upper part of the core and control rods which will be inserted into the core by gravity, when an accident occurs.

(A2) Reactivity Insertion Accident (RIA): Although the added reactivity is small there is a possibility that the accident results are severe, because the effective β (= delayed neutron fraction) of MSR is only 0.1% ΔK(=1/5 of LWR). The reason is that β of ^{233}U is 0.26% which is about half of ^{235}U, and the half of β is lost when the fuel-salt flows outside the core.

Regarding the addition of a reactivity, mis-withdrawal of control-rod does not happen, because safety control-rods are always withdrawn when the reactor is in operation. Regarding the power increase by mis-insertion of the graphite control rod, it is small owing to the very small rod reactivity.

It might be a cold loop start up accident that the largest reactivity is added to the reactor as a reactivity insertion accident. It is an accident which can have a positive temperature reactivity coefficient when the stopped pump starts, and the fuel salt of relatively low temperature enters the core and then the absorption of neutrons by the Doppler effect becomes small. Since the reactivity insertion of 3 dollars (= 0.3% ΔK) is due to a 100^{o}C decrease of the fuel salt

temperature, this event is really a reactivity insertion accident. However, this event terminates by scram with the negative temperature coefficient of fuel-salt, although the fuel-salt temperature increases to some extent. In addition, since the prompt neutron lifetime of MSR is about 10 times longer than LWR, the power increase is mitigated for the prompt accidents.

(B1) Fuel Salt Loss Accident: The possibility of piping rupture is very low due to the low pressure and no steam existence. Meanwhile, it is possible to collect the lost fuel-salt to the drain tank (para.5.4].
(B2) Heat-transfer Piping Rupture Accident of HX (Heat Exchanger): In this case, secondary coolant salt enters the core side, because the secondary side contains higher pressure than the primary side. The boron content of coolant salt mixes with the fuel salt and the reactor stops.

(B3) Heat transfer Piping Rupture Accident of SG (Steam generator): In this case, it is necessary to evaluate the influence, because the steam of 200-250 bars flows into the secondary coolant-salt. However it is said that the molten salt does not cause a chemical explosion unlike Na, and therefore any serious influences on the primary system will not be induced.
(B4) Disruptive Accident in Off-gas System: Since MSR always removes gaseous FP from the primary system, off-gas treating facility accumulates a large amount of radioactive gas. Moreover, the cover-gas system of secondary loop accumulates Tritium generated from Li in fuel-salt, although T is transformed to water and easily controllable. Anyway, since it is a static facility unlike the main body of the reactor, correspondence is not difficult. Of course, countermeasures against the disruptive accident of off-gas systems are necessary.
(B5) Mis-operation of Fuel-salt Adjustment Equipment: This equipment is necessary in MSR to make up the salt components. It is necessary to design it so that a large amount of fissile materials is not inserted by this equipment. Since the inventory in this equipment is very small compared to that of cores, rapid reactivity insertion does not happen.

5.4 Severe Accidents

Based on the above review on DBAs, the following three main events are examined [12]:
(1) *Fuel-salt Flow Decrease Accident*: As a severe accident of MSR, it is necessary to assume *Scram Failure*, and *All Primary - and Secondary-Loop Pumps Stop*. Since ΔT (temperature increase between core fuel-salt inlet and outlet) is proportional to P/W (Power/Flow), temperature increase (reactivity decrease) by W becoming 1/10 from the related value and temperature decrease (reactivity increase) by P becoming 1/10 from the related value will balance.

By the way, if the speed of the pump of MSR is changed, it is possible to change the power output using the above phenomena, and this is one of the advantages of MSR.

As explained above, when the flow decreases, reactivity decreases by temperature rise, but a small positive reactivity is inserted by the increase of delayed-neutrons. This is because the delayed-neutron precursors, taken away outside the core, stay in the core.

Shimazu concluded by a quantitative analysis [13] that “The flow decreases to a power level of about 10% after 10 seconds, according to the analysis assuming that the flow becomes zero when all pumps stop. The exist temperature rises from 973K to 1170K”.

In an actual situation, it is necessary to remove the decay heat. If both the primary and secondary loops circulate naturally, the decay heat of the core can be exhausted outside the rector. When natural circulation is impossible, the decay-heat removal system is actuated (para.5.2 [b]). Therefore it is safe enough even if both the primary loop pumps and the secondary loop pumps stop in a severe accident case.

There is a *Flow-Path Plugging* with debris, which is one of the other scenarios of the flow reduction. This scenario is reviewed relating with MSBR [5] and it says, "If the fuel salt temperature reaches the boiling point, there may b a problem caused by a positive void coefficient. But there are hundreds of channels in a core, and even if 100% void happens at 20 channels simultaneously, void reactivity is only 1$. In addition there is an effect that the fuel itself disappears, and it is unlikely to become a problem. However, further examination is necessary".

(2) *Reactivity Insertion Accident*: It might be a *Cold Loop Start-up Accident* that the largest reactivity insertion is forecast as a reactivity insertion accident. Since the temperature coefficient of the fuel salt is about -3×10^{-5} $\Delta K/K/^{o}C$, the inserted reactivity is about 3$ (0.3% ΔK), because the fuel salt temperature decreases about 100 ^{o}C.

This scenario is calculated on MSBR by Shimazu [14] and it says, "At zero-power or full-power condition, 3$ reactivity insertion with scram failure assumption, the fuel-salt negative temperature coefficient mitigates the event, and the highest fuel-salt temperature is 1473 K. This temperature is lower than the melting point of Hastelloy N (1640 K), assuming that the temperature of the core vessel is the same as the temperature of the fuel-salt." Therefore the MSR has enough safety for the reactivity insertion accident.

(3) *Fuel Salt Loss Accident*: Basically, the *Fuel Salt Loss Accident* happens only as a severe accident. As a result of any pipe rupture accidents in MSR it is possible to terminate the accident if the system is designed to collect the lost fuel-salt into the drain-tank. Also it is necessary to design the drain tank system using natural heat radiation in order to endure a long-term cooling of the decay-heat. Since the fuel-salt becomes a solid (a stable glass) below the melting point at a final stage, it is not necessary to consider a so-called China Syndrome. If the drain system with natural heat radiation is designed, the integrity of containment is secured. Therefore, in MSR, it is possible to prevent the worst severe accident scenario such as the containment failure = China Syndrome = a large amount of radioactivity release.

Moreover, the *Re-Criticality Accident* does not occur. This depends on the fact that concentration of fissile material in the fuel-salt is low, and the fuel salt does not become critical without an appropriate amount of moderator such as graphite.

In addition, since the gaseous fission products are always collected in MSR, the amount of radioactivity release is small, even if there is a radioactivity release accident.

6. ADVANTAGE IN NUCLEAR-PROLIFERATION ISSUE

THORIMS-NES brings high proliferation-resistant nuclear fuel cycles to the world through covering fissile material in the near future from Pu to ^{233}U. Advancements in proliferation-resistance will be observed in the following three view points [15]:

(1) Macroscopic View in Global Fuel Cycles: plutonium in spent fuels of various thermal; reactors are steadily increasing in the world. Especially vast amounts of them are expected in developing countries in the near future through the promotion of nuclear power generation, mostly with LWRs.

Plutonium brings proliferation risks even when it remains in spent fuels. They should be subject to more stringent safeguards compared to new fuel made from low enriched uranium. But there is always the risk of theft or diversion, especially in the case of solid spend fuel, which is easier to handle than liquids or solidified fuel-salt of MSRs. Even if spend LWR fuels would be disposed in a deep geological stratum, they might form a potential future plutonium -mine because radioactivity of fission product decays out in a long time.

However, when spent LWR fuels are reprocessed from the reasons of waste volume reduction or the issue of energy resources - that will be very likely - proliferation risks will further increase unless we have a good scheme for utilizing separated plutonium. When the plutonium is used again in LWRs, i.e. LWR-MOX cycle the problem will not be much changed from the usual LWR cycle and remain unsolved. On the other side, if the plutonium is used in FBR cycle it will bring more issues, to be described in the following paragraph (2).

So, thorium fuel cycle development through plutonium incineration by THORIMS-NES is the best scheme we have for this purpose, since it actively reduces and simultaneously suppresses new production of spent fuels containing plutonium.

Plutonium utilization in MSR which brings power generation and converted ^{233}U simultaneously might be the only possible way to let effective use and nonproliferation of nuclear materials be compatible, because it has the following advantage over FBR fuel cycle. Therefore THORIMS-NES would be able to make a great macroscopic contribution to global fuel cycles.

(2) Plutonium vs. ^{233}U (FBR vs MSR): Significant quantity (SQ) in nuclear safeguards is not so much different between plutonium (8kg as element-total) and ^{233}U (8 kg in isotope) but diversion resistance will be significantly larger in ^{233}U.

One core fuel assembly for FBR usually contains about 1 SQ of plutonium and it is rather small and easy to handle and conceal for diversion or theft. Blanket fuel assembly for FBR has lower plutonium concentration than core assembly, and several blanket assemblies are required to get 1 SQ of plutonium. But their plutonium is very near to the weapon grade and attractive to the potential divertor. On the other hand, fissile material concentration in MSR fuel is low as is described in (3), and it is difficult to get 1 SQ because of the required large amount (1-2 tonnes) and the inconvenient form for theft. Moreover, plutonium in MSR-Pu is usually too old for weapon use and ^{233}U accompanies strong radiation as described below.

^{233}U usually contains more than 500 ppm ^{233}U and its daughter nuclides, some of which emit strong high energy (208I1 2.6 MeV) gamma rays. They bring lethal doses of 1-2 Sv/hr at 50 cm distance from 1 SQ (8 Kg) ^{233}U. To shield it more than 20 cm thick lead is necessary, which emit strong high energy ($^{208}I1$ 2.6 MeV) gamma rays. They bring a lethal dose of 1-2 Sv/hr at 50 cm distance from 1 SQ (8 Kg) ^{233}U. To shield it more than 20 cm thick lead is necessary, which in fact makes it impossible to steal and fabricate nuclear explosives.

To procure pure ^{233}U it is necessary to separate its precursor ^{233}Pa. However the separation of dilute ^{233}Pa is chemically not easy work, and its half-life lasts only 27 days.

^{233}U can easily be denatured by adding ^{238}U if required. Even in this case ^{238}U concentration in MSR fuel is maintained fairly low, about 1/10 of the main fertile material thorium, because of low concentration of ^{233}U. This prevents not only to spoil the nuclear characteristics but also to produce Pu and higher nuclides [Am, Cm etc.], which have the potential to easily become weapon material. This liberation from TRU elements is the great merit of Th- ^{233}U fuel cycle, and the U-Pu fuel cycle never gets out of this yoke.

FBR fuels must be recycled in fairly short periods to retain their breeding power at a practical level. So annual throughput of plutonium in FBR fuel cycle will become very large and bring significant safeguards and transportation problems. Required plutonium inventory in one FBR (1 GW(e)) is several tonnes of plutonium, for example, 1% of them becomes several SQ. Hold-up of the order of 1% will be apt to occur.

The situation in MSR/THORIMS-NES is much easier, because the ^{233}U.inventory in MSR is about ¼ of plutonium in FBR and it will become effectively fuel self-sustaining near breeder. These will result in few transportation occasions and little fissile material throughput.

(3) Microscopic View in reactor Site: Fissile material concentration in MSR fuel is low in both cases of MSR-Pu and MSR-^{233}U, and the typical concentration will be about 1 wt% of them. Therefore the fuel salt containing 1 SQ (8 kg) of plutonium or ^{233}U weighs 800 Kg with the volume of about 250 litres. In practice these fissiles will be dirty and need larger amounts of salts. This makes theft effectively impossible.

MSR does not have large excess reactivity. So even when a diversion by the operator is made, the fact can easily be detected b the inspector. This will be effective to deter theft. MSR has a further merit in that it has only a little additive fuel and spent fuel at its site.

High gamma dose level of ^{233}U cycle fuel serves also to provide easy detection of the irregular tranfers in the normal fuel handling route. In case of FBR there is a proposal to intentionally add radioactive TRUs into plutonium. But in the case of ^{233}U the radioactivity accompanies naturally, and it brings no obstacle in nuclear characteristics of the reactor.

Reprocessing and re-preparation of MSR liquid fuel is simpler and easier than those of FBR solid fuel. This will reflect the possible difference of theft and diversion between the two reactor types. Transportation - the vulnerable point in fuel cycle - can also be much reduced in MSR, because it is principally a self-sustaining "*Near Breeder*" and it usually has on-site processing and re-preparation of the fuel. These advantages can similarly be held in he case of AMSB (accelerator molten salt fissile producer). AMSB and the fuel-salt processing facilities will be non-utility/process plants in essence, and will be accommodated inside Regional Centers heavily safeguarded. This separation plan of the breeding facilities from the very little consuming power stations is a good management scheme of nuclear materials.

To summarize the above, it should be strongly recommended to convert plutonium to "the hardest and least desirable fissile material for weapon - ^{233}U through MSR-Pu and gradually to shift to MSR- ^{233}U fuel cycle on a global scale. The effectuation of the Comprehensive Test Ban treaty makes it impossible to make the explosive test for the ^{233}U bomb development. This condition also suggests a more safe world using ^{233}U than plutonium.

7. IMPROVEMENT IN SOCIAL ACCEPTANCE

The nuclear energy community is suffering serious criticisms from the public not only on safety, radio waste and nuclear proliferation issues, but also inflexibility/instability in public relations. This depends mostly on the influence of the past *Cold world war*. Now a new Nuclear Era should be reconstructed following the recommendation of the late David E. Lilienthal encouraging "a revival of its positive, affirmative fighting spirit" of scientists [16].

For such purpose the THORIMS-NES will be able to contribute as shown in the following lists:

Socio-Philosophical Advantages of Thorium Molten-Salt Nuclear-Energy Synergetic System [THORIMS-NES]

Notation:

(Q): Old Development Philosophy based on *Current Nuclear-Energy* technology approach
[A]: New Development Philosophy based on THORIMS-NES approach

(Q1) Introduction of "Controlled Society" derived from "Controlled Management of Nuclear Materials.
[A1] Normal Society protected by **enhanced resistance to Nuclear Proliferation/Terrorism** depending on Th-U Fuel Cycle: elimination of Pu & Trans-U elements, and intense 2.6 MeV gamma of ^{232}U.

(Q2) Huge Protection Work on **Radioactive Exposure**
[A2] Wide application of Remote Operation/Maintenance, Curtailed Maintenance, Handling and Processing of Fuels & Radio-Wastes based on Fluid-Fuelled Reactor: Molten-Salt Reactor.

(Q3) Comprehensive Restraint to achieve **"Material Quality-Control"** and "Operation/Controllability" for Hazard-Protection.
[A3] Fundamental "Reactor Safety" enhancement such as **"No Severe Accident**": no core melt down, no re-criticality, restricted radio-activity release, and resistance to military attacks or sabotage.

(Q4) Burden of Future Generation: Radio-Waste Management for centuries and millennia.
[A4] No Production of Pu, Am, Cm [Trans-U elements] limited Dilution of High-level Radio-Waste and minimized Amount of Low-Level Radio-Waste due to reduced Maintenance/Process Works [cf.[17]).

(Q5) Large Efforts and Emphasis on **R&D** to facilitate Political Control, Monopoly, Power Centralization.
[A5] "Short Term", "Low Cost" and "Simple (few items, esp. in fuel development)" **R&D Program**, based on "Nuclear Chemical Engineering [liquid medium]" Principle of Nuclear Energy System.

(Q6) Big Complex Science: Non or Costly testing for elaborate **System Size and Sophistication.**

[A6] Simple, small and Testable [no Severe Accident] **Power Stations** owing to Separation of Power Reactors and Fissile-Producers, denying "Fission Breeder Power Reactor" concept.

(Q7) Compelled "Public Acceptance" from the side of Nuclear Energy Promoters.
Loss of Individuality and Persona Liberties, and **Human Estrangement.**
[A7] Return to Original **Scientific Spirit,** and should prepare a really safe, flexible and economical "PUBLIC INDUSTRY', depending on rational/practical Principle of Nuclear Energy Technology.

It will be optimistic, but we need such technology. And the THORIMS-NES concept is young and will have potential for further improvement. Therefore the above will be recognized as a promising target of our effort. We have to proceed for preparing the future "**Open Society**".

8. CONCLUSIONS

The biggest handicaps to the Th-MSR concepts originated by ORNL are the unbelievable excellence in the scientific and technological basis, not requiring significant money and personnel, and resulting in no accident during the MSRE project, 30 years ago. Although all their results had been published, it is not easy to obtain those.

Now we have to start improving he excellent ORNL results to the most suitable form in the next century. The most effective first measure will be the demonstration of integral MSR technology by the simple pilot-plant: 7MWe miniFUJI with a reactor-vessel size of 1.8 m diameter and 2.1 m high [4,6].

As a conclusion, the following final sentence in the last book, **"Atomic Energy: A New Start",** by David E. Lilienthal [16], a notable American will be shown in the hope that our work might be useful as a trial reply to his sincere wish:"**What I have reflected upon and written about is not merely a new source of electrical energy, nor energy as an economic statistic. My theme has been our contemporary equivalent of the greatest of all moral and cultural concerns - fairness among men and the endless search for a pathway to peace."**

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A PROLIFERATION RESISTANT FUSION BREEDER FOR THORIUM FUEL CYCLE*

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Abstract. The fissile breeding capability of a (D,T) fusion-fission (hybrid) reactor fueled with thorium is analyzed to provide nuclear fuel for LWRs. Three different fertile material compositions are investigated for fissile fuel breeding: 1) ThO_2 2) ThO_2 denaturated with 10 % natural-UO_2 3) ThO_2 denaturated with 10 % LWR spent fuel. Two different coolants (pressurized helium and Flibe "Li_2BeF_4") are selected for the nuclear heat transfer out of the fissile fuel breeding zone. Depending on the type of the coolant in the fission zone, fusion power plant operation periods between 30 and 48 months are evaluated to achieve a fissile fuel enrichment quality between 3% and 4%, under a first-wall fusion neutron energy load of 5 MW/m^2 and a plant factor of 75%. Flibe coolant is superior to helium with regard to fissile fuel breeding. During a plant operation over four years, enrichment grades between 3.0% and 6% are calculated for different fertile fuel and coolant compositions. Fusion breeder with ThO_2 produces weapon grade ^{233}U. The denaturation of the ^{233}U fuel is realized with a homogenous mixture of 97% ThO_2 with 3% natural-UO_2 as well as with 3% LWR spent nuclear fuel. The homogenous mixture of 97% ThO_2 with 3 % natural-UO_2 can successfully denaturate ^{233}U with ^{238}U. However, at the early stages of plant operation, the generated plutonium component is of weapon grade quality. The plutonium component will be denaturated after a plant operation period of 24 and 30 months in Flibe cooled and gas cooled blankets, respectively. On the other hand, the homogenous mixture of 97 % ThO_2 with 3 % LWR spent nuclear fuel remains non-prolific over the entire period for both, uranium and plutonium components. This is an important factor with regard to international safeguarding.

1. INTRODUCTION

Presently, light water reactors (LWRs) supply the increasing demand on nuclear energy production, followed by Canada deuterium uranium (CANDU) reactors. LWRs require substantial quantities of slightly enriched (3-4 %) nuclear fuel over their operating lifetime of 30 to 40 years. Sooner or later, this energy strategy would lead to a bottleneck in the provision of the nuclear fissile fuel in addition to the generation of substantial quantities of spent fuel as nuclear waste.

Only non-fissile (external) neutron sources can supply the required quantities of low-enriched nuclear fuel for the continuing nuclear energy production based on the well-established LWR-technology. The idea of the production of abundant fissile fuel through fusion breeders or electro-nuclear breeders is quite old [1-8]. Studies show that a fusion breeder can produce up to 30 times more fissile fuel than a FB per unit of energy. Typically for a hybrid reactor with suppressed fission [5],

$$\frac{\left(\frac{BR-1}{E}\right)_{HR}}{\left(\frac{BR-1}{E}\right)_{FB}} = \frac{\left(\frac{1.8-1}{27}\right)}{\left(\frac{1.2-1}{200}\right)} = 30 \text{ will be obtained.}$$

* 1998 meeting.

World thorium reserves are estimated to be about three times more abundant than the natural uranium reserves. The known thorium reserves in Turkey exceed 300.000 tonnes. Hence, thorium as future nuclear fuel source has special importance for Turkey.

Early work has investigated the possibility of ^{233}U in a fusion-fission (hybrid) reactor [9-13]. However, nuclear fuel, produced in fusion breeders can become of nuclear weapon quality with ^{239}Pu or ^{233}U in the fissile components. Hence, considerations for the denaturation of these new nuclear fuel sources become very important [14]. The subject of the present work is to analyze the breeding potential of a thorium fusion breeder with inherently enhanced protection precautions against nuclear weapon proliferation. Calculations are conducted using a (D,T) fusion neutron driver for the hybrid reactor.

2. BLANKET GEOMETRY

For the reasons of consistency and comparison of data with previous work, the neutronic analysis is performed on an experimental hybrid blanket geometry, which was presented to the international scientific community on different occasions [10-20]. Figure 1 shows the basic structure of the hybrid blanket adopted in this work. This geometry is not a representative of one of the mainline fusion reactor design concepts. However, the geometry of the blanket is not a crucial criterion for the generic investigations within the framework of the present work. In this concept, a line neutron source in a cylindrical cavity simulates the fusion plasma chamber. A first wall made of 304-stainless steel surrounds the latter. Recent work has shown that a SS-304 first wall without Mo and Nb components would give a C-class nuclear waste material in fusion reactors, after a plant life time of 30 years which will be suitable for shallow burial after the decommissioning of the reactor [21-22]. However, a first wall made of SS-304 can be selected only if water is not used as coolant material. Because Mo and Nb is needed in stainless steel mainly for corrosion resistance against of water. As the blankets selected in this work do not use water as coolant, SS-304 is to be preferred instead of SS-316.

In the present study, for the purpose of denaturation, the fissile zone is made of fissile fuel breeder rods with two different compositions:

1) ThO_2 denaturated with natural-UO_2.

A fusion breeder with fast neutron spectrum in the fissile zone produces highly prolific fissile fuel. In a fusion breeder with ThO_2 alone, the isotopic percentage of ^{233}U would be close to 100 %. Hence, precautions to denaturate the fissile fuel are an important issue for a fusion breeder. An easy way of denaturing ^{233}U can be possible by mixing it homogeneously with natural uranium. ^{238}U reduces the weapon grade quality of ^{233}U. However, while ^{238}U can denaturate ^{233}U, it will be partially converted into ^{239}Pu in the hybrid blanket so that the key issue for such a MOX fuel composition would be shifted to the denaturization of ^{239}Pu with even plutonium isotopes.

2) ThO_2 denaturated with pressurized LWR spent fuel with plutonium recycle after a burnup grade of 33000 MW·d/MT [23].

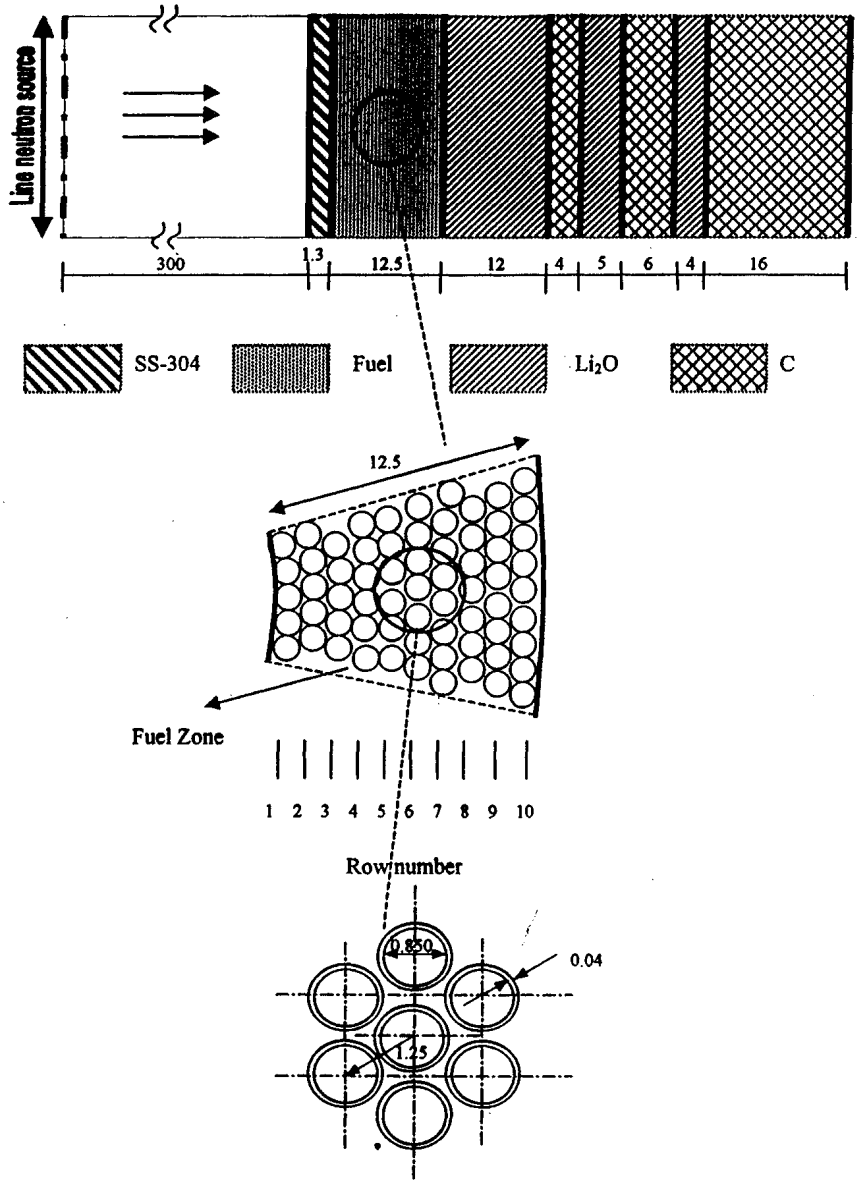

Figure 1. Cross sectional view of the investigated blanket (dimensions are given in centimeters)

Natural uranium contains only 0.7% of ^{235}U as a fissile component. Thermal reactors have a modest conversion ratio (0.6-0.7 for LWR), so that the plutonium production becomes relatively modest. Hence, LWRs can exploit only about 1% of the natural uranium fuel and still 99% of the fissionable natural uranium resources are not being used for energy production. Furthermore, LWRs produce great amount of actinides as nuclear waste, which is one of the nuisances of present day reactors. Previous work has highlighted the possibility and advantages of the utilization and also regeneration of spent nuclear fuel of critical reactors in fusion-fission (hybrid) reactors. It has been analyzed in detail and shown that the burn-up of the nuclear waste actinides in fusion reactor blankets and the rejuvenation of the spent nuclear fuel for multiple recycling in critical nuclear reactors have a solid background and realistic prospects with regard to neutron physics [15-20]. This suggests to investigate the potential of spent nuclear fuel for denaturing a thorium fusion breeder, as light water reactor spent fuel with plutonium recycle will contain, already at startup, sufficient even uranium and plutonium isotopes to denaturate both fissile fuel components.

In the fissile zone of a fusion breeder, the nuclear heat production will tend to have an exponentially decreasing character in the fusion neutron source driven medium. A non-uniform fission power density is the major source for temperature and radiation gradients and for complicated fuel shuffling scenarios. On the other hand, a quasi-constant fission power generation has several advantages, such as reduced material stresses, uniform exploitation of the fissile zone, higher fuel burn-up grades, etc. An elegant and easy way of fission power flattening is possible by increasing Σ_f in radial direction in order to compensate the decrease of the neutron flux (Φ) to realize a constant fission rate ($\Sigma_f \cdot \Phi$) over the entire fissile zone.

To obtain a quasi-flat nuclear heat generation, the UO_2 (or LWR spent fuel) fraction with a higher neutron multiplication has been increased in radial direction, on the cost of the ThO_2 fraction with a lower neutron multiplication. The UO_2 (or LWR spent fuel) volume fraction in the fuel rods has been increased gradually, with increasing row number (see figure 1) from 0.5 %, 1%, 2 % to 5 %, 10 %, 20 %. Table I shows the investigated fuel zone models for different mixed fuel composition by a volume fraction of $V_c/V_f = 2$ (coolant and fuel, respectively), where the fuel zone is cooled with pressurized helium which occupies a volume fraction of 62.6 %.

The radial reflector is made of Li_2O and graphite in sandwich structure. This measure reduces the neutron leakage drastically and leads to a better neutron economy [15].

3. NUMERICAL RESULTS

3. 1. Calculational Methods

The neutronic calculations have been performed by solving the Boltzmann transport equation with the neutron transport code ANISN [24] by using the neutron transport cross sections of the CLAW-IV data library [25] and the activity cross sections of the data library TRANSX-2 [26]. The integration of the angular neutron flux has been done in S_{16}-P_3 approximation by using Gaussian quadrature sets [27] to obtain a high accuracy. ANISN results have further been processed with the help of the auxiliary code ERDEMLI [28].

Table I: Variation of the mixed fissile/fertile fuel fraction by volume in radial direction in the fuel zone.

Coolant:	Helium		FUEL ZONE									
			Row 1	Row 2	Row 3	Row 4	Row 5	Row 6	Row 7	Row 8	Row 9	Row 10
MODEL 1 (ThO_2 + LWR)	%ThO_2	a	99.5	99	98.5	98	97.5	97	96.5	96	95.5	95
	%LWR		0.5	1	1.5	2	2.5	3	3.5	4	4.5	5
	%ThO_2	b	99	98	97	96	95	94	93	92	91	90
	%LWR		1	2	3	4	5	6	7	8	9	10
	%ThO_2	c	98	96	94	92	90	88	86	84	82	80
	%LWR		2	4	6	8	10	12	14	16	18	20
MODEL 2 (ThO_2 + UO_2)	%ThO_2	a	99.5	99	98.5	98	97.5	97	96.5	96	95.5	95
	%UO_2		0.5	1	1.5	2	2.5	3	3.5	4	4.5	5
	%ThO_2	b	99	98	97	96	95	94	93	92	91	90
	%UO_2		1	2	3	4	5	6	7	8	9	10
	%ThO_2	c	98	96	94	92	90	88	86	84	82	80
	%UO_2		2	4	6	8	10	12	14	16	18	20

Temporal effects in the blanket are evaluated for a neutron energy flux load of 5 MW/m^2 on the first wall. The plant factor (PF) is taken as 75%. For neutronic calculations, the fuel zone is divided into 10 equidistant subzones, which corresponds to the ten fuel rod rows.

The temporal change of the fuel composition during hybrid reactor plant operation is evaluated for discrete time interval Δt, as explained in ref. [19]. The nuclear quality of the fuel has been followed in each fuel rod row individually while considering variations in the neutron spectrum and in the atomic densities of all fissionable isotopes over the radial coordinate within the fissile zone $\Delta t = 15$ days.

3. 2. Overall Performance of the Blankets

Table II shows the most pertinent integral neutronic data for the investigated cases at the beginning and at the end of a plant operation period of 48 months (4 years). At start-up, the blankets are barely self-sustaining with respect to tritium breeding (TBR > 1). However, in the course of plant operation, tritium production in lithium (mainly in ^{6}Li isotope) increases almost linearly, caused by an increase of the neutron population due to the accumulation of fissile fuel in the blanket. After a plant operation period of 1 year, TBR exceeds 1.05 for sufficient tritium supply.

Table II: Pertinent integral neutronic data in the blanket

	Model 1a		Model 1b		Model 1c		Model 2a		Model 2b		Model 2c	
Time	0	48	0	48	0	48	0	48	0	48	0	48
TBR	1.005	1.197	1.011	1.207	1.024	1.223	1.001	1.195	1.010	1.205	1.020	1.219
$\nu\cdot\Sigma_f$	0.156	0.570	0.168	0.582	0.191	0.608	0.154	0.573	0.163	0.577	0.183	0.603
Σ_f	0.045	0.203	0.048	0.206	0.055	0.213	0.044	0.204	0.047	0.204	0.053	0.211
M	1.956	4.011	2.003	4.058	2.098	4.157	1.947	4.025	1.984	4.030	2.062	4.129
^{232}Thγ	0.313	0.321	0.306	0.313	0.293	0.299	0.314	0.322	0.304	0.310	0.290	0.296
^{238}Uγ	0.0070	0.0073	0.0141	0.0146	0.0287	0.0296	0.0080	0.0083	0.0160	0.0165	0.0324	0.0334
Γ	1.463	1.179	1.383	1.167	1.249	1.142	1.477	1.177	1.411	1.176	1.293	1.150
L	0.065	0.075	0.065	0.076	0.065	0.076	0.064	0.075	0.064	0.075	0.065	0.076

TBR: Total tritium breeding ratio
Σ_f: Integral fission rate
$\nu\cdot\Sigma_f$: Integral fission neutron production
^{232}Thγ: ^{233}U breeding ratio
M: Blanket energy multiplication
^{238}U□: ^{239}Pu breeding ratio
Γ: Peak-to average fission power density ratio in the fuel zone
L: Radial neutron leakage fraction

Total energy generation in the blanket can be expressed with the help of the energy multiplication factor M. One can observe a doubling of M over 4 years of plant operation, also caused by an increase of the fission events due to the accumulation of fissile fuel in the blanket. This increase is relatively low for a gas cooled hybrid reactor with a (D,T) driver. Note that a quasi-invariable energy production over the operation period is essential to exploit the installed non-nuclear island of the plant (turbines, generators, heat exchangers, etc.) at an optimal level.

The spatial non-uniformity of the fission energy density, defined with the help of peak-to-average fission power density ratio "Γ" decreases with time. This is a very favourable effect with regard to a uniform exploitation of the fissile zone in the blanket, and is a direct result of the flattening procedure.

The radial neutron leakage out of the blanket "L" increases slightly, again caused by an increase of the neutron multiplication in the blanket.

3. 3. Fissile Fuel Breeding

The quality of the nuclear fuel can be measured with the help of the so called cumulative fissile fuel enrichment (CFFE) grade which is the sum of the isotopic percentages of all fissile isotopes, in our case mainly that of ^{233}U, ^{239}Pu and ^{241}Pu. Figure 2 shows, for model 1c, the variation of the CFFE over a total operation period of t = 48 months for selected fuel rods. The spatial variation of the temporal growth of CFFE throughout the fuel zone is more or less uniform, except for row # 10. In all other investigated models, CFFE show a similar, but more uniform behaviour for different fuel rods.

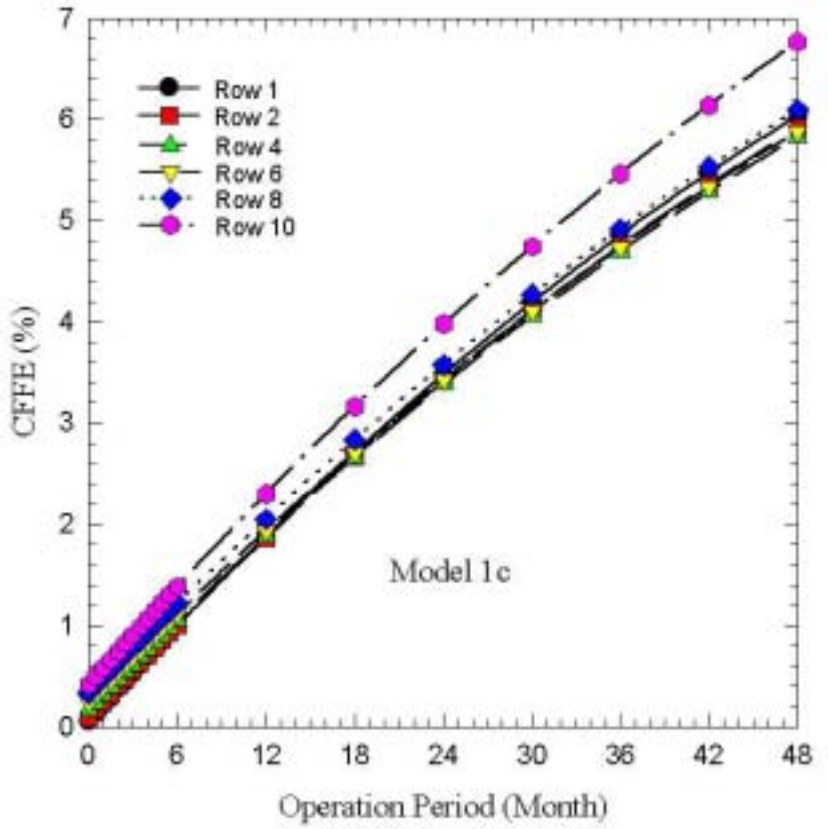

Figure 2. Temporal increase of the fissile fuel quality in the blanket

As the neutrons penetrate through the fuel zone towards the Li_2O and reflector zones, the neutron spectrum will be shifted to lower energy regions. The softening of the neutron spectrum at the outer regions of the fissile zone will increase the (n,γ) absorption rates in the resonances and thermal energy region in the fertile isotopes ^{232}Th and ^{238}U so that the production of ^{233}U and ^{239}Pu begins to rise towards the periphery of the fuel zone despite a radial reduction of the neutron flux.

The fuel quality exceeds CFFE>1% and >2% within <6 and <12 months, respectively. This would make it suitable for utilization in advanced commercial HWRs. Recent studies indicate that the burn-up rate in a CANDU reactor can be increased substantially if the fuel charge is slightly enriched [29], namely up to 1 to 1.5 %. An advanced CANDU breeder concept with thorium fuelling would require an average enrichment level of 1.5 to 2.0 % at start-up [30] to

realize a commercially reasonable breeding capability of ^{233}U from ^{232}Th. This would open new dimensions for the well established CANDU reactor technology.

A CFFE value between 3 and 4 % can be reached after an irradiation period of about 24 months, see Figure 2, suitable for utilization in LWRs. Higher CFFE values would allow to increase the burn-up rate in LWRs, substantially.

3. 4. Fuel Burn-up Grade

During the plant operation, some of fissionable fuel produces energy in the hybrid blanket *in situ*, along with the spent fuel rejuvenation. This can be measured with the help of fissile fuel burn-up. The calculational procedure of the average burn-up of the fissile fuel in a hybrid blanket had been described in chapter III. E, in ref. [19].

Figure 3 shows the fissile fuel burn-up in the hybrid blanket for a neutron energy flux of 5 MW/m^2 at the first wall with a plant factor of 75% as a function of plant operation period. The burn-up levels after 4 years can reach those observed in LWRs.

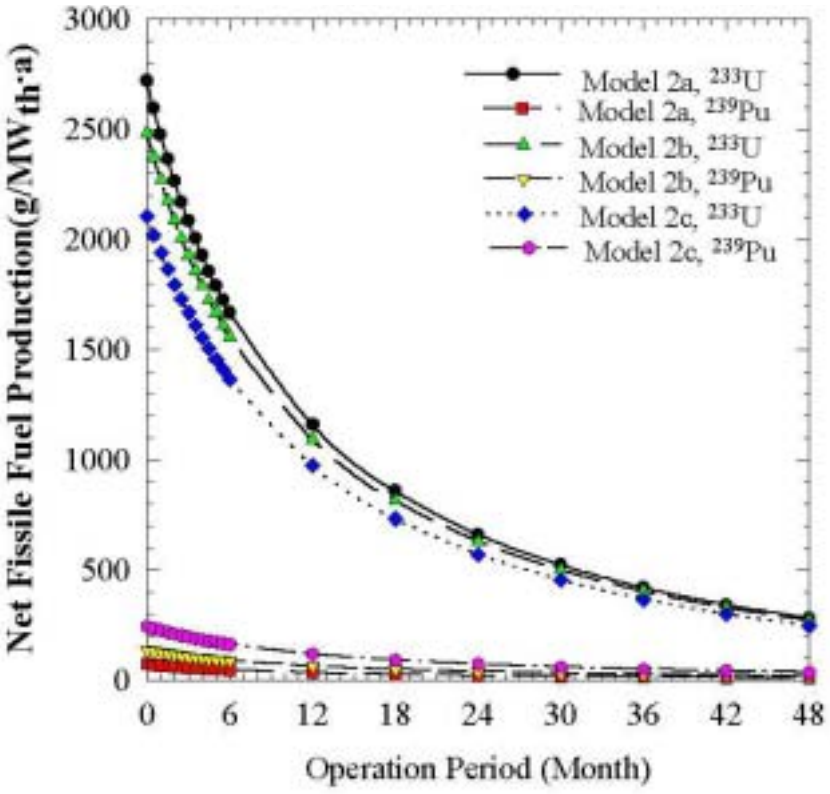

Figure 3. Temporal increase of fissile fuel burn-up in the blankets

3. 5. Figure-of-Merit

The fuel breeding ability of hybrid reactors can be defined in form of a figure-of-merit (FOM) as the ratio of the amount of net ^{233}U and ^{239}Pu mass generated to the fission power output of the hybrid blanket. The mathematical formulation of FOM in net ^{239}Pu [g/(MW(th)a)] or [kg/(GW(th)a)] has been described in chapter III. F in ref. [19], and will not be repeated here.

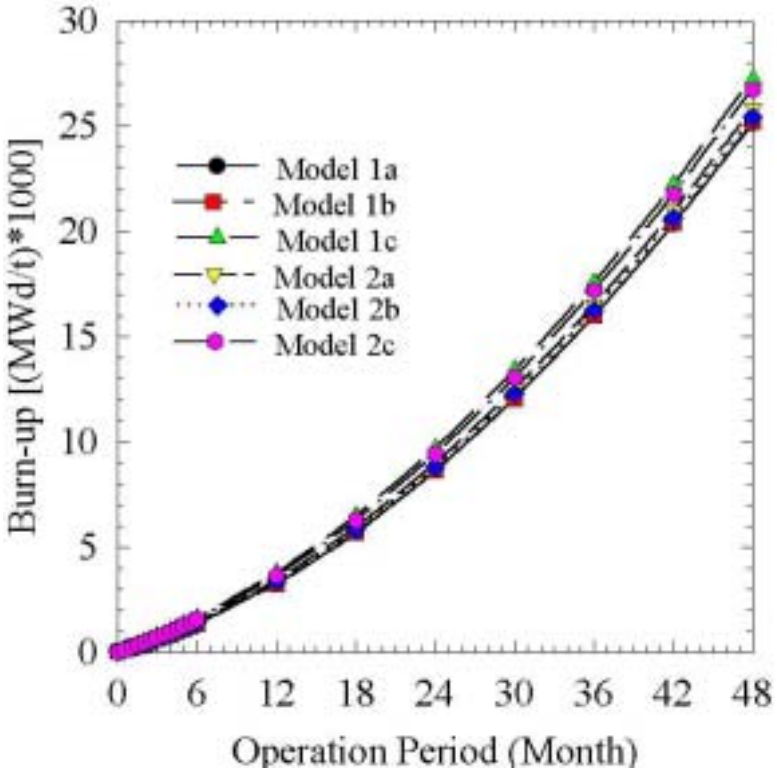

Figure 4. Temporal variation of net fissile fuel production in grams per MW(th) fission energy release in the blankets

Figure 4 depicts the FOM values for the models 2a, b, c. Models 1a, b, c show similar behaviour. At start-up, the FOM values are high. In the early stages of power plant operation, there is a rapid accumulation of the fissile fuel. This causes a faster increase of the fission power production in the reactors, and burns the fissile fuel more efficiently in the blanket, *in situ*. In all cases, FOM values decrease during plant operation along with an increased fission burn-up, which leads to an increase of the blanket energy multiplication M in Table II.

One can recognize in figure 4 that the breeder produces mainly ^{233}U as fissile fuel. However, despite of the low UO_2 percentage in the MOX fuel, there is also substantial plutonium production -in the range of tens to hundreds of kg/(GW(th)a)- which needs special attention for safeguarding.

3. 6. Safeguard Aspects of the Fissile Fuel

A major point of this work for thorium cycle is focused on the prolific aspects of the fissile fuel in a fusion breeder. Figure 5 shows the temporal variation of the percentages of the uranium isotopes in the first row with the lowest LWR spent fuel or natural UO_2 fraction. The ^{238}U content is burnt up, along with the accumulation of ^{233}U. In models 1a and 2a the small ^{238}U fraction in the first row is converted rapidly into ^{239}Pu so that ^{233}U fuel (with > 80%) may be considered prolific after ~ 12 months of plant operation. Models 1b and 2b become prolific after ~ 30 and 36 months, respectively. Whereas in models 1c and 2c, the ^{238}U content (> 2% all over) is sufficient to denaturate the ^{233}U fuel successfully.

On the other hand in models 1a, 1b, 2a, 2b, only those fuel rods which contains a natural-UO_2 fraction > 2 % (R > 304 cm), ^{233}U fuel remains fully denaturated (^{233}U < 80 % even after 48 months), as it can be observed in figure 6. In figure 6, the isotopic composition of uranium is plotted as a function of the radial dimension in the fuel zone for model 2a. The MOX fuel starts with 0.5 % nat-UO_2 in the rods on the left side and ends up with 5 % nat-UO_2 on the right side. While the MOX fuel in the 1st and 2nd row may be still within the classification of prolific nuclear materials, it can be considered sufficiently denaturated beyond the 3rd row, as shown in Figure 7.

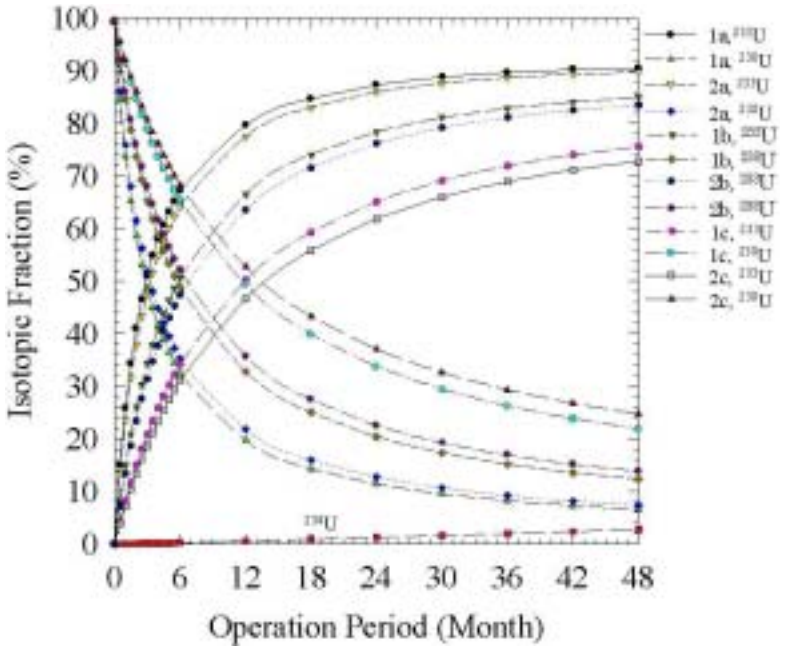

Figure 5. Temporal variation of the percentages of the uranium isotopes in the blankets (in the first row)

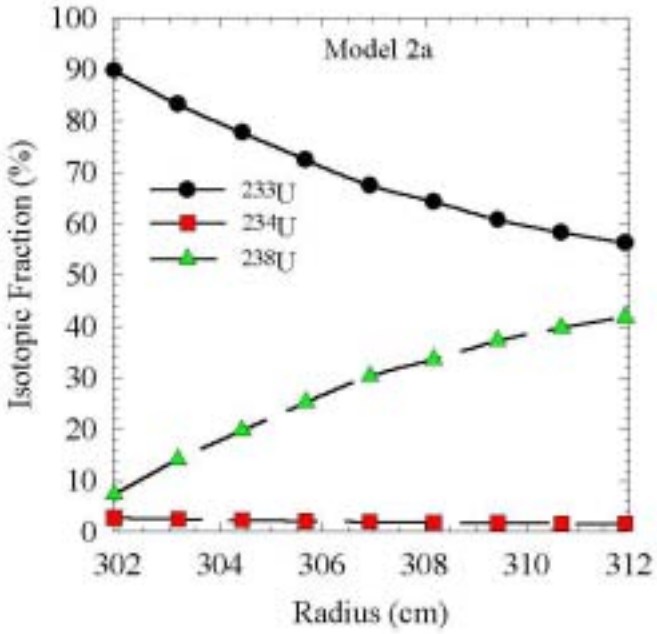

Figure 6. Isotopic composition of uranium fuel along the fuel zone after 48 months of plant operation period

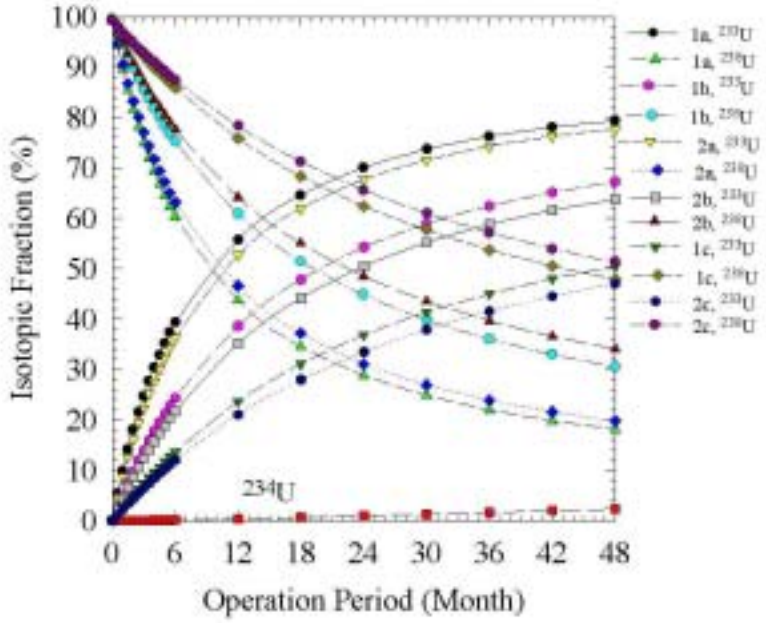

Figure 7. Temporal variation of the percentages of the uranium isotopes in the blankets with ThO_2 and natural-UO_2 (in the third row)

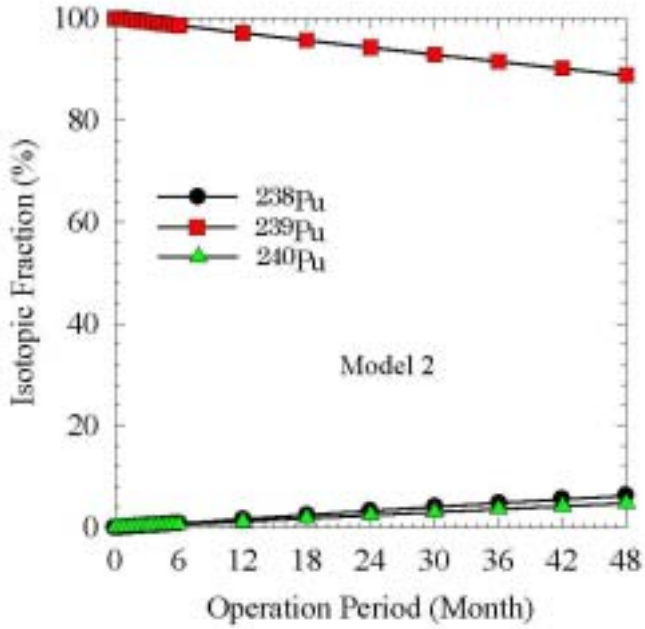

Figure 8. Temporal variation of the percentages of the plutonium isotopes in the blankets with ThO_2 and natural-UO_2

The intensity of spontaneous fission neutrons in ^{238}Pu and/or ^{240}Pu is inversely proportional to their respective spontaneous fission half lives of $T_{1/2} = 4.9\times10^{10}$ and 1.2×10^{11} years [31], and is about 50000 and 110000 times higher than in ^{239}Pu ($T_{1/2} = 5.5\times10^{15}$ years). Hence, only a few percent of ^{238}Pu and/or ^{240}Pu would already denaturate the generated plutonium to a non-prolific level due to their high neutron background levels [32]. The denaturation effectiveness of ^{238}Pu is about twice higher than ^{240}Pu. Previous analyses has indicated that the ^{240}Pu content must be <5 % in weapon grades plutonium fuel [33-35]. Therefore, a total ^{238}Pu +

^{240}Pu content of > 5 % can be considered as sufficient to denaturate the plutonium, safely. On the other hand excessive depression of the isotopic ratio of the ^{239}Pu component in plutonium fuel is not desired, as it would reduce the nuclear quality of the fissile fuel.

Figure 8 shows the variation of the percentages of the plutonium isotopes in the blankets with ThO_2 and natural-UO_2 in the course of the breeding process. At the beginning, plutonium fuel is of highly weapon grade quality. Hence, careful international safeguarding is required. In critical thermal reactors, plutonium is rapidly denaturated (within few days) due to the accumulation of the even ^{240}Pu isotope. Under the high energetic fusion neutron environment, the accumulation of the denaturating even isotopes with high spontaneous fission yield (^{238}Pu and ^{240}Pu) proceeds very slow. The production of ^{238}Pu is faster than that of ^{240}Pu. One can recognize in Figure 8, that an irradiation period > 24 months would be required to barely denaturate the plutonium component of the fissile fuel.

In order to produce a fully denaturated fissile fuel out of ThO_2, the fertile fuel has been mixed with LWR spent nuclear fuel. The latter consists of fissionable material which contains a high grade of even plutonium isotopes. Hence, such a MOX fuel is already and inherently denaturated at the start-up. Figure 9 shows the variation of the percentages of the plutonium isotopes in the blankets with ThO_2 and natural-UO_2 in the course of the breeding process. In this MOX fuel, both ^{233}U as well as ^{239}Pu are successfully denaturated throughout all stages of the plant operation, as it can be observed in Figures 5 (models 1b and 1c) and 9 (models 2a, 2b and 2c), respectively. In a thorium fuel cycle, a MOX fuel consisting of thorium and LWR spent fuel has a high grade of proliferation resistance.

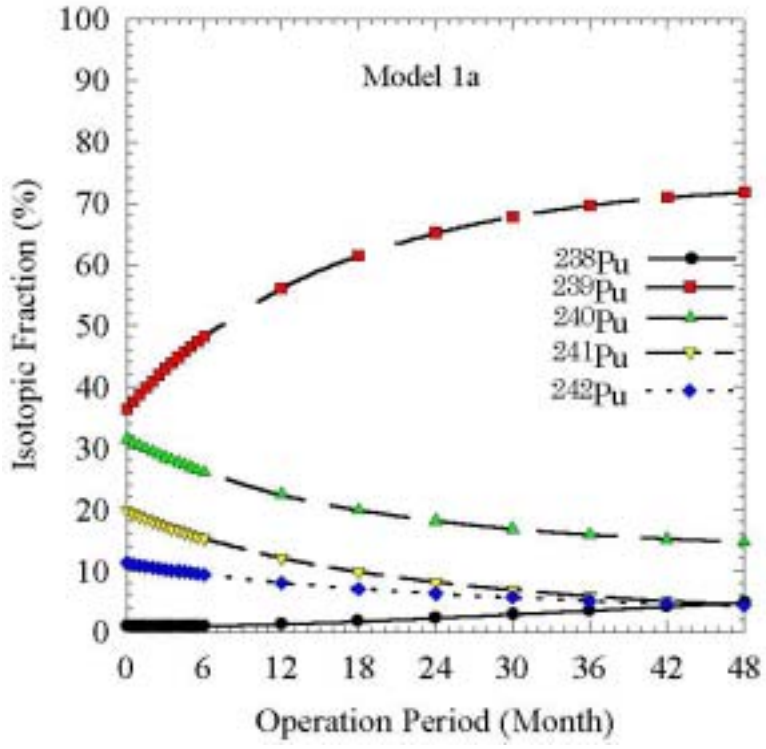

Figure 9. Temporal variation of the percentages of the plutonium isotopes in the blankets with ThO_2 and LWR spent fuel

4. POWER DENSITY FLATTENING IN THE FISSILE ZONE

A fusion driven hybrid reactor has an exponentially decreasing fission power profile. However, a quasi-constant FPP in the blanket would have many advantages from an engineering point of view with respect to a simpler fuel management scheme (better fuel utilization), higher total power output, lower temperature, and radiation damage gradients throughout the blanket. Different approaches were suggested for power flattening in a hybrid blanket. In recent work, a straight-forward numerical-graphical method had been evaluated for power flattening in fusion-fission (hybrid) reactors and a MOX fuel (ThO_2, natural UO_2 and nuclear waste actinides) with a variable fraction of components has been used in the fissionable zone of a hybrid blanket [36-38]. In the present work, ThO_2 has been mixed first with natural UO_2 and afterwards with LWR spent fuel to obtain a quasi-constant fission power profile. Table I shows the variable fraction of components for different models. Figure 10 shows the nuclear heat production density in the fissile zone for the models 1a, b, c. The quasi-constant power shape is saved over 48 months, which was one of the major aims of this work.

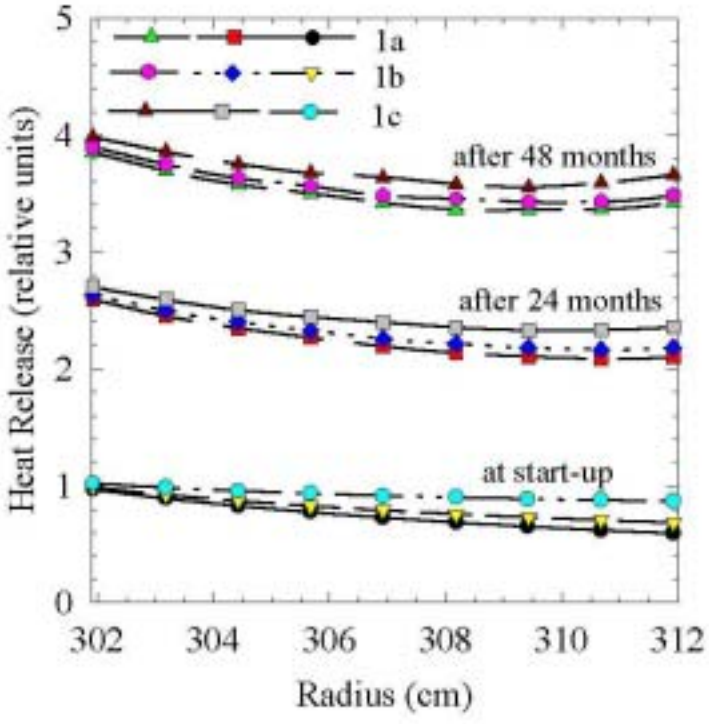

Figure 10. Nuclear heat production density in the fissile zone

5. CONCLUSIONS AND RECOMMENDATIONS

In this study, a possibility of denaturing the ^{233}U fuel produced in a fusion breeder has been investigated. The main conclusions are as follows:

- A fusion breeder containing ThO_2 would produce weapon grade ^{233}U. It must be denaturated for commercial utilization [14].

- The denaturation of the fuel in a fusion breeder can be performed by mixing ThO_2 either with natural UO_2 or with LWR spent nuclear fuel in sealed fuel rods for reutilization in critical reactors without fuel rod reprocessing. ThO_2 mixed with natural uranium content of > 2% produces denaturated ^{233}U, but the ^{238}U component in the natural uranium begins to produce highly prolific ^{239}Pu. Only after long operation periods in the fusion driver > 24 months, both fissile fuel components can be denaturated.

- ThO_2 mixed with LWR spent nuclear fuel content of > 2% can produce fully denaturated fissile fuel.

- It is possible to obtain a quasi-constant nuclear heat production density in the fissile zone by mixing ThO_2 either with natural UO_2 or with LWR spent fuel variable fraction of components. This has significant advantages in plant operation and also allows a uniform utilization of the nuclear fuel in the fissile zone.

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THE USE OF THORIUM FOR PLUTONIUM UTILIZATION IN REACTORS*

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Abstract. Fuel cycle with a full-scale use of thorium is only a far perspective for Russia. Resources of uranium for NPPs with thermal neutron reactors will be sufficient for decades in Russia and involvement of fast neutron reactors into power system makes the problem of raw resources less actual. However, in Russia research works on uranium-plutonium cycle were closely followed by those on thorium-based one. Of course scales of those research works were incomparable. But recently the peculiarities, problems, and perspectives of thorium-based cycle have been discussed more actively in Russia and abroad. Besides, experts have realized that application of thorium-based cycle, at least in the nearest future, will be most probably evolutional, which does not demand any radical changes in the existing fuel cycle. At the same time, some useful features of thorium can be used in a short term. The possibility of using thorium in the process of plutonium utilization is one of such useful properties; in this case, unlike MOX fuel, the plutonium breeding is excluded. The feasibility to include plutonium and thorium into a fuel cycle of Russian light-water reactors WWER-1000 is also touched upon in this report. Once-through fuel recycle has been considered, the final decision on its further utilization being postponed. Basic fuel parameters and properties, important from safety standpoint, are presented. The minimum changes in the structure necessary to ensure safety of plutonium-thorium fuel at the level of conventional WWER-1000 are needed. A wide range of problems investigated and to be investigated is outlined in the paper.

1. GENERAL AND PARTICULAR ASPECTS OF INVOLVING THORIUM INTO NUCLEAR POWER

Necessity of involving thorium into nuclear power of Russia at present is not caused by depletion of uranium raw material resources. Uranium resources for supplying operating NPPs with thermal reactors are available for decades, and with involving into nuclear power fast neutron reactors the issue of raw materials becomes to be not so important. At the same time, in Russia practically straight away after development of studies on uranium-plutonium cycle the works were begun on thorium cycle as well. Of course, scopes of these studies are not comparable. The works on thorium cycle were conducted for both studying aspects of development of nuclear power and the ways of involving thorium into it as an additional resource (long-term outlook), and studying those useful qualities which can be introduced by the use of thorium in operating reactors (short-term outlook and medium-term aspect).

Involving of thorium, accumulation and use of ^{233}U bring in essential good points in nuclear power:

- Fuel base increases double or more.
- Characteristics of operating thermal reactors (WWER, PWR, HWR) are improved in the use of fuel, safety, nonproliferation, accumulation of long-lived radioactive nuclides. It provides possibility of essential extension of life of the developed types of reactors in nuclear power and use of their enormous operating experience.
- Elaboration of thorium fuel cycle may stimulate renewal of types of reactors put aside (MSR, HTGR).
- Combining thorium and uranium-plutonium cycles with using thermal and fast reactors gives to nuclear power significant flexibility and possibility of optimal use of accumulated plutonium and rejects of depleted uranium, accumulation of ^{233}U and involving of thorium.

* 1999 meeting.

- Transmuters (critical and subcritical) with thorium blanket can help in solving the problem of burning-out actinides and fission products without second buildup of minor actinides.

In the last few years peculiarities, problems and aspects of thorium fuel cycle have been discussed in the international nuclear community much more actively than it was 10-15 years ago. In so doing one of the most important incentive to studying possibility of involving thorium fuel cycle in the context of present day is a problem of improvement of fuel cycle safety from viewpoint of nuclear material nonproliferation and decreasing in stocks of accumulated plutonium.

From the other hand, the use of Pu along with thorium and initial accumulation of ^{233}U can be considered as the first and essential step for going to closed fuel cycle based on ^{233}U and thorium, where the use of many valuable features of ^{233}U and thorium is able to the utmost. However, a wide variety of objective factors restrain development of thorium cycle:

- Sufficiency of raw material base even with the most optimistic prognoses of usual nuclear power development.
- Large sums has been invested in elaboration and creation of uranium cycle, although up to now closing of the cycle has been performed on a small scale. At the same time accumulation of plutonium in spent nuclear fuel by existing and future nuclear power serves as an incentive to ensuring the use of this plutonium.
- Characteristics and reactor safety in uranium-plutonium cycle are improved and, most probably, will be acceptable for the society, and merits of thorium cycle taking into account economic factors can be considered as long-term outlook for a long time.

The said above means that the rate of progress to developed thorium cycle with the use of ^{233}U will be determined by different factors of national economy and power engineering policy. At the same time, partial involving of thorium can turn out to be useful from the standpoint of a number of particular problems of nuclear power. Here there are some examples of possible developments and decisions, which realization are able at the first stage.

- Reactors of the WWER type with partial or full inventory with thorium and plutonium oxides can be used in resolving the problem of decreasing weapons- and reactor-grade plutonium stockpiles.
- The WWER reactor with fuel based on mixture of thorium and 20 % enriched uranium may be interesting from the position of delivery to foreign countries because its fuel is protected in addition from proliferation of nuclear materials (both fresh and spent fuel).
- Thorium blankets of fast reactors (with postponed reprocessing and extraction of ^{233}U until the time when there is demand for this material) utilize neutron leakage instead of accumulating piles of uselessly activated steel reflectors.
- Thorium blankets of accelerator driven subcritical systems can be used in burning out actinides and fission products.

Taking this into consideration, resolving of thorium issue can not be put aside for a long time. It is worth to continue its development in a sequential coordinated manner but not forced. Minatom's institutes, Kurchatov Institute, Moskow Institute of Physics and Engineering, Obninsk Institute of Nuclear Power Engineering virtually have not stopped independent researches in the area of thorium cycle, and currently supported by new works on studying reactor concepts, physics of thorium systems, technology. Reflection of these works are recent publications [1-13].

In 1999, in order to coordinate research works performed in the Russian Federation, Minatom entrusted SSC RF IPPE with the duties of head organization of the industry on the problems of thorium fuel cycle.

2. SCIENTIFIC BASIS OF THE FIRST STAGE OF INVOLVING THORIUM IN NUCLEAR POWER

2.1. Neutronic studies

At present, there is less nuclear data available for elaboration of the thorium fuel cycle than for uranium-plutonium cycle [6]. Experimental data on cross sections depending on neutron energy has considerable dispersion, for many nuclear reactions there is null data, and files are constructed based on incomplete theoretical models.

At present time, at the phase of searching developments and conceptual studies, integral experiments, making on critical facilities and power reactors, support designers. Tests on the COBR critical facility has been made for mediums containing Th and ^{235}U, with different spectrum of neutrons due to different moderator content [7]. These experiments are important for calculation verification of critical parameters and ratios of different average cross sections in centers of assemblies.

Very informative experiments of another type are associated with irradiation of samples of Th, ^{233}U, ^{234}U in power reactors. Experts of IPPE in cooperation with reactors personnel carried out comparative irradiations in terms of different spectra (core and blanket of fast reactor, core of thermal reactor). Measurements of isotope composition of irradiated samples, which partially have been made, provided information on absorption cross sections and inelastic neutron scattering by thorium nuclei, on cross section of ^{231}Pa fission. The data have been acquired on accumulation of ^{232}U in ^{233}U produced.

Considerable work on estimation of the experimental results, obtained both from critical facilities and reactors, as well as on creation of the test models for checking used libraries of nuclear data and calculation methods, lies ahead.

2.2. Fuel fabrication technology for the first stage of involving thorium in nuclear power

There are no difficulties expected in technology of production of pelletized thorium oxide [1,9]. There are no considerable problems anticipated in fuel fabrication from mixtures of uranium or plutonium oxides with thorium oxides. Therefore, we can expect that at the first stage of development of thorium cycle technologies for oxide fuel will be sufficient.

The use of metal thorium or its alloys shows considerable promise (for example, as matrix in disperse fuel). In blanket of fast reactor with sodium cooling the use of metal thorium may turn out to be quite acceptable. There is a number of proposals on fuel compositions with inert matrix [9], on new methods of fuel regeneration, containing thorium, uranium and plutonium. Harmonious combination of requirements on fuel compositions for all stages of fuel handling are important: fuel should be practically feasible, workable, economical and capable for reprocessing.

2.3. Outlooks for future reprocessing thorium fuel and ^{232}U problem

In the event of realization closed fuel cycle, extraction of accumulated ^{233}U after irradiating thorium oxide is made difficult by the fact that thorium oxide is a poorly soluble in nitric acid, but addition of hydrofluoric acid solves this problem. Laboratory experience of these technologies has been acquired by now [1, 11]. The use of metal fuel for fast reactors with sodium coolant would facilitate extracting of accumulated ^{233}U.

Industrial technolgy for the thorium cycle must take into account high rate of accumulation of hard γ-emitters in ^{233}U after its extraction from thorium due to ^{232}U decay. ^{232}U content of up to 10 ppm retains the possibility to work with such uranium in gloveboxes, at the greater concentrations protected automated technological lines are necessary.

^{232}U concentrations in fuel will run to hundreds and thousands of ppm in power reactors with economically acceptable expositions of fuel. Restriction on the time after extraction of ^{233}U and removal of decay products of ^{232}U can make work with open fuel easier. However, we must be inevitably oriented to automated processes with heavy biological protection. In this connection it is advisable to develop dry methods of regeneration with following use of fuel vibro-packing. In the Russian Federation technological problems of thorium fuel cycle are under study and development in VNIINM, Radium Institute, IPPE, Kurchatov Institute, NIIAR [8-12].

For experimental adjustment of fuel reprocessing technology based on ^{233}U it is necessary to have ^{233}U with ^{232}U content of less than 10 ppm. Such uranium can be obtained in thermal reactor in circumstances where there are short thorium expositions when accumulating ^{233}U in thorium approximately of 1-2 g per kg [8]. At the same time, it is obviously, that small accumulation of ^{233}U in thorium can be acceptable only for the purpose of experiment when adjusting technologies of fuel based on ^{233}U.

In fast reactor blanket it is able to produce ^{233}U with ^{232}U content of less than 10 ppm with approximately treble accumulation of ^{233}U in thorium. These data has been proved experimentally [8].

Detailed calculation results given in [1] show that in thorium blanket of fast reactor of the BN-800 type it is able to accumulate tens (50-100) kg ^{233}U with ^{232}U content of less than 10 ppm per year. This can provide more easy conditions for conducting studies, development and tests of fuel compositions and fuel elements on the basis of ^{233}U.

Industrial technologies of thorium fuel cycle should be oriented to high ^{232}U content in fuel. High levels of activity attendant to ^{233}U can be considered at the same time as additional barrier for preventing proliferation of this nuclear material.

3. PLUTONIUM-THORIUM FUEL IN THE WWER REACTORS

Recycling reactor-grade plutonium abroad has relied heavily in the PWR reactors for a long time. In recent years the problem of the use of MOX fuel in the WWER reactors is studied in Russia as well. The presence of ^{238}U in such a fuel with necessity leads to considerable decrease in the rate of burning-out of plutonium or even to increasing in its stocks in the case of recycle in reactor with partial MOX fuel loading.

When using fuel on the base of mixture of plutonium with thorium two problems can be solved: plutonium utilization (as this take place, rate of plutonium burning-out rises compared

to MOX reactors) and involving thorium resources in nuclear power. In doing so at the first stage of thorium introduction spent fuel assemblies with thorium fuel can be not reprocessed but stockpiled, expecting better times when technology of reprocessing thorium fuel will be worked through. High γ-activity of fuel irradiated for a long time, determined by great ^{232}U content (thousands of ppm), deteriorated plutonium composition, and difficulty of reprocessing thorium dioxide make unauthorized extraction of fissile material quite unattractive and very complicated.

The most easy option of involving thorium and plutonium seems to be a fabrication of fuel assemblies for the operating WWER on the base of thorium and plutonium dioxides on retention of cycle duration and their construction [4]. Such an approach would allow to fit into existing WWER-1000 construction at the most, that would remove the problem of justification of thermal, hydraulic and thermomechanical characteristics of the core. Necessary level of safety could be assured due to little changes in the reactivity compensation system and fuel assembly reloading scheme, but without changes in the number and construction of fuel assemblies. As an example let's consider some fuel characteristics of the WWER-1000 reactor with partial or full replacement of uranium fuel with PuO_2-ThO_2 mixture. For comparison purpose, there are analogous data for reactor with MOX fuel.

As one can see from the Table I, the use of thorium instead of uranium causes considerable improvement of characteristics of plutonium utilization. Thus, for instance, the amount of burned reactor-grade plutonium for the option with 100 % inventory increased from 398 kg in MOX fuelled reactor to 850 kg in reactor with PuO_2-ThO_2 fuel, as this take place, fraction of burned plutonium relative to loaded plutonium changed from 28 % to 47 %. In so doing the extent of degradation of discharged plutonium was increased: ^{239}Pu fraction in discharged plutonium declined from 50.5 % in MOX reactor to 36.3 % in PuO_2-ThO_2 reactor. It should be noted, that the option with partial PuO_2-ThO_2 inventory reduces plutonium stocks, while reactor with partial MOX inventory just uses power potential of plutonium and deteriorates its isotope composition and increases its stockpile.

From this figures it will be obvious that reactor-grade plutonium recycling does not tend to essential change in radiotoxicity: at the initial stage of storage (tens of years) radiotoxicities are closely allied, then radiotoxicity when recycling plutonium appears to be slightly below radiotoxicity of open fuel cycle, and at the end of given period they prove to be close. By and large it can be noted that the use of thorium for decreasing in radiotoxicity does not give clear merits, both as compared with open cycle, and recycle in the form of MOX fuel.

Intergovernmental agreements between the USA and RF determined strictly fixed amount of weapons-grade plutonium to be utilized. Thus comparison of the options of reactor-burner for such plutonium is worthwhile to make based on radiotoxicity, divided by the mass of weapons-grade plutonium consumed per year. Figures 3 and 4 show radiotoxicity of spent fuel per 1 kg of plutonium consumed in the form of MOX or PuO_2-ThO_2 fuel. It is obvious from the figures that in the interval up to 10000 years there is some advantage of the option with thorium fuel, while as a whole it is difficult to prefer one or another of the options.

Without changing the mode of operation of the WWER-1000 basic option, operation of reactor with fuel mixture of enriched uranium oxide and thorium can be realized. In this instance of special interest is the use of uranium of 20 % enrichment mixed with thorium. Core of this type is protected from unauthorized using fission material for fresh (enrichment of no more than 20 %), and spent fuel (great ^{232}U content). Whilst in this case plutonium is produced, its amount will be small, and isotope composition - non-weapons.

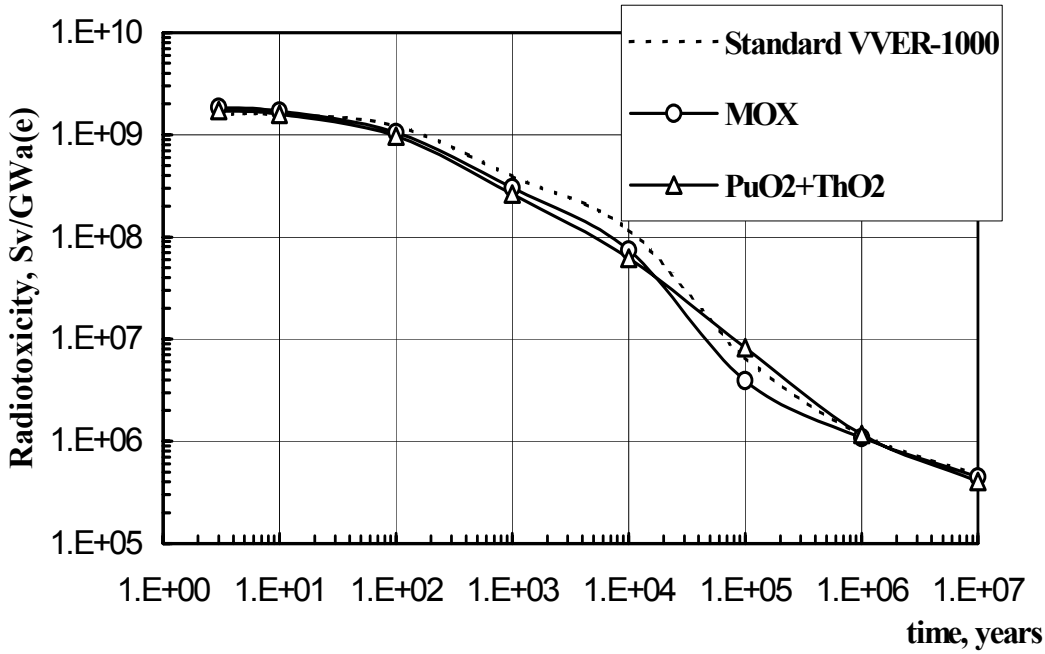

Figure 1. Ingestion hazard of heavy metals (one-through cycle of plutonium discharged from standard WWER-1000 in MOX and (PuO_2-ThO_2) WWER-1000 type reactors).

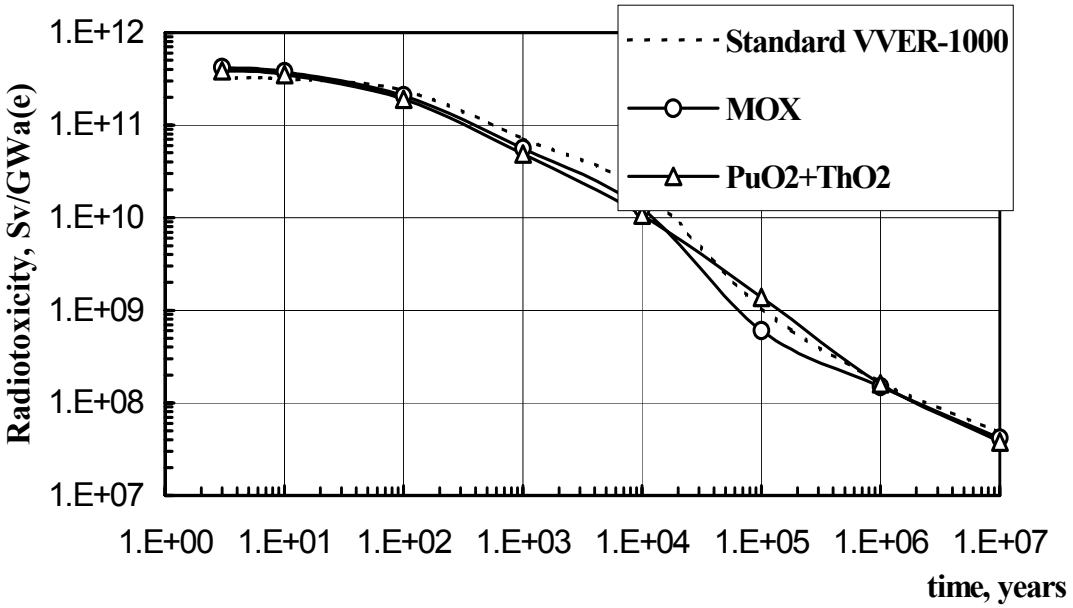

Figure 2. Inhalation hazard of heavy metals (one-through cycle of Pu discharged from standard WWER-1000 in MOX and (PuO_2-ThO_2) WWER-1000 type reactors).

Currently the possibility of thorium cycle for decreasing radiotoxicity of long-lived wastes is under investigation. Within the framework of Research Project coordinated by IAEA "Potential of thorium-based Fuel Cycle to Constrain Pu and Reduce Long-term Waste Toxicities" we made comparative analysis of radiotoxicity of the standard WWER-1000 reactor spent fuel and spent fuel when recycling plutonium in MOX or PuO_2-ThO_2 fuel in considered above light-water reactors of the WWER-1000 type.

Comparison has been made on the basis of incoming with air or water doses, calculated using international standards on Dose Coefficients of Intake recommended by ICRP (ICRP publications, 1991, 1994). Radiotoxicity of recycled fuel was determined as sum of radiotoxicity of spent fuel of uranium WWER-1000 after extracting from it 99 % of plutonium isotopes and radiotoxicity of spent fuel of reactor, in which plutonium recycle is made (reactor-burner).

A relationship between capacities of uranium WWER and reactor-burners was defined based on plutonium mass balance. Only heavy atom and their fission product radiotoxicity was taken into account. Calculation results were normalized to 1 GWa(e) of energy produced and presented in Figures 1, 2.

4. SOME OTHER REACTOR CONCEPTS

At the second stage of involving thorium in nuclear power, with using ^{233}U-Th fuel in light-water-reactors, a new optimization of fuel assembly construction will be needed on the relationship between water and fuel in order to make the most use of neutronic characteristics of ^{233}U and Th and ensure safety characteristics not worse than of existing reactors.

As it is known, in light-water reactors, operating on ^{233}U-Th fuel, an efficient use of fuel can be realized and under certain conditions even extended reproduction of ^{233}U can be achieved. Demonstration of such a possibility took place many years ago in Shippingport (USA) on a reactor, that is called now Radkowsky seed and blanket reactor. At present this conception is developed by Ben-Gurion University (Israel), Brookhaven Laboratory (USA), and in Russia with essential distinctions (the VVER-T reactor) – by Kurchatov Institute [5]. New incentives to its development are the possibility of utilization of weapons plutonium and uranium in seed region and high safety with respect to proliferation of nuclear materials. Construction and technology of the core are specific, and such a core obviously is not able to fit in with existing construction of the WWER reactor in full measure.

In order to accumulate ^{233}U for its subsequent using in thermal reactors fast reactors can be used with radial thorium blankets as well. It is known than accumulation of ^{233}U in the blanket has a set of advantages as opposed to accumulation in the core of thermal reactor [1]. If there is excess plutonium in the system, when extended reproduction does not required, constructions without radial uranium blanket could be used, for example, with steel blanket. Better – with thorium one, ^{233}U will be accumulated in it. Reprocessing of this thorium may be postponed till accumulated ^{233}U be called.

The use of ^{233}U in the core of fast reactor is also not improbable. Mixed cycle stands up attractive:

- in central part of the core – ^{233}U mixed with ^{238}U,
- in peripheral part – plutonium with ^{238}U,
- in the blanket – thorium.

Such a scheme ensures secondary accumulation of ^{233}U and has beneficial properties on safety and neutron balance.

Table I. Fuel cycle characteristics

Plutonium	Weapons-grade				Reactor-grade			
Charge	Partial		Full		Partial		Full	
Characteristics	MOX	PuO_2-ThO_2	MOX	PuO_2-ThO_2	MOX	PuO_2-ThO_2	MOX	PuO_2-ThO_2
Annual charge of $^{235}U/Pu+^{241}Am$, kg	625/268	612/355	41/978	0/1220	625/369	612/519	41/1436	0/1804
Annual discharge of Pu, kg	330	266	661	462	407	401	1038	953
Annual discharge-charge balance, kg of Pu	+62	-89	-317	-758	+38	-118	-398	-850
Fraction of burnt Pu in reference to its charge, %	-	25	32	62	-	23	28	47
Fissile Pu isotopes in discharged Pu, $^{239}Pu/^{241}Pu$, full, %	50.9/16.4, 67.3	44.6/19.7, 64.3	50.5/16.8, 67.3	36.3/22.4, 58.7	50.9/19.1, 70.0	42.0/19.2, 61.2	42.0/19.2, 61.2	29.5/22.3, 51.8
^{232}U fraction in discharged U, ppm	0	3860	0	3675	0	3550	0	3322

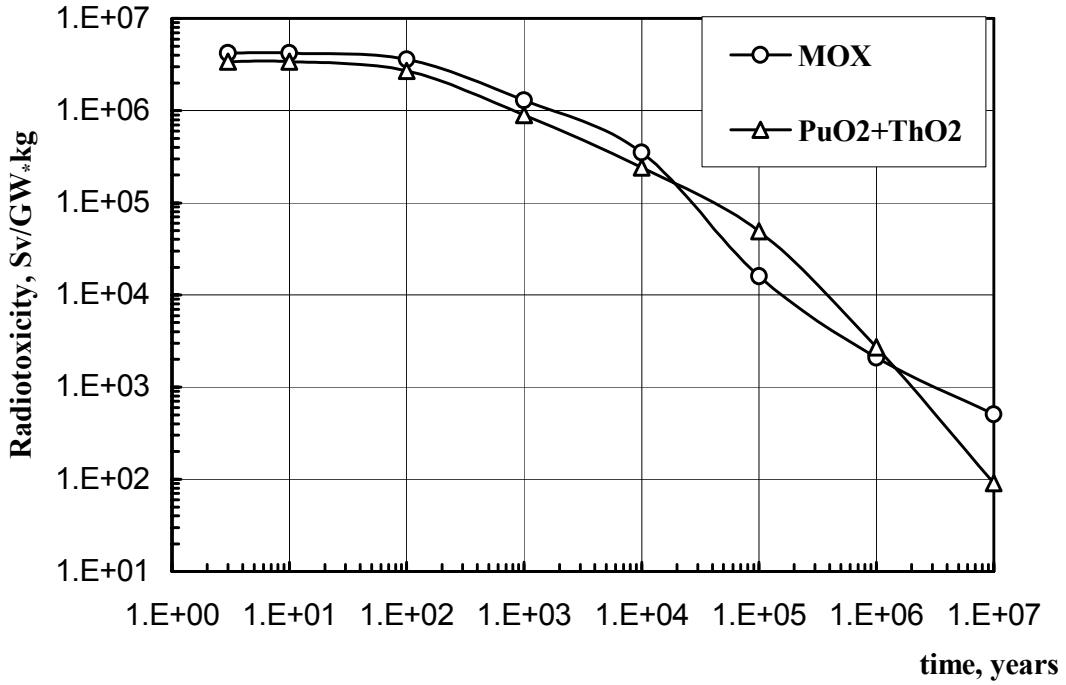

Figure 3. Ingestion hazard of heavy metals from spent fuel based on weapons-grade Pu

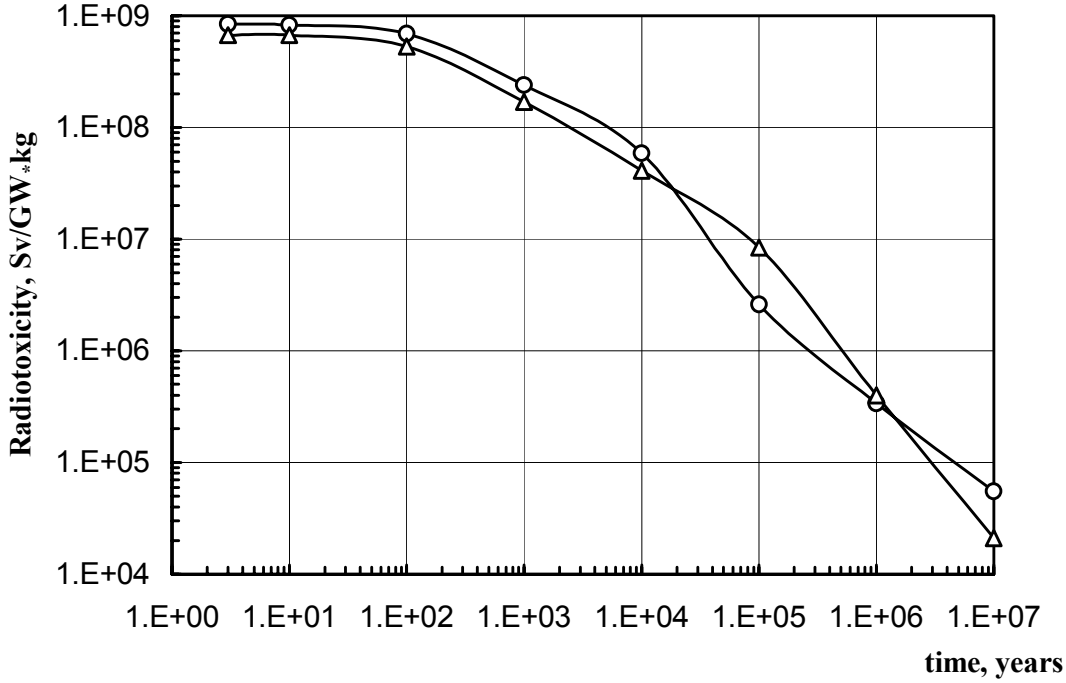

Figure 4. Inhalation h azard of heavy metals from spent fuel based on weapons-grade Pu

In the course of development of nuclear power essentially all reactor concepts were considered as applied to thorium cycle as well. At present these studies are conducted once again, taking into account attained standard of knowledge, today's demands, and notions of the future of nuclear power [1-5].

Considering extensiveness of the data, let's restrict our attention to just brief mention of some of it.

Many experts study molten-salt thorium systems today, both critical and accelerator-driven subcritical with continuous (or periodical) extraction for regeneration. At the same time a practical implementation of these technologies is arduous. Kurchatov Institute, VNIITF, ITEF are concerned with this problem in Russia.

Thorium fuel cycle in HTGR is worthwhile (by reactor physics) assuming its closing [5]. Usually considered for this purpose fuel based on microspheric particle in graphite matrix is not reprocessed by methods of water chemistry. For reprocessing of this fuel methods of dry fluoride technology can be applied [5].

^{233}U application may appear to be useful for creation of space reactors for global communication satellites and global television. Compared to reactor with ^{235}U, reactor with ^{233}U can has greater resource and higher reliability of elements of direct conversion. Proposals of this sort were discussed in IPPE [1]. Conceptual developments of HWR with thorium cycle are carried out in ITEF.

CONCLUSIONS

1. Involving thorium in nuclear power will be evolutional by its nature, assuming passing through a series of stages. The rate of advance to developed thorium cycle with using ^{233}U will be defined by different factors of national economy and power policy.
2. Reactors of the WWER-1000 type without essential changes in their construction with thorium can be taken up in resolving the problem of reduction of weapons and reactor-grade plutonium surplus just at the first stage of involving thorium in nuclear power. The rate of plutonium utilization in such reactors rises considerably as compared to MOX reactors. There are no considerable problems expected to be when fabricating fuel based on mixtures of thorium and plutonium dioxides. Spent fuel assemblies of thorium reactors can be stored with postponed decision about their reprocessing.
3. At the following stages in developed fuel cycle with using thermal and fast reactors consuming both uranium and thorium, the possibility will be to combine efficiently the best qualities of each of reactor type and each of fuel type.

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PHYSICS DESIGN OF ADVANCED HEAVY WATER REACTOR UTILISING THORIUM*

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Abstract. An Advanced Heavy Water Reactor (AHWR) is being developed in India with the aim of utilising thorium for power generation. AHWR is a vertical pressure tube type reactor cooled by boiling light water and moderated by heavy water. It has been optimised for the thorium cycle. The main design objective is to be self-sustaining in ^{233}U with most of the power from the thorium fuel using plutonium as the external fissile feed. It incorporates several advanced safety features namely, heat removal through natural circulation and a negative void coefficient of reactivity. The reactor has been designed to produce 750 MW(th) at a discharge burnup of 20,000 MWd/H(e). The physics design of AHWR has followed an evolutionary path ranging from a seed and blanket concept to a simplified composite cluster to achieve a good thermal hydraulic coupling. We have designed a composite cluster using both kinds of fuel namely, (Th-UO_2 and (Th-Pu)O_2. With plutonium seed, negative void coefficient can be achieved by making the spectrum harder. This was done by using a pyrocarbon scatterer in the moderator. The void coefficient strongly depends on plutonium. As plutonium burns very rapidly, it is not possible to achieve uniformly negative void coefficient with burnup in this cluster. Alternatively, burnable poison can be used within the cluster to achieve negative void coefficient taking advantage of the flux redistribution and change in spectrum upon voiding. Here, it is possible to achieve almost constant void reactivity with burnup resulting in a good thermal hydraulic coupling. The cluster design presently incorporates a central burnable absorber region. Boiling light water coolant requires that the core power distribution be optimised with thermal hydraulic parameters. The peaking factors inside the cluster should be low so as to have significant margin in operational conditions and to avoid burnout in accident conditions. The variation of reactivity from cold clean to hot operating has been evaluated. In this paper, results of the core calculations, neutronic-thermal hydraulic coupling, reactivity swings and kinetic parameters were presented.

INTRODUCTION

With the aim of utilising the vast reserves of thorium for power generation, India is currently engaged in the design of an Advanced Heavy Water Reactor (AHWR). This is a reactor which is being optimised for the thorium cycle. AHWR is a vertical pressure tube type boiling water-cooled, heavy water moderated reactor, designed to produce 750 MW(th) power. The major design objectives are that:

- most of the power should come from thorium fuel,
- the system should have negative void coefficient,
- the system should be self-sustaining the ^{233}U
- the discharge burnup should be higher than 20,000 MWd/t(e) and
- initial plutonium inventory should be low [l].

The design philosophy was to use the existing expertise of the heavy water reactors while incorporating other advanced passive safety features [2]. The design incorporates the pressure tube concept and on power fuelling with low temperature and low-pressure moderator.

* 1999 meeting.

DESIGN BASIS

The fuel cycle chosen is the Th-^{233}U cycle and the objective is to produce about 75% of the power form thorium. In order to go to high discharge burnups, we chose to use plutonium as the external fissile feed. Since ^{233}U is not available in large amounts, any system utilising ^{233}U should become self sufficient in ^{233}U in the equilibrium cycle. Studies have shown that it is possible to achieve a self-sustaining cycle in PHWR, but the discharge burnup attainable is only 12,OOOMWd/t(e) [3]. It is possible to increase this by adding some make-up fissile content in the form of plutonium. However, the void coefficient in such a PHWR is positive in spite of using heavy water as coolant.

Thorium oxide has got excellent fuel performance characteristics, and is capable of going on to very high burnups. Since this has to be matched by reactivity considerations, the initial plutonium enrichment could be very high. This would have the undesirable consequence of too high a fraction of power coming from plutonium. If the plutonium is in segregated pins, these pins could be refuelled more frequently then the Th-^{233}U pins, so that a lower enrichment would suffice, and the fractional power production from plutonium will be correspondingly lower.

The thermal absorption of thorium is three times that of ^{238}U. Therefore, the conversion would be more efficient and it is possible to consider the use of light water coolant. This opens the way to direct cycle and in-core boiling. With boiling coolant, the reactor has to be vertical, and one can think in terms of 100% heat removal by natural circulation with passive safety features. But one has to also make the coolant void reactivity negative from the point of view of safety as well as control stability

When coolant boils in the channels, there will be a strong coupling between thermal hydraulics and neutronics The cluster peaking factors and overall peaking factors have to be within the acceptable range from the point of view of heat removal by natural convection.

THE CLUSTER DESIGN

The preliminary objective was to achieve a negative void coefficient of reactivity with light water as coolant. Initially we were concentrating on a seed and blanket concept [4]. The cluster design gradually evolved into a composite cluster concept where two types of fuel are being used. The fuel assembly arrived at was a 52 rod cluster designated as D-3 [20]. The pins are placed in a square pitch of 1.37 cm inside a cylindrical pressure tube. The arrangement resulted from spacer design and coolant pressure drop requirements. Figure 1 shows the cross section of one such cluster. The cluster was optimised with respect to the fissile content, maximum attainable reactivity, negative void coefficient and low cluster power peaking factors. The cluster had 32 (Th, ^{233}U)O_2 pins and 20 (Th,Pu)O_2 pins. The enrichments used were 2.94% ^{233}U in the inner 32 pins and 2.7% Pu in the outer (Th,Pu)O_2 pins. The objective here was that maximum power should come from the thorium fuel. Table I shows the description of the fuel assembly.

The physics studies showed that in order to get negative void coefficient, the lattice pitch should be around 18.0 cm as shown in Figure 2[1].

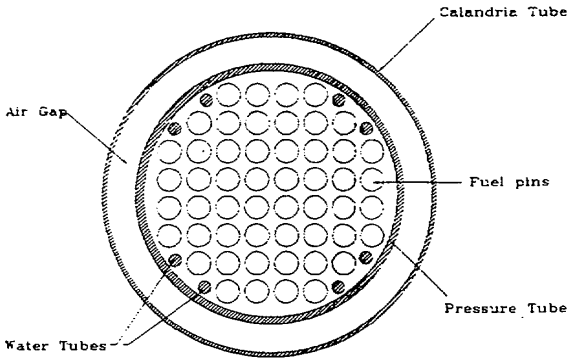

Figure 1 Cross section of AHWR Cluster (D3).

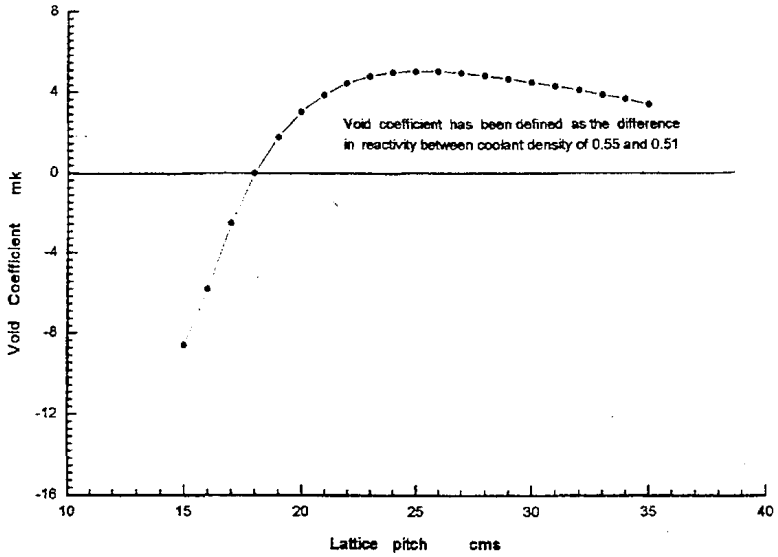

Figure 2 Void coefficient in D3 cluster as a function of pitch.

The spectrum is then relatively hard and the light water carries out the function of both coolant and moderator. It was extremely difficult to engineer such a tight pitch. The lowest pitch which could be designed was 29.4 cm. This would make the spectrum extremely soft. In order to make the effective pitch as 18 cm, many low neutron absorber materials were tried as fillers in the moderator region. Finally, it was decided to use pyrocarbon as the filler/scaterrer. The cluster design was optimised for a moderator region consisting of 80% Pyrocarbon and 20% heavy water. This was achieved by using pyrocarbon blocks in the region around the calandria tube. This introduces a loss of reactivity of about 40 mk.

The plutonium composition has a strong bearing on the void coefficient. Although it was possible to achieve negative void coefficient at the beginning-of-cycle, the void coefficient becomes positive as plutonium burns. Negative void coefficient can still be achieved by lowering the pyrocarbon density, which is a difficult task. Safety was being the paramount consideration; it was decided to look into other possibilities of achieving negative void coefficient.

Table I. Description of the AHWR fuel assembly

	D-3(20)	D-4
Fuel pellet radius cm	0.49	0.49
Sheath outer radius, cm	0.56	0.56
Pellet gap, mm	0.1	0.1
Number of pins in the cluster	52	36
Central absorber region	NIL	20% Dy in Graphite matrix
Radius of the central absorber region cm		2.2
Clad outer radius cm		2.4
Number of plutonium bearing pins	20	20
Pu enrichment (RG)	2.7%	4,0%
Number ofthorium-U^{233} pins	32	16
U233 enrichment	2.94%	5.5%
Fuel pellet density, g/cc	9.6	9.6
Pitch of the pin lattice(square), cm	1.37	1.37
Clad thickness, cm	0.06	0.06
Pin-to-pin gap, cm	0.25	0.25
Coolant tube inner radius, cm	6.0	6.0
Coolant tube outer radius, cm	6.35	6.35
Calandria tube inner radius, cm	7.7	7.7
Calandria tube outer radius, cm	7.9	7.9
Number of water rods	8	8
Water rod inner radius, mm	2.5	2.5
Water rod outer radius, mm	3.0	3.0
Material of clad	Zircaloy	Zircaloy
Material of water tube	Zircaloy	Zircaloy
Material of pressure tube	Zr-2.5% Nb	Zr-2.5% Nb
Material of calandria tube	Zircaloy	Zircaloy
Length of active fuel, cm	350.0	350.0
Mass of heavy metal in an assembly, kg	116	80
Coolant material	Light water	Light water
Coolant density (average), g/cc	0.55	0.55
Coolant in water tubes	Light water	Light water
Water tube water density, g/cc	0.771	0.771
Maximum cluster peaking factor	1.54	1.43

Several cluster designs were tired out. Certain liquid moderating materials also were studied. But these designs resulted in a very low reactivity. It is possible to achieve negative void coefficient by using an absorber in the fuel or in isolated pins in an inert matrix [5]. The modified cluster has a central absorber region of 2.2 cm radius consisting of Dysprosium in a Graphite matrix. The amount of dysprosium has been optimised to obtain negative void coefficient. The cluster now has 36 pins, with inner 16(Th, $^{233}U)O_2$ pins and outer 20 (Th,Pu)O_2 pins. The description is given in Table 1. The cluster has been designated as D-4. The cross section of the D-4 cluster is given in Figure 3.

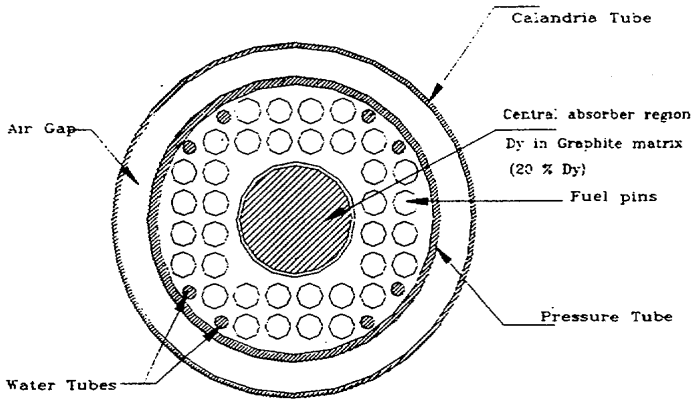

Figure3 Cross section of the D4 cluster.

The presence of dysprosium alters the absorption profile inside the cluster, thereby making the void coefficient negative. The void coefficient remains negative throughout the burnup regime and the power profile also remains flatter providing a good thermal hydraulic coupling. In this design, the outer moderator region consists of heavy water only The loss in reactivity due the presence of dysprosium is partly compensated by the removal of pyrocarbon in the moderator. As the number of fuel pins have decreased, the fuel enrichment will have to be increased in order to get reactivity or a higher discharge burnup. The enrichments are 5.5% ^{233}U and 4.0% Pu for the two type of pins respectively. Table II gives the optimisation of the D-4 cluster.

PERFORMANCE CHARACTERISTICS OF THE TWO CLUSTER DESIGNS

The cluster was optimised to meet the design objective of achieving maximum burnup and being self-sustaining in ^{233}U. The lattice level variation of reactivity and the void reactivity with burnup is given in Figures 4 and 5 for the two cluster designs respectively. The burnup profile of the two types of pins will be different. Plutonium will bum faster and therefore these pins will reach 20.000 MWd/t(e) earlier than the (Th, ^{233}U) pins. The ^{233}U produced in the outer 20 pins will be governing the self-sustaining criteria. Since this has to be matched with the reactivity criteria, it is required to reconstitute the (Th-Pu)O_2 pins. The variation of the fissile content and the power profile during this reconstitution phase has been worked out.

Table II. Optimisation of the D-4 cluster.

Case No.	No. of Pu pins	Pu %	No. of U^{233} pins	U^{233} %	K-inf.	Void reactivity mk	Cluster peaking factor	Dy in Graphite
Central absorber 3.1 cm								
1	8	3.0	28	4.5	1.243	-0.776	1.31	5.0% Dy
2	20	3.0	16	4.5	1.161	-0,985	1.39	-do-
-^	20	3.0	16	4.5	1.171	-0.784	1.39	4.0% Dy
4	20	3.5	16	5.0	1.224	-0.482	1.41	3.5%Dy
Central absorber 2.2 cm								
5	20	3.5	16	5.0	1.253	+0.214	1.38	6.0% Dy
6	20	3.5	16	5.0	1.245	+0.027	1.38	8.0% Dy
7	20	3.5	16	5.0	1.239	-0.054	1.38	10%Dy
8	20	3.5	16	5.0	1.230	-0.243	1.39	15%Dy
9	20	4.0	16	5.5	1.268	-0.058	1.41	15%Dy
10	20	4.0	16	5.5	1.262	-0.184	1.42	20% Dy

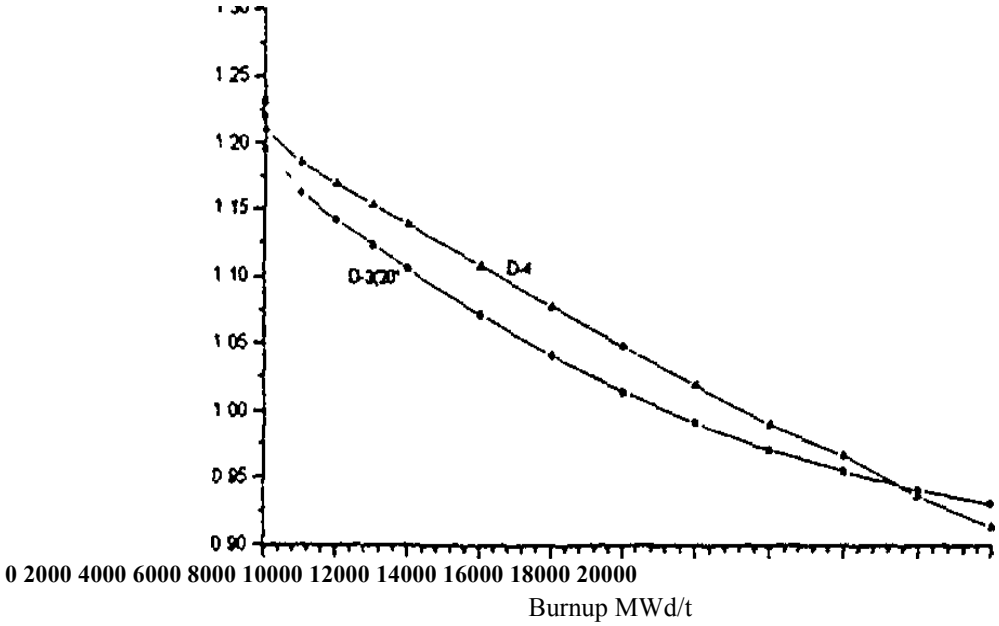

Figure 4 Variation of k-effective in D3 and D4 clusters of AHWR.

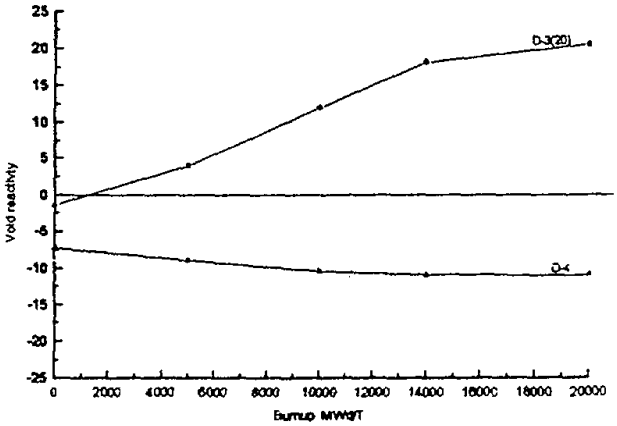

Figure 5 Variation of void reactivity in D3 and D4 clusters of AHWR.

The operational parameters for the two cluster designs have been summarised in Table III. In the D3 design the void reactivity is very sensitive to the ^{239}Pu concentration.

The void reactivity becomes positive as ^{239}Pu burns. However, if we use (U, Pu)O_2 pins instead of (Th,Pu)O_2 in the D-3 cluster, there is in-situ generation of ^{239}Pu, the void reactivity remains negative. But since, thorium is not used, ^{233}U will not be produced in these pins, which governs the self-sustaining criteria.

In the D4 design, the void reactivity continues to be negative even when ^{239}Pu content decreases with bumup. The power profile along the cluster shows that about 20% is being produced in the inner 16 pins of the D-3 cluster. The heat ratings of the 36 pins of the D-4 cluster will be enhanced by a similar amount if the fuel assemblies are to produce the same power. The optimisation of channel power is being done from the thermal hydraulic calculations.

Table III Reactivity coefficients for D-3(20) and D-4 clusters for AHWR

	D-4	D-3(20)
K-effective 0 MWd/t(e) 8000 MW·d/t(e) 12000 MW·d/t(e) (Avge. Coolant config.)	1.23145 1.07871 1.01084	1.22014 1.04207 0.99147
Void Coefficient 0 MW·d/t(e) 10000 MW·d/t(e) (40-50% voids)	-0.184mk -0.397 mk	-0.107mk +0.756 mk
Void reactivity 0 MW·d/t(e) 10000 MW·d/t(e) (0-100% voids)	-7.18mk -10.35 mk	-1.43 mk +11.90mk
Temperature coefficients d-Rho		
a) Tf=Tc=Tm (300°K - 353°K)	+2.36 mk	+3.54 mk
b) Tf=Tc (353°K-558°K) Tm=353°K	+3.6 mk	+22.0 mk
Tf=Tc=558°K, Tm=353°K Tf=(558°K-898°K)	-3.27 mk	-6.9mk
Cluster peaking factor	1.44	1.54
Average energy of thermal neutrons	0.08 ev	0.098 ev
Prompt neutron lifetime	0.55ms	0.47ms
Fuel inventory in cluster a) Heavy metal b) Plutonium c) U-233	 80 Kg 1.91 Kg 1.96 Kg	 115.6 Kg 1.29 Kg 2.10 Kg

CORE OPTIMISATION STUDIES

The AHWR core has been housed in a calandria of 860 cm diameter and a height of 500 cm. The active core will have a height of 350 cm and. a radius of 340 cm. There will be a radial reflector of about 60 cm of heavy water. The bottom and top reflectors will be 75 cm each of heavy water. The power distribution has been optimised to obtain

- maximum discharge burnup,
- maximum attainable reactivity,
- efficient heat removal through natural convection.

The coolant in this reactor being in the two-phase state, there is very strong interaction between the neutronics and thermal hydraulics. The axial power distribution in the average channel of each set was also calculated. This power distribution is used in the thermal hydraulic analysis to arrive at the core void distribution or coolant density profile. This process was repeated until an optimum power distribution was achieved. The channels in the core were grouped into nine sets, and the average channel power in each of these sets was obtained A radial power factor was defined as the maximum-to-average power in that particular group of channels. This was the deciding factor to ensure an optimum power distribution from heat removal capabilities. The ratio of maximum-to-minimum radial power factors is limited to 1.5 to 3.0 in the core. The AHWR core optimised with D3 clusters consisted of 428 fuel positions, 4 regulating rods, 4 absorber rods, 32 locations SDS-1, and 32 locations for SDS-2. The core had a three zone refuelling scheme with reconstituted

Table IV. Description of AHWR at the present stage of evolution

Reactor power MW(th)	750
Fuel description	
Number of pins	36
Number of Pu bearing pins	20
Number ofthorium-U^{233} pins	16
Number of water tubes for ECCS	8
Plutonium content in MOX%	4.0%
^{233}U content in thorium	5.5%
Coolant water density (varies in the range)	0.55 g/c^3 0.55-0.50
Total number of channels	452
Number of seeded clusters	424
Number of clusters with all "thorium-U^{233}" pins	28
Number of fuelling zones	2
Number of reconstitutions	1
Thorium pins discharge burnup MW·d/t	20,000
MOX pins discharge burnup MW·d/t	20,000
Lattice pitch cm	29.4
Active fuel length cm	350
Moderator and reflector	D_2O
Calandria radius cm	430
No. of adjuster rods	16
Worth of adjuster rods mk	14.0
No SDS-1 rods	36
Worth SDS-1 mk	70
Performance data in equilibrium core	
Radial form factor	1.63
Hot spot factor in the seeded cluster	1.43
Hot spot factor in the thorium-U^{233} cluster	1.37
Maximum channel power MW	2.70
Maximum-to-minimum radial power factor	2.6
Fraction of power from thorium%	59.0

clusters in the outer regions. 84 (Th-$^{233}U)O_2$ clusters have been used for power flattening and maximising the power output from thorium region, The power from thorium was around 75%. The hot spot factor in the seeded cluster was 1.54 and the maximum-to- minimum radial power factor was 2.16.

However, the equilibrium core had a positive void coefficient. The fuelling requirements have also been worked out. The core was nearly self-sustaining in ^{233}U with the gain coming from the 20 $(Th,Pu)O_2$ pins of the D3 clusters.

The description of the AHWR core at the present stage of evolution is given in Table IV. The AHWR core with D4 clusters is being optimised. We are currently using 452 fuel positions, 32 locations for SDS-1 and 16 locations for adjuster and regulating devices. Here again, 40 (Th, $^{233}U)O_2$ clusters are used for power flattening and to maximise the output from the thorium region. A simple two zone refuelling scheme has been adopted.

The void coefficient remains negative in the equilibrium fuel loading. The power from thorium is around 59%. The hot spot factors and the radial power factors are not very different from the core loaded with D3 clusters.

REACTIVITY SWINGS

The reactivity variations in an AHWR core will govern both the control functions during normal operating conditions and safety functions while shutting down. Although the actual coefficients will come from the core calculations, the lattice evaluation gives the extent of the reactivity variations. The various temperature coefficients at the lattice level during operational conditions are given in Table 4. The Doppler reactivity due to fuel temperature is about 3,33 mk in the D-4 cluster whereas it is around 6.9 mk in the D-3 cluster. The coolant temperature coefficient is slightly positive initially in both the clusters. The channel temperature coefficient is a combination of reactivity effects of the fuel temperature and coolant density. We have calculated the Doppler reactivity and the coolant temperature effects separately. The reactivity swings from cold to hot operating for both the cluster designs in shown in Figure 6.

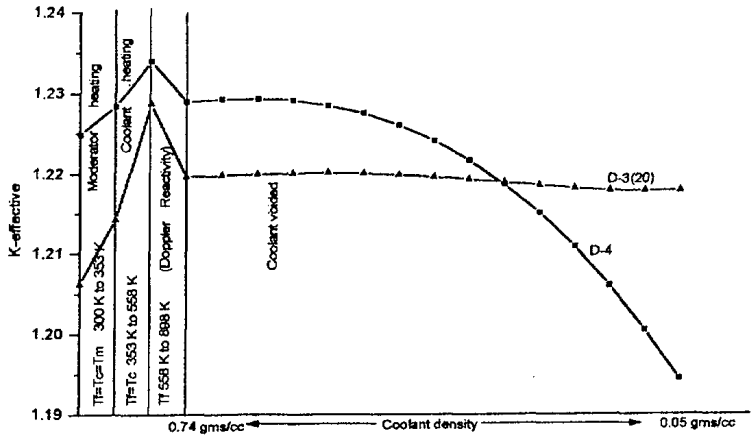

Figure 6 Reactivity swings from cold to hot operating in D3 and D4 clusters.

The operating thermal flux is $5X10^{13}$ neutrons/cm^2s. The equilibrium xenon load is about 20 mk for both the clusters. Since the operating fluxes are lower, the xenon buildup during poisonout period is not expected to be as high as in PHWRs. The xenon override reactivity is within 10 mk for 100% power reduction. The regulating rods can take care of this reactivity variation.

The initial excess reactivity of about 220 mk has to be compensated by a combination of thoria clusters and chemical shim. The thorium clusters are used for power flattening until equilibrium is reached. In the present design since all the area surrounding the calandria tube is filled with heavy water, it is possible to have boron introduced into the heavy water moderator. The boron worth in the D4 cluster is very high which is 19 mk/ppm. An initial boron concentration of about 5-10 ppm is expected to take of all the operating loads The coolant void reactivity remains negative even in the presence of boron. In the D3 cluster, since the amount of heavy water is less, the initial excess reactivity cannot be compensated by boron. A large number of thoria clusters have to be used for reactivity control in the initially.

CONTROL DEVICES AND SHUT-DOWN SYSTEMS

The control requirements for AHWR are low due to on-line refuelling. The power cycles which the fuel assembly will see will also decide the flux tilt function of the control rods. The AHWR core using D3 clusters was optimised with four regulating rods and four adjuster rods. The control devices for core with the D4 clusters are being optimised.

With both plutonium and ^{233}U as fuel material, the delayed neutron fraction is low. The $ß_{eff}$ for AHWR has been calculated as 2.8 mk. Since, the void collapse will introduce positive reactivity; it will be one of the design basis accidents. The worth of the control and safety devices will have to be worked out at all the stages through which the reactor operates, namely, initial, most reactive phase, hot-operating and refuelling and reconstitution stages.

The average neutron energy for thermal neutrons is around 0.08 ev implying a slightly harder spectrum than the PHWRs using natural uranium fuel. The prompt neutron lifetime also is shorter compared to heavy water reactors. The prompt neutron lifetime is 0.55 ms in the D4 cluster and slightly lower in the D3 cluster.

AHWR has two fast acting shut down systems with diverse functions. With this in mind, we are currently working on mechanical shut-off rods falling under gravity as one of the shut down system (SDS-1) and liquid poison tubes (LPTs) as the other shut down system (SDS-2) with the D3 cluster loading. We are presently considering an absorbing element consisting of boron carbide sandwiched in stainless steel. The other shut down system will have Lithium pentaborate solution rising in Zircaloy tubes. With D4 clusters the SDS-2 can be poison injection into the moderator like the modem PHWRs and thus we can use the locations of LPTs for fuel.

FUTURE DEVELOPMENT

The design as it stands is still at the developmental stage. A lot of uncertainties at the lattice level can be overcome by validated nuclear data for the thorium cycle. Monte Carlo techniques are being used to accurately estimate the pin power distribution and void reactivity. The core optimisation studies are going on presently. Multi zone refuelling schemes are being studied to optimise the control requirements. Physics and thermal hydraulic iterations will have to be done to ensure proper coupling. Kinetics and safety analysis are being are being done.

A critical facility for AHWR is being designed at BARC. Several experiments will be carried out using the AHWR cluster. This will validate our calculational methods and models and also benchmark thorium cycle data, performance and handling of thorium fuel.

CONCLUSIONS

We have presented the two cluster designs being developed at present. The D3 design is better from fuel cycle point of view which operates in the near self-sustaining mode as regards to ^{233}U. The D4 cluster has a negative void coefficient throughout the residence and hence has better control and safety features. But the power from thorium is less in this design. The ^{233}U content is being optimised for obtaining self-sustainability. The fuel cycle aspects will have to be optimised with respect to the void reactivity, other operational loads and refuelling

strategy. The development of the pyrocarbon scaterrer in D3 cluster configuration is an engineering challenge. The good points of both the designs could be combined to arrive at an optimum design of the cluster.

The Advanced Heavy Water Reactor is an attempt to design a reactor for the thorium cycle. It will pave a way for utilising the potential of thorium.

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THORIUM PRE-BREEDER/BREEDER ROUTE TO WIDEN THE NUCLEAR MATERIAL BASE FOR GENERATION OF ELECTRIC POWER*

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Abstract. Fission nuclear power is generated almost exclusively from uranium in the power reactors operating in the world today. Though the potential of thorium had been recognized very early, no cost-effective reactors have been designed that can use thorium in a major way. Eventually depleted uranium and thorium would become comparable candidates in the sense that both would need some man-made fissile isotope to continue fission nuclear power. Plutonium will be the only seed material that would be available from the fuel discharged from uranium reactors. In this paper an early induction of thorium in uranium reactors is advocated in order to widen the nuclear material base for continuation of fission nuclear power.

INTRODUCTION

The energy need has grown at a phenomenal rate in the 20th century. While the energy consumption rate has already reached its peak in the developed nations of the West, it is slated for similar such growth in rest of the world in the 21st century. Use of conventional fossil fuels like coal, oil and natural gas for meeting these energy needs is rather easier since the technologies for the same is proven. However, the fossil fuel resources are unevenly distributed and are limited. In addition, it is recognized that the emission from the burning of the fossil fuel leads to the Green House effect and the phenomenon of Global Warming. It would therefore be prudent to conserve these fossil fuels for the purpose of transportation and other domestic uses. It is necessary to look for alternative means of electricity production that would be eco-friendly and can last for several centuries to come.

In this context, energy from nuclear fission can be considered as a timely boon to mankind. Despite the two moral shattering accidents of Three Mile Island and Chernobyl, the fission nuclear power reactors can be deemed to have reached a high level of sophistication. They are capable of being operated with a degree of simplicity equal to or even better than the thermal power stations using fossil fuel. The amount of nuclear waste, especially in a closed fuel cycle, is much smaller in volume and is contained. Technologies are constantly being developed and improvised for its long term storage and disposal. Notwithstanding the apprehensions of the not so knowledgeable public or even the elite class, the nuclear power is likely to be pursued far more vigorously in the next century especially by countries with limited possibility of energy growth from fossil fuels.

It is noted that the present day power reactors use uranium almost exclusively. Thorium is not inducted in any major way since no cost-effective reactors have been designed. Introduction of thorium directly in the existing power reactor designs poses problems of reactivity load adjustment, disruption of power distribution etc. These problems manifest themselves in some form of economic penalty and hence use of thorium is not earnestly pursued by all countries. It is possibly necessary to conceive a new reactor system that is tailor-made for thorium, taking into account the factors like economy, safety and operation etc. India has a special interest in developing new reactor concepts suitable for large scale utilization of thorium, since its thorium reserves are six times that of uranium reserves.

* 1999 meeting.

The physics disadvantages of thorium inhibiting its early use in power reactors are: i) thorium has no intrinsic fissile content, ii) the thermal absorption cross section of ^{232}Th, is nearly three times that of the fertile ^{238}U in uranium. A thorium breeder reactor (ATBR) concept was conceived in which these disadvantages were turned into advantages [1-6]. A conceptual design of a 600 MW(e) reactor was described in the above references. This reactor concept envisages essentially two phases. In phase-I, it is a pre-breeder, i.e., an efficient ^{235}U to ^{233}U converter. Phase-II will be a breeder or at least a self-sustaining reactor system with (^{232}Th-^{233}U) oxide fuel. The physics design principle will be briefly described here.

PHYSICS DESIGN PRINCIPLES

If natural thoria rods, without any external seed, are placed in the ambience of large thermal neutron flux, a fairly high rate of fertile to fissile conversion occurs. Initial conversion rate is nearly three times that for uranium rods subject to the same neutron fluence. In a thermal reactor neutron spectrum, the asymptotic stable concentration of ^{233}U in thorium is about 1.5% and is distinctly higher than the plutonium formed from uranium. Apart from lower thermal capture cross section of ^{238}U, the thermal absorption cross section of plutonium isotopes is more than two times that of ^{233}U. Hence plutonium production rate is low and consumption rate is high. Plutonium content in uranium rods therefore does not rise much above 1%, even after a long residence time at high neutron flux. In reactors using natural uranium, the residence time is short and hence the plutonium content in discharged fuel is just about 0.3%. The even isotopes ^{240}Pu and ^{242}Pu of uranium burnup chain are non-fissile. They accumulate much more than the even isotopes ^{234}U and ^{236}U of thorium burnup chain. ^{233}U is a far superior fuel compared to plutonium for reuse in thermal reactors owing to its much larger η value. Plutonium is a better fuel for fast reactors for the same reason, but in fast energy range. Fast reactors need however much more fissile material inventory for a given reactor power. When one is contemplating large scale utilization of thorium, thermal reactor is a better option. As can be seen, there are certain advantages of irradiating thorium in comparison to uranium, if one desires a superior and larger residual fissile content in the discharged fuel.

In order to exploit the above physics advantage, we must explore the ways and means of increasing the thoria loading in a power reactor design. Reactors using natural uranium, after reaching equilibrium condition, have practically no excess reactivity. They cannot accommodate thoria rods without any external seed or without some penalty in the already limited discharge burnup of natural uranium. Reactors using enriched uranium have large excess reactivity. This reactivity is compensated normally by some control absorbers in the form of control rods, burnable poison rods containing Gd, ^{10}B etc. and/or soluble boron in moderator. This leads to wasteful neutron captures with no tangible returns. If these neutrons are used for fertile captures in thoria rods, one can use the excess fission neutrons far more effectively. Thoria rods, which behave like absorber rods to start with, turn into regular fuel rods after they accumulate adequate content of the fissile isotope ^{233}U. The geometrical disposition of these thoria rods has to be suitably devised.

A light water breeder reactor (LWBR) was operated successfully at Shipping Port, Pennsylvania, U.S.A. This reactor employed (^{232}Th-^{233}U) fuel. An initial inventory of 501 kg of ^{233}U was used and after five years of operation from 1977 to 1982 a net breeding ratio of 1.013 was reported [7]. It may be noted that the above LWBR started with the man-made fissile isotope ^{233}U. The reactor power was 90 MW(e). Tight lattice spacing was used to

enhance the neutron flux in resonance energy range. Varying ^{233}U contents was used in the seed and blanket regions.

In light water reactors, which need higher enriched uranium, the thermal flux level is somewhat lower than in heavy water reactors of same power. The thermal flux level could be lower by a factor of five. When tight lattice spacing is used, epi-thermal flux is enhanced and one would obtain an intermediate neutron spectrum. If thoria rods without any seed material are to be placed in such a spectrum, there are several factors inhibiting the rapid fertile to fissile conversion in thoria rods.

- Higher thermal neutron capture cross section of ^{232}Th cannot be exploited.
- Intermediate spectrum would require higher seed enrichment. Thoria rods would have to compete with the seed fuel rods that are placed in close proximity due to tight lattice spacing.
- Due to poorer rate of neutron capture, the thoria rods are designed to reside for much longer period. In the WWER-Thorium reactor design, the thoria rods are allowed to reside for as long as nine years while the seed is changed every year [8].
- In intermediate spectrum, neutron capture by ^{233}Pa is enhanced, since it has significant resonance or epithermal capture cross section. In this case the direct formation of ^{234}U isotope is enhanced and a ^{233}U isotope is lost.
- Power mismatch between seedless thoria rods and enriched fuel is substantial and would pose problem in thermal hydraulic design.

In view of the above physics reasoning, it was felt that one must design a core in which there are some islands of high thermal neutron flux trapped in pure moderator regions with low thermal capture probability in the moderator itself. D_2O is the best moderator satisfying this requirement. Fig. 1 illustrates this physics phenomenon.

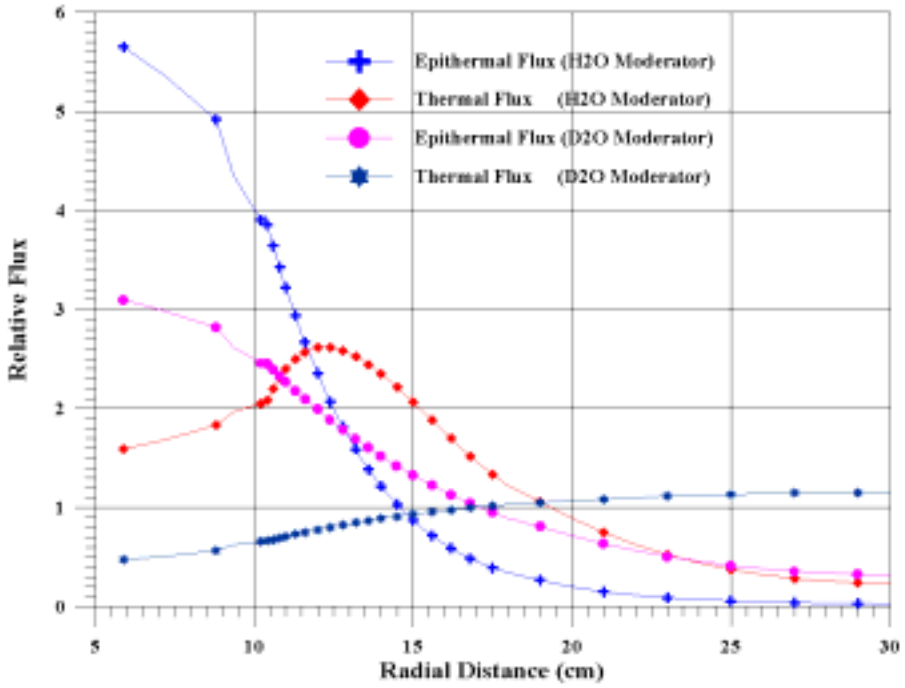

Figure 1. Relative flux distribution across the fuel assembly (H_2O moderator vs. D_2O moderator).

A theoretical study was made in which some seed fuel rods are surrounded by thick moderator regions of either light water or heavy water. The fluxes are normalized such that total absorption in the problem domain is unity. It is seen that in case of light water both epithermal and thermal neutron flux fall rapidly while in case of heavy water moderator the thermal neutron flux increases and remains flat for significant radial distance. It must be added that there was practically no loss of reactivity with increased moderator thickness in case of heavy water while in case of H_2O, the reactivity decreased rapidly. Thus with respect to critical system, the D_2O moderator case is more realistic. One would be able to find ample space for accommodating thoria rods.

D_2O moderated and boiling H_2O cooled reactors have been designed and operated in the world. These reactors are called by different names. In U.K., it was called Steam Generating Heavy Water Reactor (SGHWR) [9]. This type of reactor is ideally suited for irradiating fresh thoria rods in a seed and blanket type arrangement, where every blanket type thoria cluster can be surrounded by seed fuel clusters.

We have designed a new reactor with SGHWR like geometry. We consider a vertical pressure tube type reactor arranged in hexagonal type lattice structure. Fig. 2 gives the cross sectional view of the optimized core loading for an equilibrium core with a typical five batch refueling scheme. Fig. 3 gives the cross sectional view of a blanket type thoria ring cluster with 30 ThO_2 rods. Fig. 4 gives the cross sectional view of the seed fuel cluster with 54 enriched UO_2 fuel rods and 30 ThO_2 rods.

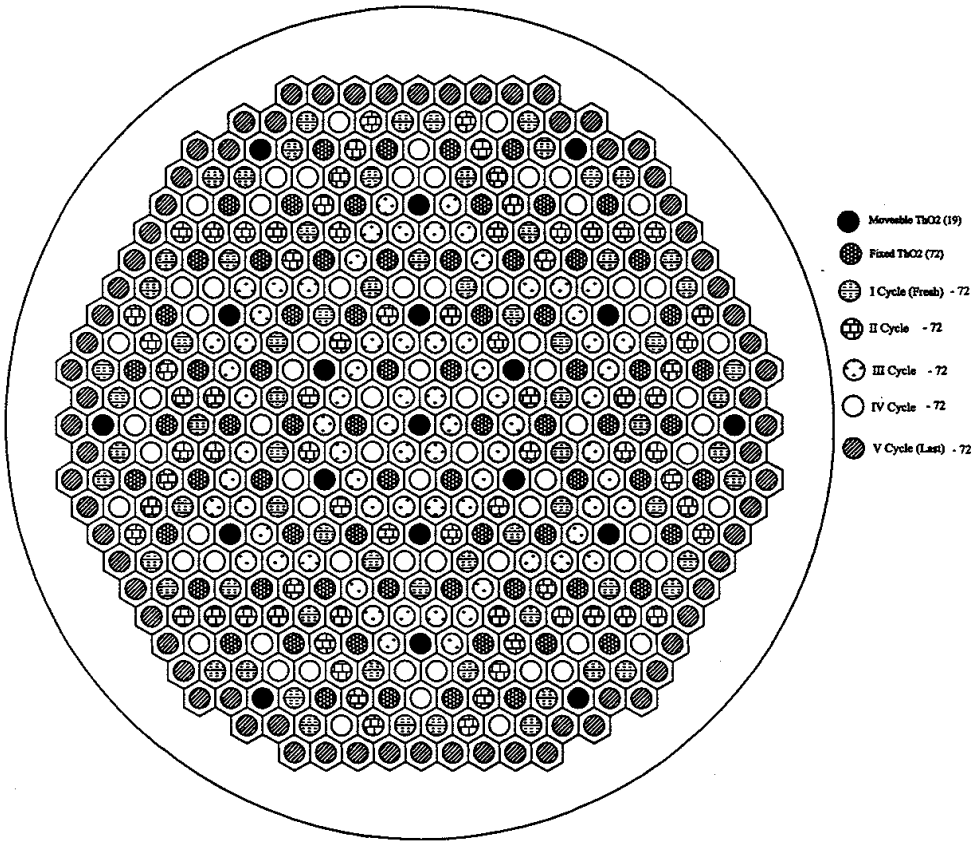

Figure 2. ATBR Core - 360 (eUO_2+ThO_2) + 91 ThO_2 Fuel Clusters 72 Assemblies/Batch - 5 Batches - Optimized Loading Pattern.

A new and unique feature of the core design is that the thoria rods require no external feed enrichment. The core consists of at least one batch size of such 30 rods thoria clusters. They are spread through out the reactor core except the one or two peripheral layers with twice the fuel assembly lattice spacing. They face the high thermal neutron flux similar to the one described in Fig. 1. By residing in the reactor core for one fuel cycle duration, they accumulate adequate ^{233}U. The irradiated thoria clusters are integrated with fresh enriched UO_2 seed fuel rods placed in two inner fuel rings. The integrated clusters undergo five more fuel cycles of operation following the shuffling scheme illustrated in Fig. 2. At the end of five cycles the thoria rods as well as enriched UO_2 rods attain a fairly high discharge burnup of

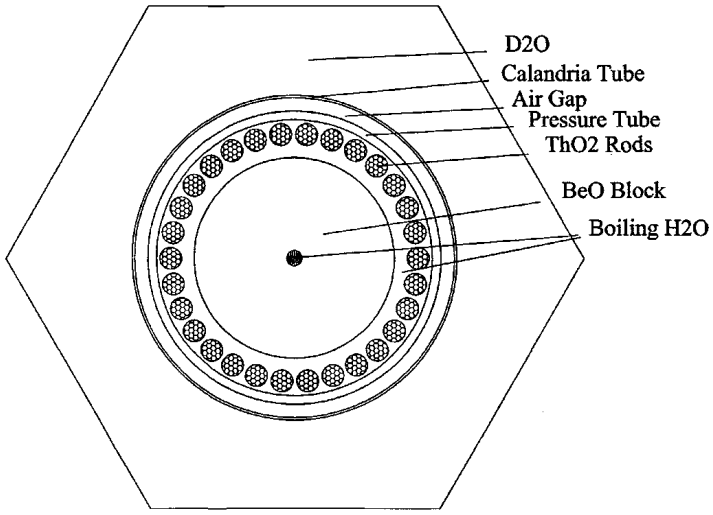

Figure 3. ATBR - 30 Rods ThO2 Fuel Cluster.

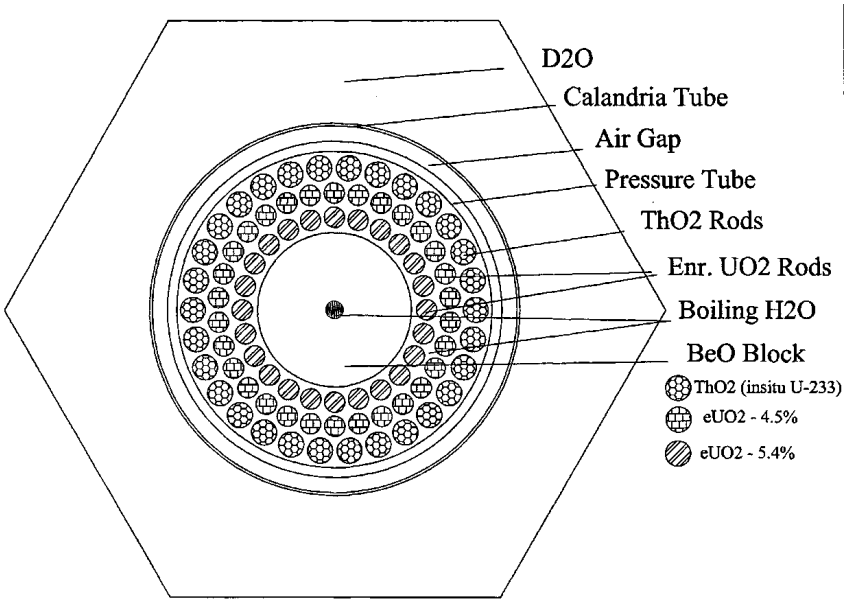

Figure 4. ATBR - 54 eUO2 + 30 ThO2 Rods Fuel Cluster.

32 GWD/T. The enrichment in UO_2 seed is about 5% ^{235}U . The fuel clusters consider some filler scattering material block like BeO in the centre. This arrangement was needed since the fuel cluster size was deliberately chosen to be large to achieve negative void coefficient. To minimize the power peaking within the fuel cluster, the central 37 fuel rods were removed and replaced by a scattering medium. More details of the core design are available in the references and are not reproduced here.

There are several interesting features of the new core design.

- At full power operation, there is practically no need for external reactivity control mechanisms since the K_{eff} variation is only 4 mk in 300 effective full power days. This can be nearly met by coolant inlet enthalpy variation.
- The power distribution is intrinsically maintained with comfortable thermal margins.
- The xenon override reactivity is about 20mk for full power operation. This can be provided by withdrawal of 19 moveable thoria clusters.
- The core can be deemed to be inherently safe since the most common transients involving reactivity excursions like rod ejection, loss of coolant, cold water addition etc are either absent or far less severe for this reactor.
- There is a sizeable production of ^{233}U which is intrinsically proliferation resistant due to formation of the isotope ^{232}U and high gamma emitting daughter products thereof. ^{232}U formation is however much lower owing to the neutron spectrum which is essentially a thermal one.
- Equilibrium loading of uranium and thorium is 50:50 by weight.
- There is no need for fuel reprocessing, if enriched UO_2 is available. Even in the closed fuel cycle options, the reprocessing load would be nearly halved, since 50% of the core can continue to use fresh ThO_2 in its natural form.
- Other types of seed zones employing either ^{233}U in natural uranium/thorium or plutonium in natural uranium/thorium are possible. Of these, the option of ^{233}U in thorium has the potential of being developed into a thermal breeder.

SUMMARY AND CONCLUSIONS

For utilization of thorium a new reactor concept with SGHWR like geometry is proposed. This reactor has two operational phases. In phase-I, it is a pre-breeder, i.e., an efficient ^{235}U to ^{233}U converter. Phase-II will be a breeder or at least a self-sustaining reactor system with (^{232}Th-^{233}U) oxide fuel. In our opinion, burning of thorium in the ambience of enriched UO_2 fuel is far superior to waiting for accumulation of plutonium from uranium reactors. Induction of thorium helps to cut down the uranium requirements. The new reactor concept has an overall better economic, operational and safety characteristics in comparison to any of the power reactor designs that are currently operational, albeit theoretically. There is no need for fuel reprocessing, if enriched UO_2 is available. Even in the closed fuel cycle options, the reprocessing load would be nearly halved, since 50% of the core can continue to use fresh ThO_2 in its natural form. This is permanent gain for future reactors. These reactors can use the same engineering design [6].

The present work is a theoretical study with the cross section data and calculation tools available with the author. Some uncertainties in the calculated results are admittedly present. Notwithstanding the above, it is claimed that the proposed reactor design has indefatigable design features which are convincingly superior to those of the power reactor designs prevalent today. It is mandatory to perform some physics experiments to refine the design parameters. The emphasis is laid more on the design philosophy rather than on the design parameters themselves.

ACKNOWLEDGEMENTS

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III. NUCLEAR AND REACTOR PHYSICS, NUCLEAR DATA AND CORE DESIGN

THORIUM UTILIZATION IN PWRs. NEUTRONICS STUDIES*

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Abstract. The main existing reactors can accept thorium fuel without fundamental modification. The aim of the study reported here and limited to PWRs is to bring neutron results up to date by using the CEA's most recent calculation codes, APOLLO 2, CRONOS 2 and PEPIN/DARWIN, associated with the last database, i.e. JEF 2.2. The study involves a 900 MW(e) PWR under three-batch core management and with annual cycles. Four types of fuel, U or Pu with a thorium carrier in the form of mixed oxides were examined. The thorium fuels produce few minor actinides and possess better conversion factors, i.e. unloaded fissile nuclei/loaded fissile nuclei, than the uranium fuels. The ^{233}U-^{232}Th fuel consumes only 12 kg/TW·h of uranium (85 kg/TW·h of ^{235}U in a conventional uranium fuel). The Pu-^{232}Th fuel offers excellent plutonium consumption potential (114 kg/TW·h) and produces a significant quantity (46 kg/TW·h) of uranium rich in ^{233}U (92%), the most effective isotope in a thermal spectrum. The kinetic and control parameters are acceptable. In open cycles, the radiotoxicity levels of unloaded thorium-fuels are lower than those of conventional uranium or MOX fuels up to 10^4 years of storage. The development of this cycle will require extensive R&D work and investment. However, a great part of the work has already been done: reprocessing by the THOREX process, remote control and shielding in the MOX industry, irradiations (Indian Point, Elk River, Shippingport etc.). Today's need to eliminate strategic materials such as High Enriched Uranium and Plutonium could launch the industrial implementation of a new fuel cycle to complement the uranium cycle.

1. INTRODUCTION

The thorium cycle in nuclear reactors, which has been studied since the Fifties, has experienced periods of frenetic activity followed by almost total oblivion. At present, there has been a revival of interest in this subject for the following four reasons:

Firstly, careful attention is being paid to managing long-lived radioactive waste, triggered in France by the Bataille law of 30/12/1991. The quantity of transuranics produced in the thorium cycles, thorium being lighter than uranium and plutonium, is clearly lower than in the conventional cycles as the transmutation stages required for them to form are longer. In practice this advantage is somewhat lessened by the fact that the essentially fertile, thorium-element fuels need to be used in conjunction with fissile elements, i.e. uranium or plutonium.

Secondly, studies have restarted on hybrid systems (Los Alamos, Carlo Rubbia), i.e. accelerators connected to a sub-critical installation implementing the thorium cycles. A proton beam hitting a target of heavy nuclei (lead, titanium, uranium, thorium etc.) creates a hard neutron flux by spallation which react with thorium or uranium fuel.

The fissile element ^{233}U is continually regenerated and the system is fuelled by thorium alone. According to the designers, these systems would be safer than conventional nuclear reactors (elimination of criticality accident), would not contribute to the proliferation of strategic materials and would produce little waste etc. These designs would require considerable technological development, in particular the implementation of high intensity proton beam, mastering the continuous reprocessing of molten salt fuels etc.

* 1997 meeting.

Thirdly, there is a policy of eliminating strategic nuclear materials [1], i.e. the highly enriched uranium and plutonium from dismantled nuclear weapons in the USA and the Former Soviet Union. Thorium, which does not generate plutonium, constitutes an excellent carrier for the "once through" scenarios where a maximum of fissile material is consumed, the remaining material being sent directly for deep disposal. The Uranium-233 created can be denatured by adding natural uranium.

Fourthly, consideration is being given to the importance of energy independence. The opinion is divided on this latter point. Some [2] maintain that abundant reserves of uranium ore, of around 20 million tonnes exist, despite the fact that at present the reasonably reliable resources at an acceptable cost (from 400 to 650 FF/kg of Uranium) are no more than 4.5 million tonnes, and virtually no prospecting is being carried out. Uranium extracted from sea water could supplement these stocks, constituting a practically inexhaustible source with 4.5 billion of tonnes dissolved in the form of carbonates, but the extraction processes which are currently known only on laboratory scale, involve costs varying from 1,300 to 2,500 FF/kg of uranium according to the different financial assumptions.

All these considerations are rather vague and involve a considerable amount of speculation. Primary energy requirements are going to double between now and 2020 with the increase of the world's population from 5 billion to 8.5 billion. For developed countries, in which a safe nuclear industry is feasible, the demand for electricity, which provides many of life's comforts, will increase even faster (in France by a factor of more than 2 over 20 years). For these countries, in some of which resources are scarce (France, Japan, Korea etc.), nuclear power offers clear advantages as it saves on fossil fuels, which are preferably kept for use in developing countries and the chemical industry.

Furthermore, nuclear industry releases are limited, which is favourable for health and the environment. Fission energy will remain indispensable and more than just financial aspects will need to be taken into consideration. Safety and stability of supply are essential. The idea of neglecting an abundant natural resource is unthinkable. According to these hypotheses, keeping in mind the relatively long time it will take for commercial start-up to take place, it is necessary to envisage mobilising nuclear energy resources straight away, thus, in addition to uranium, we could turn to thorium, the reserves of which appear to be considerable and easy to exploit at known cost. The potential of the nuclear industry and its lifetime have expanded considerably as have reliability and flexibility as a result of diversification of technologies and materials.

The main existing reactors: light water (pressurised [3] and boiling), heavy water reactors, high temperature reactors and fast reactors can accept a thorium-based fuel without requiring fundamental modification. Thermal reactors loaded with UOX, MOX or thorium fuel will produce plutonium of a given isotopic composition, which can be assimilated by fast reactors which, in turn, will provide uranium-233, the better fuel for thermal neutrons, by irradiating the thorium in the blankets.

These considerations have led us to re-open the thorium file. The study is restricted to pressurised water reactors which constitute the predominant existing technology (65% of installed nuclear power in 1996). Numerous results have been published since the Sixties, but they are fairly scattered, both as regards material flux and changes in the radiotoxicity of the unloaded fuel. The most recent calculation means developed by CEA (APOLLO2, CRONOS2, PEPIN) are used, together with nuclear data obtained from the JEF2.2 file.

We will briefly go over a few of thorium's properties and then take a closer look at how the four types of fuel change in a 900 MW PWR while generating the same amount of energy:

a) Thorium-232/High Enriched Uranium, enriched with ^{235}U, (HEU);
b) Thorium-232/First generation plutonium;
c) Thorium-232/Uranium with a high ^{233}U content (U_T) taken from spent fuel b). Two moderating ratios: 2 and 1.3;
d) Thorium-232/Medium Enriched Uranium, enriched with ^{235}U, (MEU).

All cores are homogenous, i.e. contain only one type of fuel in order to avoid zoning. The initial fissile material content required to assume a whole cycle, the balance of heavy nuclei from the beginning to the end of irradiation, a few physical parameters, the control worth, the radioactivity and the changes in radiotoxicity of unloaded fuels are assessed.

2. REVIEW OF THORIUM'S PROPERTIES

Thorium is considered to have the following three strong points:

Firstly, it is presumed to be abundant - as its half life is three times that of uranium-238 ($1.4x10^{10}$ years), and it is accepted that its reserves are larger and more advantageously distributed, however, as there is no market for it at present, it is not being prospected for.

Secondly, neutron quality is high in the thermal and epithermal fields of uranium-233, the fissile isotope resulting from irradiation of thorium-232. The fission cross-section is high, while that of capture is low. (Production/absorption by fission = 2.29, which makes it possible to envisage breeding in a thermal spectrum)[4].

Thirdly, the potential radiotoxicity of the irradiated fuel is lower. Being a lighter element than the uranium and plutonium isotopes, thorium-232 produces fewer minor actinides (americium and curium). Another advantageous parameter is the low capture cross-section of ^{233}U which restricts transmutation.

The disadvantage of using thorium-232 in reactors is that it is not fissile. The cycle must be started by mixing it with a fissile element, either ^{235}U, ^{239}Pu or ^{233}U.

Another drawback of the thorium cycle is the inevitable in-pile formation of uranium-232 (by n, 2n reaction on uranium-233). When this decays, two high-energy γ emitters are produced, thallium-208 (Eg = 2.6 MeV, T = 3 min., b) and bismuth-212 (Eg = 1.8 MeV T = 1h30, b; T = 3h, a), which complicate all handling operations (recycling, manufacture, transport, disposal etc.) and thus shielded and remote lines must be used. Paradoxically, this disadvantage could be considered as an advantage from the non-proliferation point of view.

Table I - Physical properties of the elements U, Pu and Th (metal and oxide).

Properties	**U**	**UO_2**	**Pu**	**PuO_2**	**Th**	**ThO_2**
Melting point (ºC)	1130	2760	632	2400	1750	3300
Phase change (ºC)	660				1400	
Theoretical density (g/cm^3)	18.9	10.96	19.8	11.50	11.7	10.00
Thermal conductivity (600ºC) W/cm/ºC	0.42	0.0452			0.45	0.044

Table I contains a few physical data associated with the main elements included in nuclear fuels. The melting point of thorium oxide (3,300ºC) is one of the highest of all refractory oxides. The specific power could be increased, as could the burnup (better fission product retention). The lower density (-10%) is a disadvantage for ThO_2 (lower concentration in heavy nuclei).

To specify the advantages and disadvantages of the different nuclear fuels used in several spectra and to obtain a thorough understanding of their behaviour, we will briefly reiterate a few neutron data associated with uranium, plutonium and thorium and their descendants.
Tables II and III show some of the parameters associated with fissile and fertile nuclei.

Table II - Mean fissile nuclei parameters in a thermal neutron spectrum.

	^{233}U	^{235}U	^{239}Pu	^{241}Pu
σ capture (barns)	46	101	271	368
σ fission (barns)	525	577	742	1007
$\alpha = \sigma_c/\sigma_f$	0.088	0.175	0.365	0.365
$\eta = \nu\sigma_f/\sigma_a$	2.300	2.077	2.109	2.151
Eff. β fact. pcm*)	270	650	210	490

*per cent of milli K (10^{-5} Δk/k)

Table III - Parameters of fertile nuclei.

	^{232}Th	^{238}U
σ capture (barns)	7.40	2.73
I.R.* capture (barns)	85	272
Fission cutoff (MeV)	1.5	0.8
Effective β factor (fast fission)	2030	1480

*I.R.: integral resonance in infinite dilution (0.625 eV at 20 MeV)

We propose to compare the uranium and thorium cycles by studying the behaviour of the main isotopes in the reactor [5] [6].

Fertile elements, ^{238}U and ^{232}Th

The **thermal capture** cross-section of thorium is around three times that of uranium-238, which would indicate enhanced conversion in a thermal reactor. However, the increase of absorption in the fertile element requires a greater quantity of fissile material, which would significantly affect the raw conversion factor (CF = fissile nuclei formed by capture/fissile nuclei eliminated by absorption), while reducing non-fissile capture in the moderator and the structures. On the other hand, if the spectrum is hardened (under-moderated reactors, HTRs etc.) the effect is reversed. The uranium-238 captures more in the epithermal field but the resonance self-shielding will reduce the consequences. In the fast field, the captures of the two isotopes are similar, however the UO_2 has a slight advantage as it also offers greater density (+10%).

The fission cross-section of ^{238}U in the fast field is three to five times that of ^{232}Th. In a conventional PWR, ^{238}U fission represents 7 to 8% of the total energy, whereas ^{232}Th fission only counts for 2%. Uranium is preferable to thorium as regards the conversion factor.

Fissile elements, ^{239}Pu, ^{233}U, ^{235}U, ^{241}Pu

The η factor = production/absorption ($\nu \sigma_f / \sigma_a$) characterises the potential use for an isotope. It can be noted that in a thermal spectrum, the best use is obtained by uranium-233 (low capture). In a fast spectrum, it is the isotope plutonium-239 which performs best.

The potential of ^{233}U remains substantial in a hardened spectrum (epithermal), Table IV. This property makes it possible to envisage breeding in a thermal reactor. In addition, its low capture cross-section limits the accumulation of even nuclei (^{234}U) and should enhance high burnup and multi-recycling.

Table IV - Average values of $\eta = \nu \sigma_f / \sigma_a$

Nuclei	**^{233}U**	**^{235}U**	**^{239}Pu**	**^{241}Pu**
η at 0.025 eV	2.30	2.07	2.11	2.15
η_{th} (averaged over the thermal spectrum of a standard PWR)	2.27	2.06	1.84	2.17
η_{epi} (averaged over the epithermal spectrum of a standard PWR)	2.16	1.67	1.88	2.49

Other fertile elements (^{234}U, ^{240}Pu) and non-fissile elements (^{236}U, ^{242}Pu)

^{240}Pu is better than ^{234}U because it engenders ^{241}Pu, which has a higher h factor than that of ^{235}U. The capture of ^{236}U is three times lower than that of ^{242}Pu, hence lower parasitic capture in the thorium cycle.

Properties of ^{233}Pa and ^{239}Np

^{233}Pa engenders b decayed ^{233}U, with a half life of **27 days**. In the same way, ^{239}Np engenders ^{239}Pu with a half life of **2 to 3 days**. With a constant flux, the concentration of ^{233}Pa would be 10 times that of ^{239}Np, resulting in greater losses, but this effect is limited. However, the large quantity of ^{233}Pa poses a problem on reactor shutdown, i.e. reactivity increases due to the disappearance of the latter, which absorbs neutrons and to the formation of fissile ^{233}U. For example, in a PWR loaded with ThO_2-^{235}U, reactivity increases successively from 310 to 820 to 1,210 pcm after shutdowns of 10, 20 and 30 days. There needs to be supplementary negative reactivity to compensate this effect which could reach 4,000 pcm. The absorption factor of ^{233}Pa depends on the flux level. It would be interesting to analyse the effect during a stretch-out.

Other parameters

- Fewer fission products absorption during the thorium/^{233}U cycle.
- There is less non-fission loss in the control rods, the moderator and the structures during the thorium cycle.
- There is a lower proportion of delayed neutrons during the thorium cycle (due to the ^{233}U, the effective beta factor of the ^{232}Th only affects the fast field where there are few reactions in the PWR spectrum) which accelerates the reactor kinetics.

Nuclei	^{233}U	^{235}U	^{239}Pu	^{241}Pu	^{232}Th*	^{238}U
β (pcm)	276	650	210	490	2030	1480

* fast fission

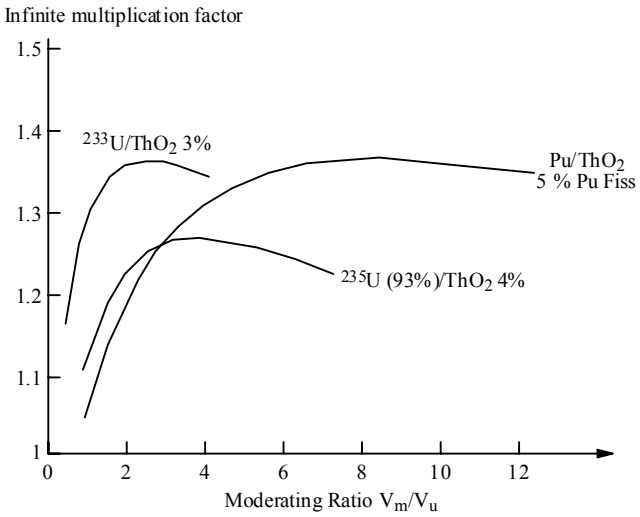

Figure 1. Change in infinite multiplication factor of a PWR cell as a function of the moderating ratio for a thorium fuel with each of the fissile isotopes.

Figure 1 shows how the infinite multiplication factor for a PWR cell changes as a function of the moderating ratio for the three fissile isotopes mixed with thorium. It can be seen that for ^{235}U-Th, the optimum is around 2 (standard value for the uranium cycle). For ^{233}U-Th the value is lower, and for Pu-Th it is greater.

To make optimum use of the ^{233}U-Th fuel, the moderating ratio must be reduced. This is also necessary to maintain a negative moderator temperature coefficient.

Nuclear data

The nuclear data primarily affect ^{232}Th and ^{233}U, then ^{233}Pa and ^{234}U produced by capture in the above nuclei. Then there is ^{232}U, which by successive decay produces the strong γ emitters ^{212}Bi and ^{208}Tl, and ^{230}Th from the decay of ^{234}U and source ^{232}U as well as ^{236}Pu. The ^{231}Pa ($T = 3.3 \times 10^4$ years) is the largest minor actinide component in this cycle. ^{228}Th is also included (chain of ^{232}U with the same γ emitters at the end), which is very radioactive (800 Ci/g) and is hazardous in the medium term.

The nuclear data used in the APOLLO code and relating to the uranium and MOX cycles in the PWRs have been extensively qualified [7] (CEA93 library from the JEF 2.2 file). The average discrepancies between experiments and calculations (effective multiplication factor) in the U and MOX lattices are weak, but there is considerable scatter (certain integral experiments used are very old).

In the case of thorium, the average discrepancy between experiments and calculations on the relatively high effective multiplication factor indicates that neutron data for the ^{232}Th and the ^{233}U are too scarce and that the experimental conditions are too uncertain (old integral experiments). For ^{233}U, a marked trend towards increasing the resonant capture cross-section can be observed. Discrepancies have been able to be reduced by taking the results from research on the trends into account. The average value of the residual discrepancy on the effective multiplication factor in the case of thorium becomes similar to those regarding uranium and plutonium. Note should be taken of the recent assessment of ^{233}U resonance parameters carried out by Derrien [8].

Current data from the CEA93 library regarding the isotopes included in the thorium chain are acceptable for the project calculations. Data has to be used in 172 groups (0 to 20 MeV) due to the importance of some cross-sections (n, 2n).

However, increased accuracy is to be desired in the future for the cross-sections of ^{232}Th, ^{233}U, ^{233}Pa, ^{234}U etc.

3. STUDY OF FUELS WITH A THORIUM CARRIER

Diffusion-evolution calculations were carried out on a conventional 17 x 17 grid 900 MW(e) PWR fuel assembly in an infinite medium using the APOLLO 2 code [9], part of the SAPHYR system (French acronym for Advanced Reactor Physics System) developed at CEA. It solves the multigroup equation of transport in space and energy, either by the collision-probability method (integral equation) or using Sn methods with nodal or finite difference techniques (integral-differential equation), in geometries of one or two dimensions.

This code, used by EDF, the operator, and FRAMATOME, the vendor, has been extensively qualified on the UO_2 and MOX fuelled PWRs and has participated in different benchmarks.

The library used (CEA93) contains an energy mesh of 99 groups ranging from 0 eV to 10 MeV and a second one of 172 groups covering the band from 0 eV to 20 MeV. The isotopes come from the JEF 2.2 assessment. The fission yields given for 10 fissile isotopes (^{232}Th, ^{233}U, ^{235}U, ^{238}U, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{241}Am, ^{242}Pu, ^{242}Am) come from the RIDER 81 compilation (77 fission products). The depletion chain for heavy nuclei, thorium and uranium, consists of 28 nuclei, 12 of which have self-shielded data: ^{237}Np, ^{232}Th, ^{233}U, ^{235}U, ^{238}U, ^{238}Pu ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu ^{241}Am and natural zirconium. These are recalculated every 10 GWd/tHM. The effects of interaction between the isotopes in space and energy are taken into account. To take into account the gradients flux, the fuel zones are divided into several rings, which are calculated separately. The depletion modules yield the isotopic concentrations for all media containing burnable nuclei, as a function of time, burnup or fluence.

Four types of fuel in the form of mixed oxides were adopted:

Case 1) Highly Enriched Uranium, (enriched in ^{235}U) (HEU)/^{232}Th

Case 2) First generation plutonium/^{232}Th.

Case 3) Uranium with a high ^{233}U content (U_T)/^{232}Th

- This uranium comes from reprocessing fuel 2.
- The study was performed with two moderating ratios, one standard (a) and one under-moderated (b). Only the latter led to an acceptable temperature coefficient at the beginning of the lifetime. To obtain $VH_2O/VUO_2 = 1.3$, the diameter of the fuel was increased from 8.2 mm to 9.4 mm. The mass of fuel in the core is increased, 31 tonnes per refuelling operation instead of 24, which complicates comparison with the other cases.

Case 4) Medium Enriched Uranium (enriched in ^{235}U) (MEU)/^{232}Th. Known as the denatured cycle.

It can be seen that cases 2 and 3 do not need direct spending on enrichment. In all cases, homogenous cores were envisaged with only one type of fuel assembly thus avoiding fuel zoning, which is costly at the manufacturing stage. Burnable poisons are not necessary, because in this study only short cycles are considered. These are 11,000 MW·d/t,

287 efpd/year (equivalent full power day), three-batch core management, average burnup 33 GW·d/t at unloading. The fissile nuclei contents are adjusted to ensure an entire cycle. The reactivity equivalence is obtained by imposing an infinite multiplication factor of 1.03 on the average core burnup at the end of the cycle (22 GW·d/t), zero boron. This choice will be confirmed subsequently by a core calculation.

The heavy nuclei balance between loading and unloading in an equilibrium state at shutdown is given in Tables V and VI for cases 3 and 3-b [after several months the ^{233}Pa (half life is 27 days) is transformed into ^{233}U]. The results are given for two burnup values, which makes it possible to see the influence of depletion. However, the fissile material contents are only adapted to the first case: unloading at 33 GW·d/t. Table VII gives some characteristic parameters for the four cases.

Table VII - Characteristic parameters (unloading at 33GW·d/t).

Fuel Type	**HEU/Th (1)**	**Pu/Th (2)**	**U_T/Th (3-b)**	**MEU/Th (4)**
Total content of fissile element (%)	3.9	6.5	3.07	19.0
Enrichment (fissile isotopes) (%)	93 (^{235}U)	70 ($^{239+241}$Pu)	92 ($^{233+235}$U)	20 (^{235}U)
Total fissile element consumption (kg/TW·h(e)) (%)	30.2 (U) 20.7	113.6 (Pu) 47.1	18.4 (U) 12.1	62.3 (U) 8.7
Minor actinide production (kg/TW·h(e)	1.91	7.13	0.50	2.14
Conversion factor* (^{233}Pa + Uf+Puf)	0.624	0.631	0.759	0.683

*CF = mass (loaded fissile nuclei/unloaded fissile nuclei)

The conversion factors (CF) are higher than in the uranium cycle (~ 0.5), hence a lower overall consumption of fissile material. In particular, the use of fuel 3-b (U_T/Th) leads to a low consumption of fissile material (12%) associated with a minimal production of minor actinides (0.5 kg/TW·h). However, the initial supply of ^{233}U is not easy: two type-2 cycles and the implementation of a reprocessing/fabrication line.

The Pu/Th fuel offers excellent potential for the use of plutonium (114 kg/TW·h) with a relatively modest production of minor actinides (7 kg/TW·h, or 6% of the plutonium consumed). It produces a substantial quantity of ^{233}U (46 kg/TW·h) which, depending on the opinion, is a useful fissile resource or else a weapons-proliferating material. In the latter case, it can always be denatured by adding natural uranium. It is to be borne in mind that this isotope is always associated with ^{232}U (0.3%) which is a strong g emitter due to its daughter products. Thorium is a good candidate for carrier when it comes to eliminating weapon grade Pu in reactors as it is strong, performs well, there are few minor actinides and the uranium formed can be easily denatured.

The kinetic coefficients were calculated into the assembly. They are shown in Table VIII.

The fuel (2) is affected by the presence of plutonium (low ppm of boron, effective beta factor and control rod worth). The low effective beta factor of the uranium-233 is apparent in the fuel (3) which, as well as with the standard moderating ratio, has a positive moderator coefficient. However, the kinetic coefficients are not fundamentally different from those of uranium fuel.

Table V - Combustible Pu/Th(2). Inventory in heavy nuclei at shutdown (kg)
900 MW(e) PWR in equilibrium: loading/unloading of 52 fuel assemblies.

BU (GW·d/t)	0		33			60	
Noyaux	Masses (kg)	Teneurs (isotopie) (%)	Masses (kg)	Teneurs (isotopie) (%)	Bilan (kg)	Masses (kg)	Teneurs (isotopie) (%)
^{228}Th	0		0.0057			0.022	
^{230}Th	0		5.05×10^{-5}			2.05×10^{-4}	
^{232}Th	22 277.		21 828.			21 308.088	
Total Th	22 277.	93.50	21 828.006	94.82	-449.	21 308.110	95.59
^{231}Pa	0		2.698			3.299	
^{233}Pa	0		20.829			25.603	
Total Pa	0		23.527	0.10	+23.527	28.902	0.13
^{232}U	0		1.032	(0.34)		2.718	(0.61)
^{233}U	0		271.700	(90.96)		370.254	(83.59)
^{234}U	0		22.440	(7.51)		56.221	(12.69)
^{235}U	0		3.327	(1.11)		12.394	(2.80)
^{236}U	0		0.216	(0.07)		1.350	(0.30)
^{237}U	0		5.34×10^{-4}	(0.)		0.003	(0.)
^{238}U	0		3.53×10^{-4}	(0.)		7.06×10^{-4}	(0.)
Total U	0		298.714	1.30	+298.714	442.941	1.99
^{237}Np	0		0.065			0.160	
^{238}Np	0		1.50×10^{-4}			4.68×10^{-4}	
^{239}Np	0		2.31×10^{-5}			3.69×10^{-5}	
Total Np	0		0.065	0.	+0.065	0.160	0.
^{236}Pu	0	(0.)	0	(0)		0.	(0.)
^{238}Pu	20.754	(1.36)	25.954	(3.21)		26.651	(6.35)
^{239}Pu	888.552	(58.11)	202.738	(25.09)		24.914	(5.93)
^{240}Pu	351.114	(22.96)	287.383	(35.56)		129.951	(30.96)
^{241}Pu	195.305	(12.77)	183.870	(22.75)		98.864	(23.55)
^{242}Pu	73.259	(4.79)	109.153	(13.51)		139.336	(33.20)
Total Pu	1 528.984	6.42	808.098	3.51	-720.884	419.716	1.88
^{241}Am	19.670		16.701			8.451	
^{242}Am	0		0.461			0.206	
^{243}Am	0		26.613			42.175	
Total Am	19.670	0.08	43.775	0.19	+24.105	50.832	0.23
^{242}Cm	0		4.887			4.537	
^{243}Cm	0		0.187			0.258	
^{244}Cm	0		12.056			32.382	
^{245}Cm	0		1.189			3.784	
Total Cm	0		18.319	0.08	+17.319	40.961	0.18
TOTAL	23 825.654	100.	23 020.500	100.	-805.154	22 291.622	100.

Table VI - UT/Th Fuel (3-b) - Inventory in heavy nuclei at shutdown (kg) 900 MW(e) PWR in equilibrium Loading/Unloading of 52 fuel assemblies (Higher fuel mass than in other cases to reduce the moderating ratio).

BU(GW·d/t)		0		33	
Nuclei	Mass (kg)	Isotopes (%)	Mass (kg)	Isotopes (%)	Balance (kg)
^{228}Th	0		0.0029		
^{230}Th	0		5.76×10^{-4}		
^{232}Th	30 347		29 383		
Total Th	30 347	96.63	29 383	97.19	-964
^{231}Pa	0		2.615		
^{233}Pa	0		44.901		
Total Pa	0		47.516	0.16	+47.516
^{232}U	3.070	(0.32)	3.175	(0.40)	
^{233}U	880.530	(91.61)	591.036	(73.95)	
^{234}U	67.570	(7.03)	158.917	(19.88)	
^{235}U	9.990	(1.04)	39.799	(4.98)	
^{236}U	0		6.324	(0.79)	
^{237}U	0		0.022	(0)	
^{238}U	0		0	(0)	
Total U	961.160	3.07	799.273	2.64	-161.887
^{237}Np	0		0.551		
^{238}Np	0		0.002		
^{239}Np	0		0		
Total Np	0		0.553	0.01	+0.553
^{236}Pu	0		3.04×10^{-6}		
^{238}Pu	0		0.142		
^{239}Pu	0		0.014		
^{240}Pu	0		0.002		
^{241}Pu	0		0.002		
^{242}Pu	0		3.05×10^{-4}		
Total Pu	0		0.161	0.01	+0.161
^{241}Am	0		2.17×10^{-5}		
^{242}Am	0		4.00×10^{-7}		
^{243}Am	0		3.76×10^{-5}		
Total Am	0		5.98×10^{-5}		
^{242}Cm	0		5.53×10^{-5}		
^{243}Cm	0		8.20×10^{-8}		
^{244}Cm	0		6.60×10^{-6}		
^{245}Cm	0		3.15×10^{-7}		
Total Cm	0		1.25×10^{-5}		
TOTAL	31 308.160	100	30 230.500	100	-1 077.660

Table VIII. - Physic parameters of the fuel assembly. Beginning of lifetime, 600 ppm, Pn.

Fuel type	HEU/Th	Pu/Th	U_T/Th (3)		MEU/Th	UO_2
	(1)	(2)	a) mod.R=2	b) mod.R=1.3*	(4)	(3.7% ^{235}U)
Soluble boron (pcm/ppm)	8.84	3.55	8.90	5.17	9.43	8.9
Doppler (pcm/°C) (650°-305°C)	-3.48	-3.29	-3.46	-4.40	-4.92	-2.6
Moderator coeff. (305°-285°C) pcm/°C	-6.0	-22.20	+5.2	-5.7	-12.2	-15.4
Global draining (pcm) (0-100% vacuum)	-105480	-68600	-97240	-95530	-97140	-70000
Effective beta factor						
0 GW·d/t	671	279	299	307	678	595
33 GW·d/t (pcm)	435	357	319	327	444	522 (core)
Control rod worth Ag-In-Cd (pcm)	36 980	23 790	33 430	33 210	36 340	34 000

*higher fuel mass (thick fuel rods)

4. RADIOTOXICITY

The DARWIN/PEPIN code [10] is used to calculate the activity (in Bq) and the radiotoxicity (in Sv) of the unloaded fuels. The code solves the differential equations of Bateman using analytical or numerical methods (Runge-Kutta). It is also possible to obtain the change in mass, residual power and the production of neutrons after reactor shutdown. This code is coupled with APOLLO 2. The decay data come from the JEF2.2 database, the dose factors come from ICRP 68 under the worst-case conditions. The cooling times vary from 5 to 10^6 years. The results (Figure 2) correspond to one tonne of heavy nuclei.

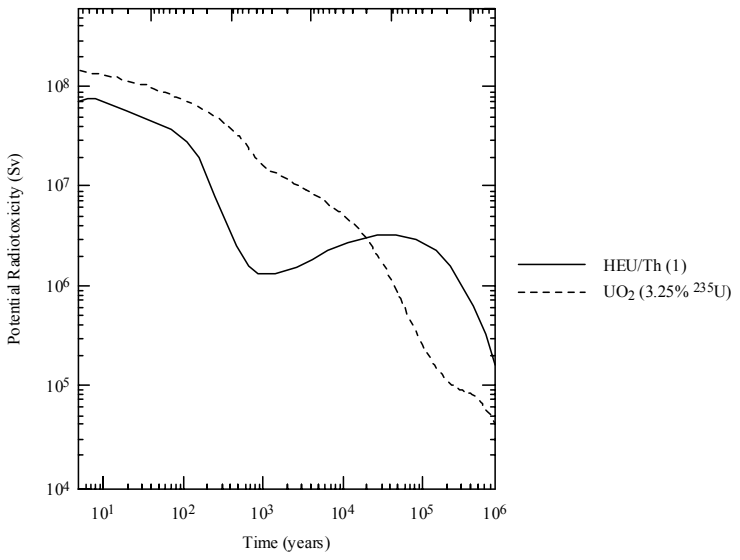

Figure 2. Radiotoxicity by ingestion for one tonne of heavy nuclei (spent fuel).

The level of radiotoxicity is lower than that of conventional UO_2 fuel. It decays rapidly for the first 10^3 years where the radiotoxicity is dominated by ^{238}Pu and ^{232}U. Beyond this, the dominant parts are ^{233}U, ^{241}Am, ^{229}Th and ^{227}Ac. At 50,000 years the dominant parts are ^{229}Th followed by ^{225}Ra.

The final level at 10^6 years is higher than that of conventional UO_2.
In reality, the thorium fuels are intended for multi-recycling and only the losses (reprocessing, manufacturing) will go into deep repositories.

5. CONCLUSION

By studying the four fuels, we have confirmed the advantage of thorium carriers which offer better conversion factors than in the uranium cycle and low minor actinide production. In particular, with ^{233}U/Th fuel, only 12% of the fissile material is consumed for a production of 0.5 kg/TW·h(e) of minor actinides (essentially ^{231}Pa). The Pu/Th fuel offers excellent potential for using plutonium with a consumption of 114 kg/TW·h(e) with relatively modest production of minor actinides (7 kg/TW·h(e)). A significant amount of uranium-233 is produced (46 kg/TW·h(e)) and could be considered as proliferating strategic material, but in this case it can easily be denatured by adding natural uranium. It would be more sensible to use it as fuel as it offers the best performance level in a thermal spectrum. The study confirms the feasibility of a PWR core loaded with thorium fuel as regards both physical parameters and control. The results as regards reducing radiotoxicity are lower than those currently stipulated in the literature but the study has been conducted in an open cycle, which does not correspond to a realistic scenario, except in the case of elimination of strategic materials. Nonetheless, this point is yet to be confirmed as is the qualification of some nuclear data.

Nuclear energy represents a durable benefit for our planet and its inhabitants, even though today it is considered fashionable to view it as merely temporary. But demographic pressure, energy demand and ecological requirements are realities which have to be faced. Thorium is an alternative which makes it possible to prolong the life of this source of energy and to diversify and stabilise supply. It would be good to take a new look at this type of fuel, in the light of the numerous studies which have already been carried out and taking into account recent technological progress, in particular in the MOX industry with regard to remote control and shielding. It should be remembered that many reactors of different types have operated with thorium-based fuel and that pilot plants for manufacturing and reprocessing (using the THOREX process) have existed. The main problem lies in the presence of gamma emitters, in the spent fuel, which complicate handling and reprocessing but which also limit proliferation (diversion and weapon design).

The proliferation aspect of a technology must gradually take second place to the increasing need for non-renewable primary energy sources. Politically stable nations are setting aside more and more resources to fight against terrorism, the elimination of which is fundamental to their survival. In addition, making an abundant energy source available at a reasonable price, will contribute to the economic development of poorer countries, which should reduce political unrest. The potential of the nuclear industry and its lifetime will be considerably increased, as will reliability, through the diversification of technologies and materials. Thermal reactors loaded with uranium, MOX or thorium will produce plutonium of sufficient quality to be used in fast reactors, which, in turn, could provide uranium-233 by irradiating the thorium in the blankets, which is a better fuel in thermal spectra. Great flexibility of supply would be achieved. The industrial implementation of thorium would require extensive R&D work to be carried out, but today considerable knowledge and a certain amount of

experience is available. This cycle is not really new. The need in the short term to eliminate strategic materials, i.e. highly enriched uranium and plutonium-239, could serve as a spring board for the industrial implementation of this cycle.

ACKNOWLEDGEMENTS

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MOLTEN SALT REACTOR BENCHMARK PROBLEM TO CONSTRAIN PLUTONIUM*

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Abstract. A molten salt reactor (MSR) based on ^{233}U-Th cycle has a positive feature from the standpoint of neutron economy, and it has been studied in Japan to utilize the MSR for the incineration of minor actinides (MA) produced in light water reactors (LWR). Therefore, Japan has proposed to study a MSR in the frame of the IAEA co-ordinated research project on the "Potential of thorium-based Fuel Cycles to Constrain Plutonium and to Reduce Long-term Waste Toxicities". It is important to study the basic character of the reactor with simple model especially for the purpose of comparison with other type of reactors. This report presents the results of the benchmark calculation and the effect of fuel volume ratio for the burnup characteristics.

1.INTRODUCTION

It is considered that a molten salt reactor(MSR) based on ^{233}U -Th cycle is one of the best reactor system from the standpoint of neutron economy, and it has been studied in Japan to utilize the MSR for the incineration of minor actinides (MA) produced in light water reactors (LWR) [1,2,3]. Therefore, Japan has proposed to study a MSR in the IAEA research co-ordinated meeting on the "Potential of Thorium -based Fuel Cycles to Constrain Plutonium and to Reduce Long-term Waste Toxicities" held in Vienna in October 1996. However, in the case of MSR, at least in principle, it is possible to make continuous fuel loading or extraction of poison material (such as Xe or ^{233}Pa), it is important to study the basic character of the reactor with simple model especially for the purpose of comparison with other type of reactors.

In this respect, a benchmark problem is prepared under the following considerations:

(1) To make comparison with the first stage of benchmark problem of IAEA LWR lattice [4], a simple two region cell of graphite moderator and fuel, in which the fuel salt flows in the central circular hole opened at the center of graphite hexagonal column. This is basically the lattice design of MSR proposed by K. Furukawa [5].
(2) Fuel salt is the mixture of LiF-BeP2-ThF(-PuF3 for the initial loading and plutonium concentration must not exceed 1 mol %.
(3) Fuel salt does not flow.
(4) To make the comparison with the High Temperature Gas Cooled Reactor which is proposed for the second stage of benchmark [6], the reactor power of 200 MW(th) is assumed, although it does not directly appear in the present calculation.

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* 1997 meeting.

(5) Plutonium vector is same as the IAEA LWR benchmark.
From the past experience, the following parameters are settled.
(6) Fuel salt volume ratio is 0.1, and the face to face distance of the moderator graphite hexagon is 40.0cm. The graphite density is 1.84 g/cm^3.
(7) The power density of the fuel salt is 100 W/cm^3 and the average temperature is 900°K.

According to the preliminary calculations, it was found that the k-inf is not sensitive to the plutonium concentration, and finally, the following fuel salt composition was determined for the initial loading: LiF-BeP2-ThF4-PuF3= 72-16-11.8-0.2 mol %. The required quantities for the calculation is same as the IAEA LWR benchmark. That is,
(1) k-inf at burnup of 20, 40, 60 MW·d/kg of Heavy Metal;
(2) nuclide densities (n/cm^3) from Th through Cm at above burnup points;
(3) the total neutron flux;
(4) average one group cross sections for capture, fission and (n,2n) reactions at a fuel burnup of 0 and 60 MW·d/kgHM;
(5) average energy per fission.

2. RESULT OF BENCHMARK CALCULATION

Figure 1 shows the definition of the problem.

Case 1 Normal Case
Specification of a cell (infinite lattice): two zone composed of hexagonal graphite column with circular channel for flowing salt at the center (Fig .1). Side length of the hexagonal is 23.1 cm and the radius of the central zone is 6.64 cm (fuel volume ratio is 1).

(Fuel: LiF-BeF2-ThF4-PuF3=72 : 16 : 11.8 : 0.2 mol% , Graphite : 1.840 g/cm^3)
Temperature : 900°K Average Power: 100 W/cm^3 for fuel salt) (constant)

Initial nuclide density (n/cm^3)

Fuel salt	
Th-232	3.778E-03
Pu-238	6.359E-07
Pu-239	3.906E-05
Pu-240	1.513E-05
Pu-241	5.018E-06
Pu-242	3.177E-06
Li-7	2.260E-02
Be-9	5.037E-03
F-19	4.978E-02

Graphite salt
C-12 9.226E-02

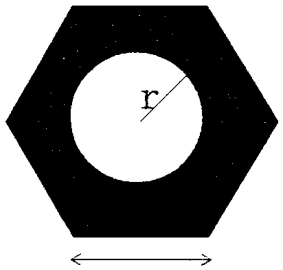

Figure 1. Specification of MSR Benchmark problem for plutonium burning for different cases.

For this problem, three institutions, Tohoku University (A), Nagoya University (B) and Toyohashi University of Technology (C), participated. The methods are: SWAT [7] in A, SRAC-95 [8] in B and SRAC [9]+ ORIGEN-2 [10] in C, respectively. SWAT is a burnup code based on SRAC and ORIGEN-2 connected by sub-codes for the preparations of cross sections for the burnup calculation and appropriate input for both main codes. SRAC-95

includes a burnup code called COREBN which is based on the analytical solution of the burnup equation. At present,29 actinide nuclides from ^{230}Th to ^{246}Cm can be treated. As for the fission products (FP), 26 nuclides and 4 pseudo FPs are included. The method used in C is same as A, but the input preparation at each burnup step is carried out manually. The cross section library based on JENDL-3.2 is used in all the calculations, however, A uses data based on ENDF/B4 for Li, Be and F.

Table I, II, III show the change in k-inf., the total neutron flux and the average energy per fission with burnup, respectively. The change in k-inf. is also shown in Fig. 2. Table IV, V, VI show the values of (total plutonium/total initial plutonium), (fissile plutonium/total plutonium) and (minor actinide/total initial plutonium), with burnup, respectively, where the minor actinide means Np, Am and Cm. Table VII shows (^{233}Pa + ^{233}U)/(total initial plutonium) with burnup which is the indication of the quantity of ^{233}U when the fuel is taken out at that burnup point. Table VIII is the summary of one group cross section at 0 and 60 MW·d/kg.

Since all the calculations are based on similar methods and cross sections, the tendency is same. As shown in Fig. 3, at first, plutonium decreases very rapidly and k-inf. also decreases very sharply. Actually most of ^{239}Pu is consumed by 20 MW·d/kg and at this point most of the fissile plutonium(~97%) is ^{241}Pu. Since the reactor power is kept constant, the flux increases and this promotes the conversion to ^{233}U from Th. Since η of ^{233}U is larger than for those of plutonium isotopes, k-inf. turns to increase and it reaches equilibrium at around 40 MW·d/kg of burnup. Total plutonium decreases steadily toward 60 MW·d/kg of burnup. The isotopic composition also changes and at 60 MW·d/kg of burnup, ^{242}Pu occupies about 96% in total plutonium.

Burnup (MW·d/kg)	A	B	C
0	1.14850	1.14165	1.13229
10	0.80344	0.74987	0.74777
20	0.77196	0.80440	0.77989
30	0.85391	0.84888	0.82595
40	0.86573	0.85542	0.83474
50	0.86369	0.85117	0.83377
60	0.85877	0.84502	0.83007

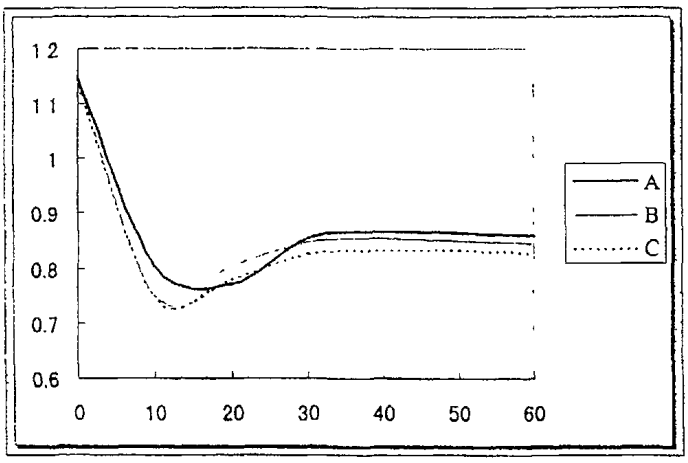

Figure 2. Change of k-inf vs. H. M. burnup (Casel)

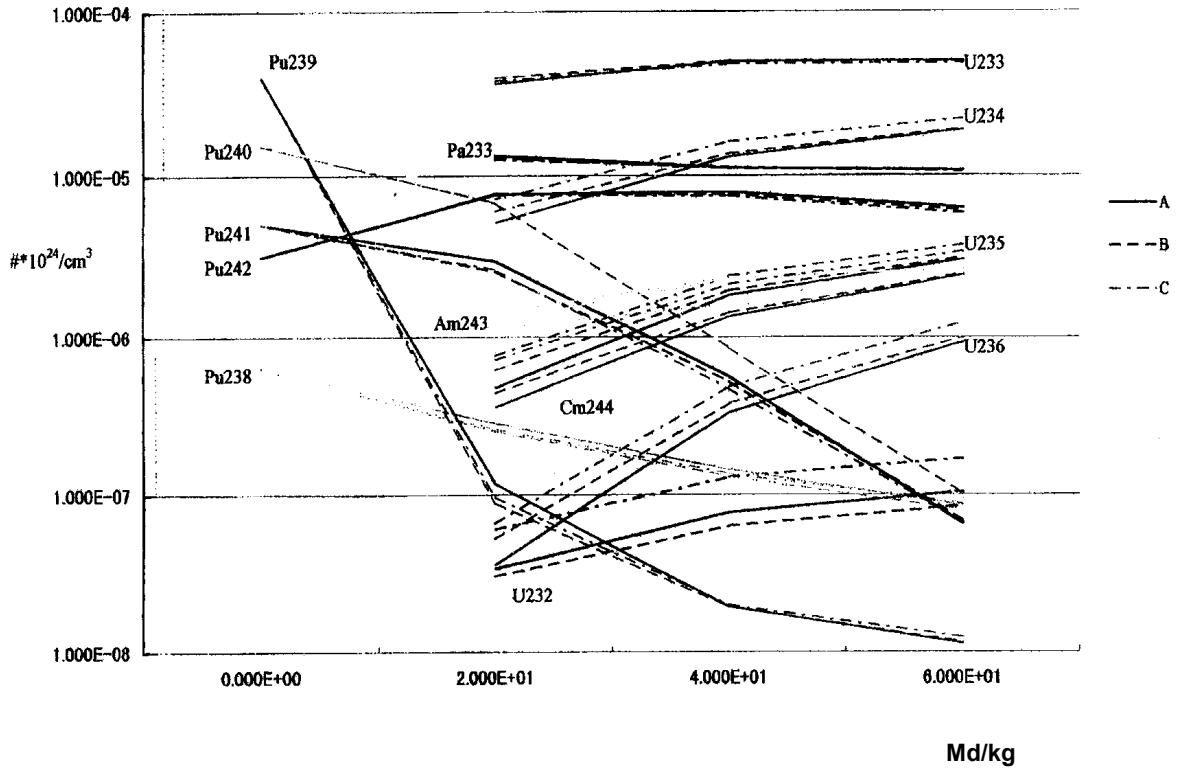

Figure 3. Change in nuclide densities with burnup.

Table I. Change in k-inf. vs. H.M. burnup.

Burnup MW·d/kg	0	10	20	30	40	50	60
A	1.14850	0.80344	0.77196	0.85391	0.86573	0.86369	0.85887
B	1.14165	0.74987	0.80440	0.84888	0.85542	0.85117	0.84502
C	1.13229	0.74777	0.779890	0.82595	0.83474	0.83377	0.83007

Table II. Total Neutron Flux vs. H.M. burnup (n/cm^{2}s).

Burnup(MW·d/kg)	0	20	40	60
A	3.510(E+14)	5.560(E+14)	5.030(E+14)	5.020(E+14)
B	3.229(E+14)	5.335(E+14)	4.903(E+14)	4.909(E+14)
C	3.27(E+14)	5.33(E+14)	4.88(E+I4)	4.74(E+14)

Table III. Average Energy per Fission vs. H.M.burnup (MeV).

Burnup(MW·d/kg)	0	20	40	60
A	210.6	202.1	201.1	201.1
B	211.8	202.4	200.8	200.6
C	211.3	202.4	200.6	200

Table IV (Total Pu/Total Initial Pu) vs. H.M. burnup.

Burnup (MW·d/kg)	0	20	40	60
A	1	0.3014	0.1513	0.1022
B	1	0.2776	0.1464	0.1022
C	1	0.280	0.144	0.0993

Table V. (Pu-fiss./Total Pu) vs. H.M. burnup.

Burnup (MW·d/kg)	0	20	40	60
A	0.6994	0.1635	0.0611	0.0120
B	0.6994	0.1520	0.0566	0.0108
C	0.6994	0.1564	0.0544	0.01

Table VI. (Minor Actinides/Initial Total Pu) vs. H.M. burnup.

Burnup (MW·d/kg)	0	20	40	60
A	0	0.0269	0.0603	0.0853
B	0	0.0318	0.0632	0.0868
C	0	0.0321	0.0644	0.0857

Table VII. ([^{233}Pa + ^{233}U]/Total Initial Pu) vs. H.M. burnup.

Burnup (MW·d/kg)	0	20	40	60
A	0	1.117	1.362	1.375
B	0	1.406	1.369	1.375
C	0	1.107	1.299	1.303

Table VIII-1. Cross sections at burnup = 0 MW·d/kg (barn).

Institution	A		B		C	
Nuclide	σ-f	σ-c	σ-f	σ-c	σ-f	σ-c
Th-232	0.0139	1.494	0.0137	1.544	0.014	1.636
Pa-233	0.0689	28.42	0.0676	28.53	0.068	31.44
U-233	84.37	10.14	88.09	10.49	94.40	11.26
U-234	0.3137	28.33	0.3085	28.92	0.306	31.23
U-235	70.63	14.44	74.61	15.04	79.70	16.23
U-236	0.2455	10.08	0.2427	10.02	0.252	11.39
U-238	0.0560	7.566	0.0551	7.477	0.056	8.263
Np-237	0.3420	48.97	0.3365	49.15	0.334	53.40
Np-239	0.363	17.91	0.3616	16.62	0.355	18.42
Pu-238	2.819	50.70	2.904	54.11	3.028	57.67
Pu-239	201.4	116.2	205.4	117.5	217.9	124.5
Pu-240	0.3781	118.2	0.3727	120.2	0.369	127.8
Pu-241	201.2	70.33	209.1	72.48	222.8	77.15
Pu-242	0.2687	31.43	0.2643	31.78	0.262	34.04
Am-241	1.384	188.2	1.347	181.5	1.430	196.2
Am-242m	1136	223.7	1206	237.7	1286	253.4
Am-243	0.3050	60.25	0.3017	60.86	328.8	69.0
Cm-242	1.415	5.124	1.416	4.863	1.469	65.38
Cm-243	121.4	19.39	125.3	20.25	134.5	21.67
Cm-244	0.6454	18.90	0.6432	18.56	0.667	21.78

Table VIII-2. Cross sections at burnup = 60 MW·d/kg (barn).

Institution	A		B		C	
Nuclide	σ-f	σ-c	σ-f	σ-c	σ-f	σ-c
Th-232	0.0110	1.869	0.0109	1.916	0.011	1.994
Pa-233	0.0547	21.75	0.0545	21.67	0.054	23.82
U-233	115.3	12.65	118.1	12.93	124.2	13.68
U-234	0.2460	25.21	0.2451	25.92	0.241	27.53
U-235	106,4	20.34	109.5	20.76	114.4	21.83
U-236	0.1836	7.076	0.1821	6.989	0.193	7.936
U-238	0.0443	5.774	0.0440	5.691	0.044	6.467
Np-237	0.2739	58.94	0.2729	58.86	0.269	60.81
Np-23	0.2575	17.24	0.2925	15.99	0.284	17.40
Pu-238	3.387	7.433	3.481	77.76	3.581	80.90
Pu-239	375.9	252.2	360.8	213.4	379.2	224.7
Pu-240	0.3246	216.9	0.3229	212.5	0.319	228.5
Pu-241	335.7	121.8	335.9	120.0	352.3	126.0
Pu-242	0.2173	21.18	0.2131	21.59	0.209	24.02
Am-241	2.198	329.6	2.018	298.8	2.119	316.4
Am-242m	1800	355.7	1855	366.9	1940	383.7
Am-243	0.2464	47.40	0.2454	47.09	0.249	52.05
Cm-242	1.62	5.582	1.589	5.238	1.634	5.618
Cm-243	155.6	25.68	157.6	26.35	166.7	27.71
Cm-244	0.5933	15.27	0.5968	14.90	0.609	16.58

On the other hand, minor actinides increase almost linearly with burnup. Looking at the minor actinides in detail, those of more than 95% are ^{243}Am and ^{244}Cm, which have relatively small absorption cross sections, though the ratio somewhat decreases with burnup. Although there are some discrepancies in the range around 20 MW·d/kg of burnup, the results at 60 MW·d/kg of burnup almost agree.

3. EFFECT OF FUEL VOLUME RATIO

It is interesting to see the effect of the fuel volume ratio since the neutron spectrum and the effective cross sections are mainly determined by the moderator volume ratio. Therefore, in addition to the above benchmark calculation (case 1), cases with $V_p/V = 0.05$ (case 2) and $V_p/V = 0.20$ (case 3) were studied with SWAT system. The volume ratio was changed by varying the radius of the molten salt region at the center. It is 4.697 cm for case 2 and 9.395cm for case 3, respectively. The initial Pu-mol% in the fuel salt was determined so that the initial k-inf. to be the same as in case 1, and it was 0.16% for case 2. However, for case 3, we adopted 0.6 mol% of plutonium since this value is recommended as the upper limit of plutonium concentration. The initial nuclide densities of fuel salt are given in Table IX.

Table IX. Initial nuclide densities for fuel salt.

Case	Pu (mol%)	Th-232	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242
case 1	0.2	3.778(-3)	6.359(-7)	3.906(-5)	1.513(-5)	5.013(-6)	3.177(-6)
case 2	0.16	3.791(-3)	5.087(-7)	3.125(-5)	1.210(-5)	4.014(-6)	2.542(-6)
case 3	0.6	3.650(-3)	1.908(-6)	1.172(-4)	4.539(-5)	1.505(-5)	9.53

Those of Li-7, Be-9, F-19 are same as nominal case.

The power density is kept to be 100W/cm^3 for the fuel salt. Thus, the volume of the reactor core should be doubled for case 2 and should be halved for case 3 from case 1, although this effect does not taken into account for the present study.

Figure 4 shows the change in k-inf. with burnup. It is interesting that k-inf. shows similar character for case 1 and case 2, but for case 3 the curve is quite different.

Burnup (MW·d/kg)	A	B	C
0	1.14850	1.14165	1.13229
10	0.80344	0.74987	0.74777
20	0.77196	0.80440	0.77989
30	0.85391	0.84888	0.82595
40	0.86573	0.85542	0.83474
50	0.86369	0.85117	0.83377
60	0.85877	0.84502	0.83007

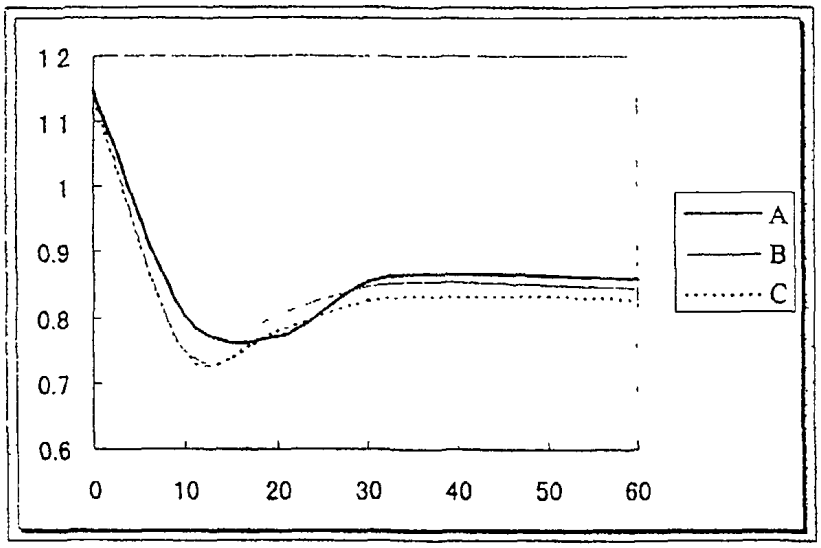

Figure 4. : Change of k-inf. vs. H. M. burnup.

This fact can be explained from the difference in the effective cross sections. For instance, the one-group effective fission cross sections at 0 MW·d/kg of burnup for case 1, 2, 3 are 225b, 303b and 107b, respectively. Due to the large fission cross sections for case 1 and case 2, ^{239}Pu is consumed very rapidly and the reduction of ^{239}Pu concentration increases the effective fission cross section even more since the neutron spectrum becomes softer with the decrease of materials with large cross sections. The effective fission cross section of ^{239}Pu becomes 379b at 20 MW·d/kg of burnup for case 1, which is 1.7 times as large as at 0 MW·d/kg of burnup and it reaches 376b at 60 MW·d/kg of burnup. Thus most of fissile plutonium isotopes are destroyed by 20 MW·d/kg of burnup. In contrast, for case 3, the effective fission cross section of plutonium at 0 MW·d/kg is relatively small and the decrease of plutonium concentration is slow, therefore, the behavior is somewhat similar to the IAEA benchmark case for LWR lattice, which gives the values of Pu-total/Pu initial = 0.16, Pu-fissile/Pu total =0.18 and Minor actinide/Initial Pu = 0.0687 at 60 MW·d/kg of burnup. Table X and XI presents the results of the total neutron flux, the average energy per fission.

Table X. Total neutron flux vs. HM-burnup (n/cm2s) with volume ratio change.

Burnup (MW·d/kg)	0	20	40	60
case 1	3.510(E+14)	5.560(E+14)	5.030(E+14)	5.020(E+14)
case 2	2.630(E+14)	4.340(E+14)	4.030(E+14)	4.050(E+14)
case 3	4.360(E+14)	5.110(E+14)	5.870(E+14)	6.180(E+14)

Table XI. Average energy per fission vs. burnup (MeV) with volume ratio change.

Burnup (MW·d/kg)	0	20	40	60
case 1	210.6	202.1	201.1	201.1
case 2	210.6	201.6	201.2	201.6
case 3	211.3	207.9	203.2	202.8

For case 1 and 2, the total neutron flux initially increases with bum up because of the decrease in fissile plutonium concentration. the flux shows a peak around 20 MW·d/kg of burnup due to the increase of ^{233}U nuclide density. For case 3,the flux increases monotonically to compensate the decrease in the fissile plutonium nuclide density. The average energy per fission clearly shows the contribution of main fissioning nuclide at that time.

From Table XII to XV, changes of (total plutonium/total initial plutonium), (fissile plutonium/total plutonium), (minor actinides/total initial plutonium) and (^{233}Pa + ^{233}U)/(total initial plutonium) for different fuel salt volume ratio with burnup are presented, respectively.

Table XII (Total Pu/Total Initial Pu) vs. HM-burnup with volume ratio change.

Burnup (MW·d/kg)	0	20	40	60
case 1	1	0.3014	0.1513	0.1022
case 2	1	0.2379	0.1302	0.0957
case 3	1	0.6530	0.3714	0.2070

Table XIII. (Pu-fiss./Total Pu) vs. HM-burnup with volume ratio change.

Burnup (MW·d/kg)	0	20	40	60
case l	0.6994	0.1635	0.0611	0.0120
case 2	0.6994	0.1447	0.0543	0.0139
case 3	0.6994	0.4994	0.2738	0.1738

Table XIV. (Minor actinides/initial total Pu) vs. HM-burnup with volume ratio change.

Burnup (MW·d/kg)	0	20	40	60
case 1	0	0.0269	0.0603	0.0853
case 2	0	0.0125	0.0330	0.0507
case 3	0	0.0170	0.0370	0,0587

Table XV. ([Pa-233 + U-233]/Total Initial Pu) vs. H.M.Bumup with volume ratio change.

Burnup(MW·d/kg)	0	20	40	60
case 1	0	1.117	1.362	1.375
case 2	0	1.406	1.633	1.625
case 3	0	0.2611	0.4560	0.5372

It is seen that (total plutonium/total initial plutonium) and (fissile plutonium/total plutonium) are smallest for case 1, though the (minor actinide/total initial plutonium) is highest for the case. From these observations, the choice of $V_p/V = 0.10$ for the benchmark calculation seems to be appropriate for the present purpose.

4. SUMMARY

For the first step to investigate a molten salt reactor for the purpose "to Constrain Plutonium and to Reduce Long-term Toxicities", a benchmark problem was constructed. Three institutions participated to solve the problem. Since their methods are basically the same, the final results are similar although some discrepancies exist in the course of burnup. The effect of fuel volume ratio which affects the neutron spectrum was also investigated. It turns out that the volume ratio of $V_p/V = 0.1$ selected for the benchmark is suitable to destroy plutonium effectively. Since the decrease of k-inf. is very rapid due to the destruction of fissile plutonium, the means should be taken to keep the change in k-inf. below certain range by the addition of plutonium salt properly. The strategy for this addition will be the task for next stage.

ACKNOWLEDGEMENT

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NUCLEAR DATA EVALUATION AND EXPERIMENTAL RESEARCH OF ACCELERATOR DRIVEN SYSTEMS USING A SUBCRITICAL ASSEMBLY DRIVEN BY A NEUTRON GENERATOR*

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Abstract. The research in the field of creation of evaluated nuclear data libraries for fissile nuclides, investigation of nuclear reaction mechanism in the range of high energies and development of calculation methods for characteristics of an electronuclear reactor are being carried out at National Academy of Sciences of Belarus since early 70s. The possibility of using low energy accelerators to investigate physical characteristics of subcritical target/blanket systems follows from the mechanism of nuclear reactions in high (1GeV) and low (15-20 MeV) energy ranges as well as from features of nucleon-mesons cascade development. It was shown that the spallation neutron source can be simulated by neutrons escaping from heavy element targets bombarded by 14 MeV neutrons. It was a reason for creation of an experimental facility consisting of a subcritical target/blanket system driven by a high intensity (1.5-2.0 10^{12} neutrons/s) neutron generator.

1. INTRODUCTION

Possibility of using thorium for ^{233}U production is very important because of its high abundance and good nuclear data which improve physics characteristics of NPPs. The thorium fuel cycle can be used in modem nuclear reactors of all types with keeping up main design peculiarities and safety of nuclear power plants. Using thorium-uranium cycle in the frame of subcritical systems driven by high energy accelerators was considered in detail by C. Rubbia [1] and by H. Takahashi [2]. K. Furukawa proposed a THO -NES concept based on using Molten Salt Reactors and Molten Salt Breeders driven by accelerator [3].

It is obvious that many characteristics of the thorium-uranium cycle including a reprocessing technology are determined by accuracy of nuclear data in a wide range of energies and mass numbers, The research in the field of creation of evaluated nuclear data libraries for fissile nuclides, investigation of nuclear reaction mechanism in the range of high energies and development of calculation methods for characteristics of an electronuclear reactor are being carried out at National Academy of Sciences of Belarus since early 70s. During this period the neutron cross-section libraries for ^{233}U, ^{235}U, ^{238}U, ^{232}Th, ^{241}Am, ^{242}Am, ^{243}Am were created and included into the BROND-2 library (CJD, Obninsk, Russia).

During the last three years these investigations were supported within framework of the Project B-03 “Actinide Nuclear Data Evaluation” (ISTC, Moscow, Russia).

2. NUCLEAR DATA EVALUATION

In contrast to uranium-plutonium fuel cycle for thorium-uranium fuel cycle experimental and evaluated data are rather scarce which leads to different libraries of nuclear data and therefore to significant differences even in such integral characteristic like k_{eff}. One can see the differences in the Fig. 1 where the data on k_{eff} are presented by participants of neutron

* 1998 meeting.

benchmark on accelerator driven systems (ADS) with thorium-uranium cycle initiated by IAEA. It is seen that the k_{eff} values considerably differ from one participant to another depending on nuclear data library and computer code used.

The differences in cross-sections σ_f, σ_{n2n} and σ_{n3n}. taken from different libraries are presented in the Figs. 2 and 3 together with our evaluation performed using nuclear systematic validated for well experimentally investigated nuclei (^{238}U, ^{235}U, ^{233}U and others).

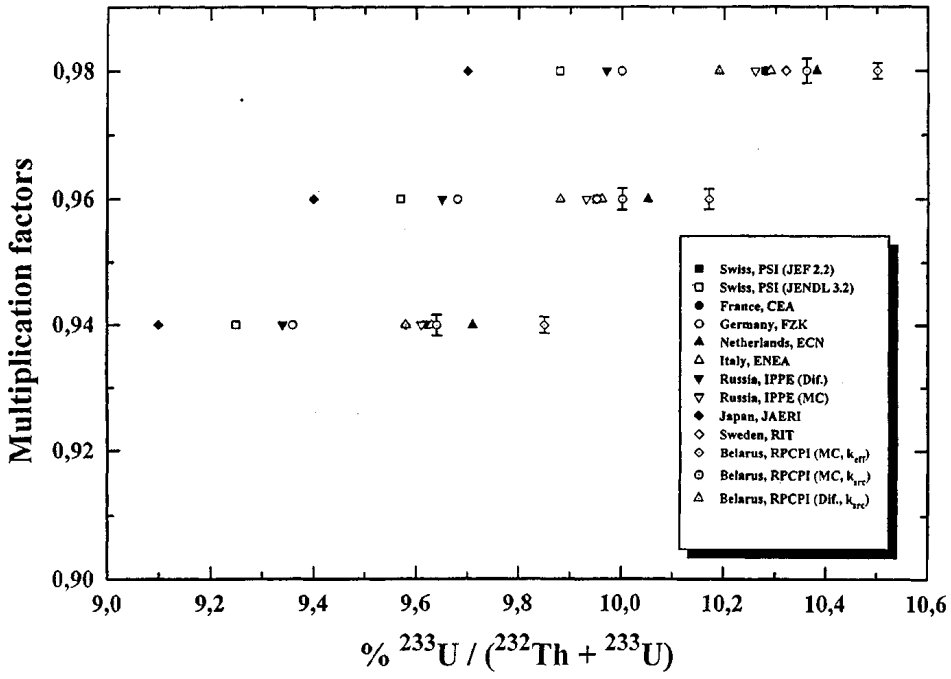

Figure 1. Results of IAEA benchmark on Th-U ADS.

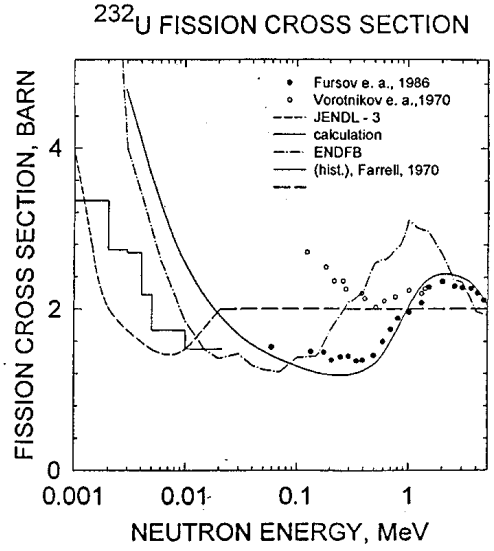

Figure 2. ^{232}U fission cross section.

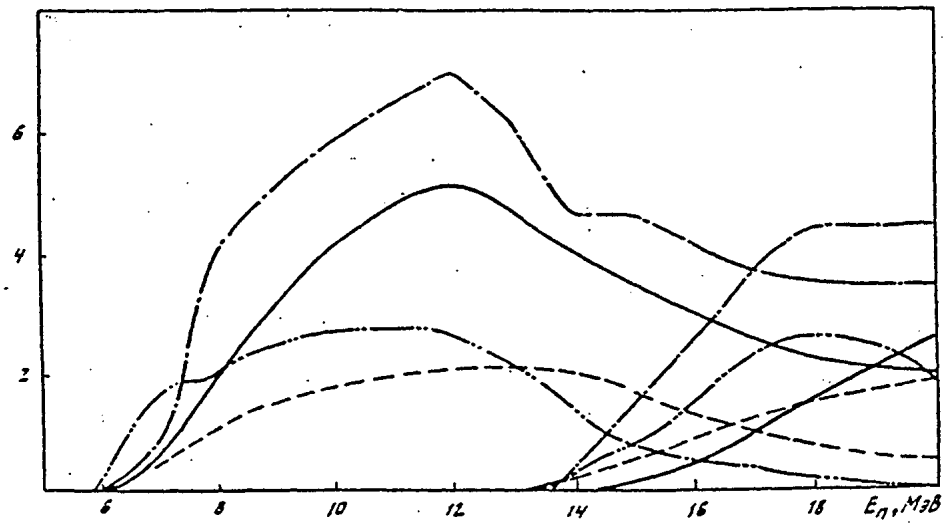

Figure 3. Comparison of evaluated (n,2n) and (n,3n) reactions cross-sections:
present evaluation ___
JENDL-2 evaluation -..-..-..-
JENDL-3 evaluation ------
ENDL-78 evaluation -.-.-.-.-.

These differences can undoubtedly result in different estimates of yields of proper isotopes for the fuel cycle, which is especially important for ADS with fast spectrum. Especially important for different estimates are fission and radioactive capture cross sections in resonance regions where their values can be extremely high. Nuclear data estimates in the regions are rather cumbersome. However using relevant systematic a self-consisted description of σ_t, σ_f, and σ_γ, have recently been obtained for ^{233}U in the energy range from 120 to 200 eV /G. Morogovsky, Fig. 4/.

Data of different libraries also differ in secondary neutron energy distributions for reactions (n, 2n), (n, n'), and (n, 3n), it is of special interest in the sense of spectrum formation for subcritical systems driven by high energy accelerators.

Large uncertainties in nuclear data required for thorium-uranium cycle and uncertainties in the region of intermediate energies are well known. In this regard for development of thorium-uranium cycle it is absolutely necessary to support any relevant experimental research and development of full modem evaluated nuclear data files. It should be also noted that the last changes made in such well known libraries like JENDL, JEF 2.2, ENDF/B-YI and other ones were made about five years ago and must be naturally expended to the region of higher energies. It is also necessary for estimates of performance of ADS.

3. EXPERIMENTAL RESEARCH

By now a lot of theoretical papers was published where basic aspects of ADS concept were discussed: production of energy, transmutation of radioactive waste, tritium production and incineration of weapon plutonium. The experimental research in this field is rather scarce because the experiments on available high energy accelerators are difficult and expensive, and in some cases even unfeasible. In this regard experimental research of various aspects of ADS on the basis of low energy ion accelerators are of great importance.

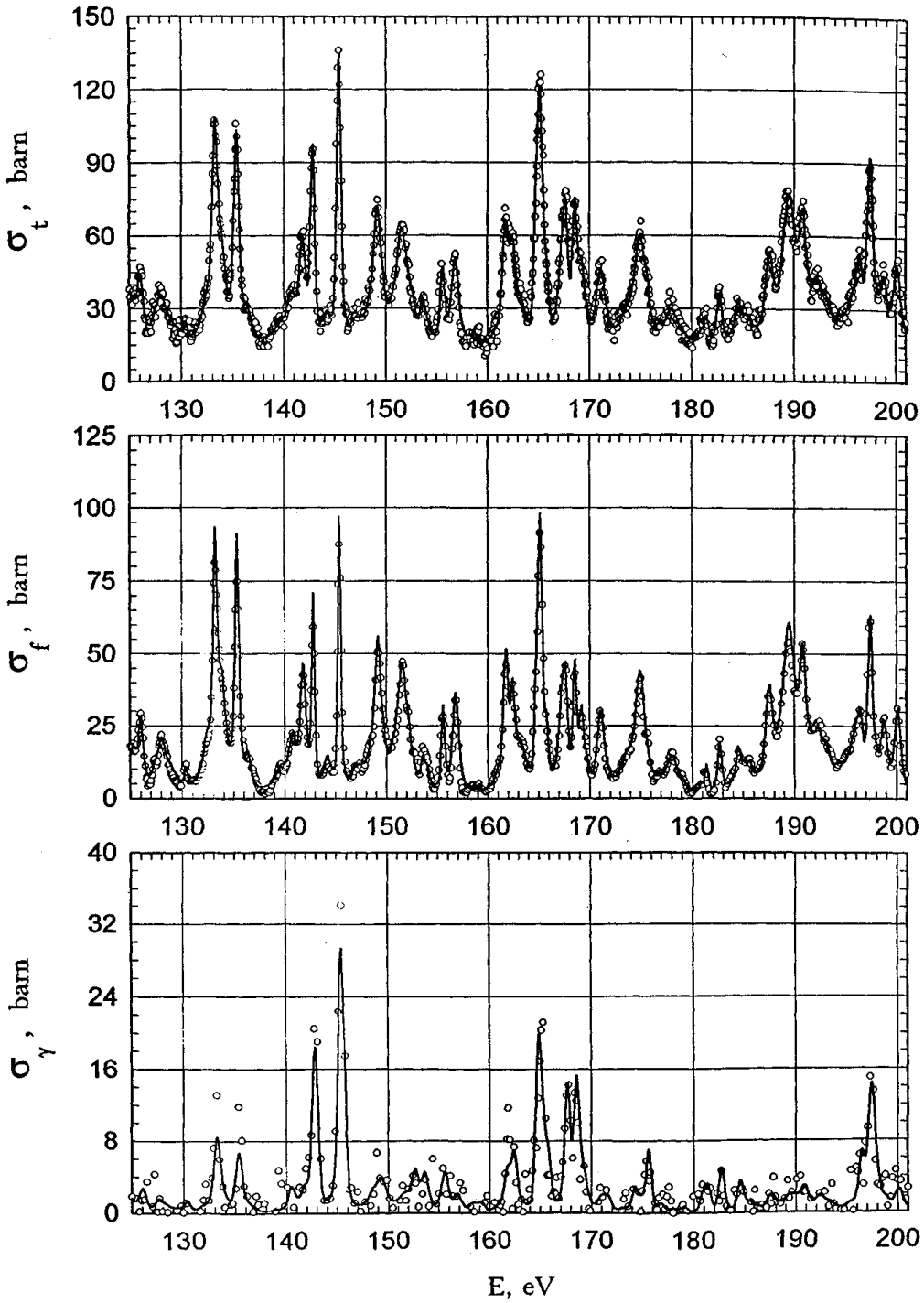

Figure 4. Comparison of experimental and calculated with MLBW parameters cross sections of ^{233}U.

The possibility of using low energy accelerators to investigate physical characteristics of subcritical target/blanket systems follows from the mechanism of nuclear reactions in high (≈1GeV) and low (≈15-20 MeV) energy ranges as well as from features of nucleon-mesons cascade development.

It was shown that the spallation neutron source can be simulated by neutrons escaping from heavy element targets bombarded by 14 MeV neutrons [8, 9]. It was a reason for creation of an experimental facility consisting of a subcritical target/blanket system driven by a high intensity (1.5-2.0 10^{12} neutrons/s) neutron generator (Fig. 5).

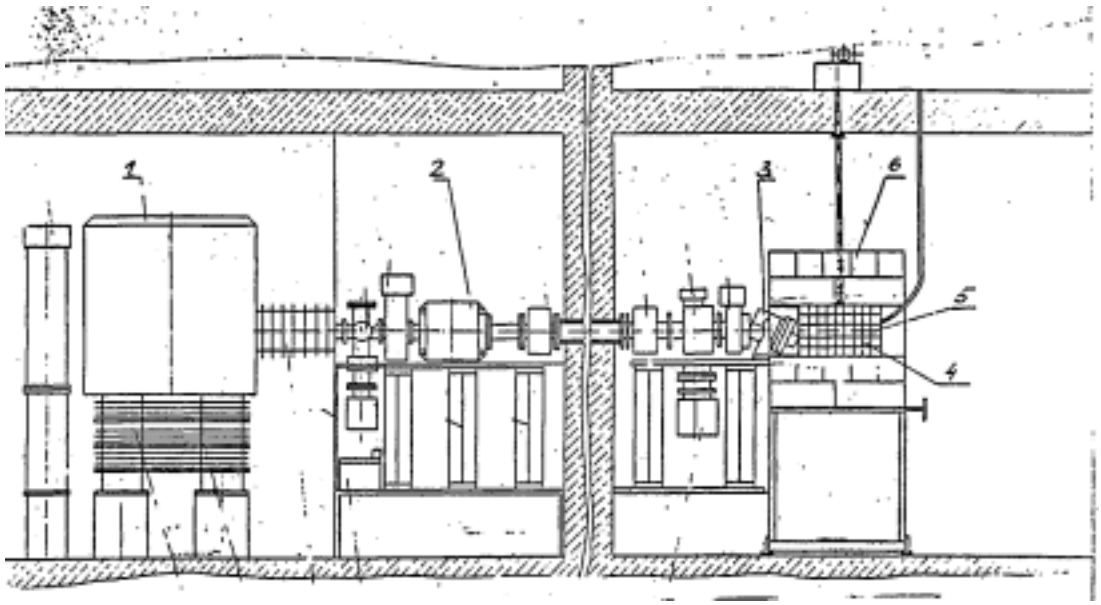

Figure 5. Subcritical uranium polyethylene assembly driven by neutron generator NG-12-1 (1-transformer, 2-magnetic analyzer, 3-target device, 4-core, 5-lead target, 6-graphite reflector).

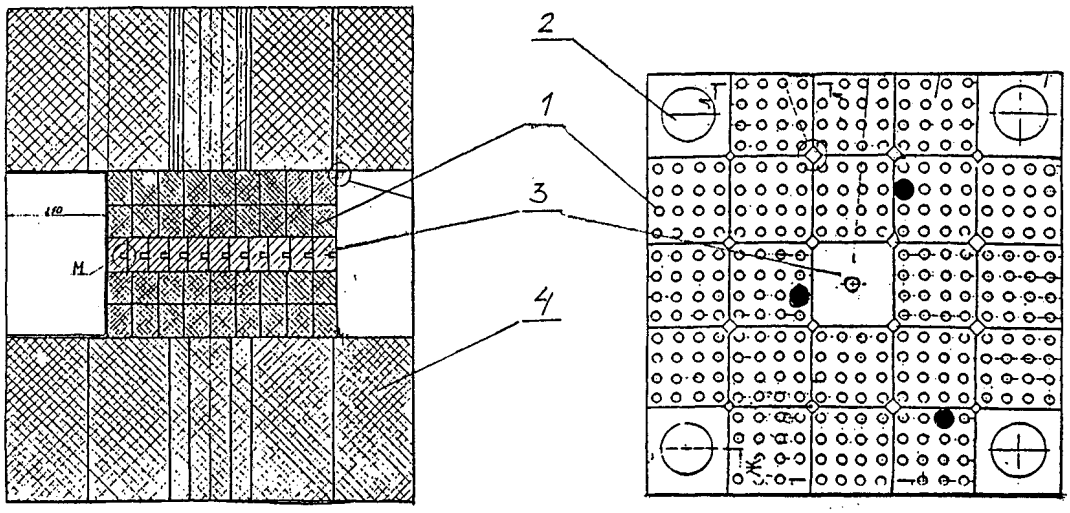

Fig. 6, Core of the subcritical assembly (1 - fuel subassembly, 2 - control subassembly, 3 - lead target, 4 - graphite reflector).

The target/blanket system includes a lead target for spallation neutron production and a subcritical assembly containing uranium rods with high enrichment, a moderator and experimental channels. Calculations have shown that it is possible to form different neutron spectra in experimental channels of the subcritical system: fast, resonance and thermal ones. In near future the subcritical assembly will be used for carrying out measurements in thermal and resonance spectra. The system measuring 4OOx4OOx600 mm^3 is assembled using cassettes with dimensions 8Ox8Ox600 mm^3 consisting of polyethylene moderator and fuel pins of UO_2 with enrichment equal to 10%. About 20 cassettes (i.e. 280 fuel pins) will be placed in the core of the target/blanket system to achieve the multiplication factor in the range of $0.9 < K_{eff} < 0.99$. The core is surrounded by a graphite reflector with dimensions 1000xl000x1200 mm^3, a cadmium layer I mm thick as well as borated polyethylene absorbing layer 0.5-1 mm thick (Fig. 6).

The subcritical assembly has three experimental channels located at a distance of 5.2, 10.5 and 16.6 cm from the assembly axis. The elements of the control system for neutron flux are placed in corners of the assembly.

It was shown by calculations that energy distributions of neutron flux density in the subcritical assembly differ from the spectra of thermal and fast reactors and are possibly characteristic of ADS with thermal spectrum.

The Fig. 7 presents the calculated neutron spectrum in the central part of the subcritical assembly. It is seen that neutrons with energies E_n<0.5 eV in the energy spectrum dominate and neutron flux varies slightly with energy in the range from 1 to 10^4 eV.

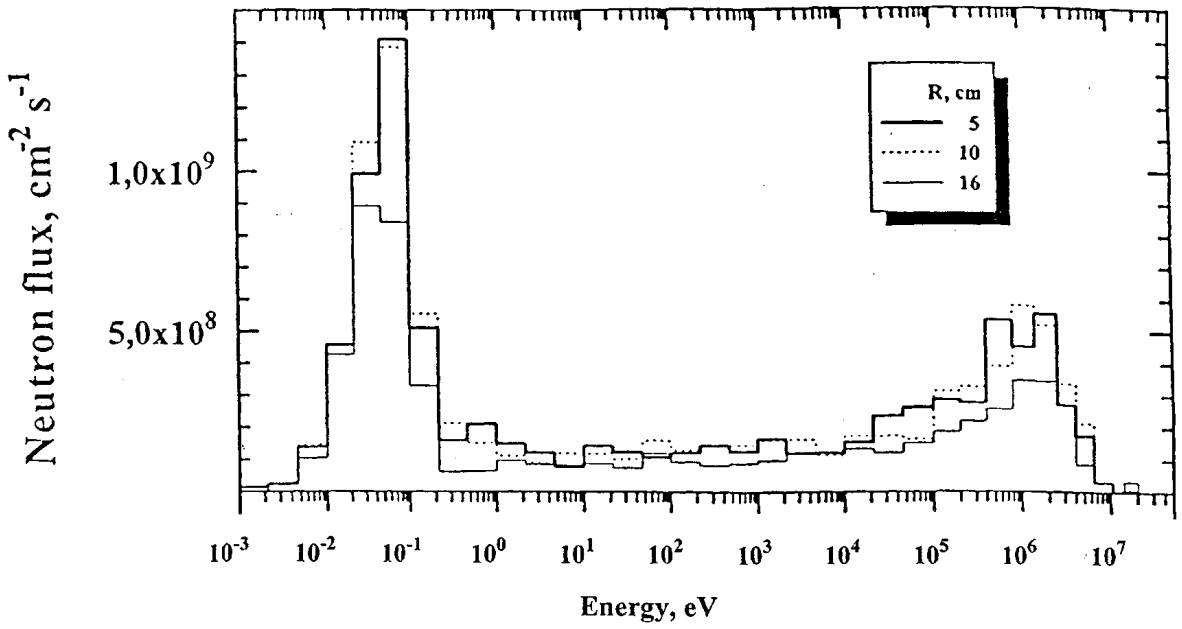

Figure 7. Calculated neutron energy distribution for the subcritical assembly driven by the neutron generator. Normalization was performed per 10^{12} neutrons per second.

Taking into account the neutron flux energy dependence as well as cross-sections σ_γ, and σ_{n2n}, for ^{232}Th and σ_f for ^{233}U one should expect that ^{232}U accumulation rate in ADS with thermal spectrum will be lower than that in thermal reactors.

Weak dependence of neutron flux density in the region of energies of 0.5 eV<E_n<10 keV gives the possibility to obtain experimental data on contribution of resonance region into transmutation rates of LLFP and MA where the values of cross-sections can be rather high.

We performed preliminary estimates for possibility of measuring transmutation rates of some LLFPs and MAs. The estimates revealed that in the assembly for a number of LLFPs and MAs reaction rates are high enough for the transmutation rates to be measured successfully (Table I).

Table I. Reaction rates of some LLFPs and MAs.
$\Phi = 10^{7}$ n/cm^2×s; t=10^4s; ε=0,1

No.	Target	σ , barn (therm)	m, g	N, n+A--A'	S_{imp}	Activity, A, Bk
1	Zr-93	2.6±1.4	1.0	1.7×10^{9}		9.3×10^{7}
2	Tc-99	20.0+1 22.9±1.3	1.0	1.2×10^{10}	1.4×10^{5}	6.3×10^{8}
3	Sn-126	0.297	0.1	1.4×10^{7}	2.7×10^{5}	1.1×10^{8}
4	1-129	27+2.2	1.0	1.3×10^{10}	8.6×10^{7}	6.6×10^{6}
5	Cs-135	8.710.5	1.0	3.9×10^{10}	1.2×10^{7}	4.3×10^{7}
6	Cs-137	0.11±0,033 0.25±0,02	0.001	4.8×10^{3}	63.0	3.2×10^{8}
7	Th-232	7.4	1.0		$3*10^{6}$	4.1×10^{3}
8	U-233	σ_f =522.6	1.0		3×10^{s}	3.6×10^{8}
9	Np-237	σ_c=169±3 σ_f=0.0019± 0.003	1.0	4.3×10^{10} 4.8×10^{6}	8.0×10^{6} 2.4×10^{2}#	2.6×10^{7}
10	Am-241	σ_C =832+20 σ_f ==3.15	0.001	2.1×10^{8} 7.9×10^{5}	1.1×10^{6} $2.5\times10^{2\#}$#	1.3×10^{8}
11	Am-243	σ_c =79.3±1.8 σ_f=0.2±0.11	0.1	2.0×10^{9} 5.0×10^{6}	1.6×10^{7} 2.5×10^{6}#	7.4×10^{s}

Yield of nuclide - 1%; the number of decayed nuclei during the time of the measurement is 10%; γ-quantum yield per one decay is 0.5;
NA - number of nuclei formed in (n,γ)-reactions;
Sj_{mp} - number of registered impulses.

From the data presented in the table one can see that if neutron flux at thermal point equals approximately to 10^7 n/(cm^{2}s) it is possible to measure reaction rates in the spectrum of the subcritical assembly driven by the neutron generator. When performing activation measurements with samples irradiated in cadmium containers one can obtain information on average cross sections for ^{232}Th and ^{233}U in the energy range with little variation of neutron flux versus energy. It is very important for updating of evaluated nuclear data libraries.

The measurements of energy spectra at different points inside experimental channels will be performed by means of activation technique having different advantages comparing to other ones. In the energy range 30keV-15MeV the measurements will be performed by means of solid-state nuclear track detectors and thin-film break-down counters.

It will allow define optimal conditions for transmutation and get information on average cross sections for the energy spectra (like resonance integrals for reactor systems) which can be characteristic ones for energy systems driven by accelerators. In addition it is possible to measure the spectral indices σ_i/σ_f [5, 6, 7] for different isotopes.

The experiments on measurement of transmutation rates of LLFP and MA in different neutron will allow to make conclusions about trends of subsequent investigations, estimate

discrepancies in evaluated nuclear data files for fission products and minor actinides as well as to compare results obtained by means of computer codes with experimental data. It is also possible to carry out experiments for research of peculiarities of dynamics of target/blanket systems driven by high energy accelerators.

The work on updating of existing evaluated nuclear data libraries and experimental research in the ADS region will be carried out at the subcritical assembly driven by the neutron generator in near future. The work will be supported by National Academy of Sciences of Belarus as well as ISTC under Project B-070

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STUDY OF THORIUM FUEL CYCLES BURNING PLUTONIUM IN THE MODULE- HTR*

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Abstract. The advantages of HTR module pebble bed reactor and thorium fuel cycles are discussed in this paper. In order to reduce plutonium stockpiles, the thorium fuel cycles are used for HTR module, and plutonium is used as fissile material. The equilibrium core is calculated and analysed for the case of the different heavy metal loading and enrichment. For the case of more than 11 g heavy metal per sphere has a negative temperature coefficient, and the maximum temperature of fuel elements under regular operation and loss of coolant accident is lower than 1500ºC. Therefore the feasibility of above scheme is studied.

1. INTRODUCTION

Since development work on the 200 MW-MODULE Pebble Bed reactor began in 1979 in Germany, the HTR module is considered as one kind of the advanced nuclear reactors with completely passive safety properties [1]. Under any accident the release of radioactivity in HTR module is not possible even without technical safety equipment in operation. The coating of the coated particle embedded in the fuel elements does not permit any radioactive gas or metallic fission products to escape from intact fuel particles up to a temperature of 1600ºC.

Residual heat can be removed from the core, even under extreme accident conditions, by means of passive heat transfer processes based on natural laws, such as heat conduction and radiation. The HTR Module also has a negative reactivity temperature coefficient. Therefore a core temperature rise can offset any reactivity increase as a result of reactivity accidents.

The spherical fuel elements are used for the HTR Module. Because these fuel elements are able to receive a very great variety of fuel cycles, this fuel permits a wide flexibility in the design of the reactor. A thorium-based fuel cycle in the HTR Module would produce a small amount of toxic fuel waste or long-lived radiotoxic waste. In order to reduce plutonium stockpiles, plutonium is used for thorium fuel cycle as fissile material in HTR Module.

2. HTR MODULE AND CALCULATION

The layout of HTR Module is shown in Fig. 1. Main design parameters are listed in Table I. The power density is 3MW/m^3 and reactor dimensions have been optimized to provide sufficiently high passive removal of the decay heat under loss of coolant, thus keeping the fuel temperature below 1600ºC. The weight of heavy metal and enrichment in the sphere are optimized for burning as much plutonium as possible, and keeping a negative temperature coefficient. Under normal operation the temperature for the spectrum calculation is listed Table II.

* 1998 meeting.

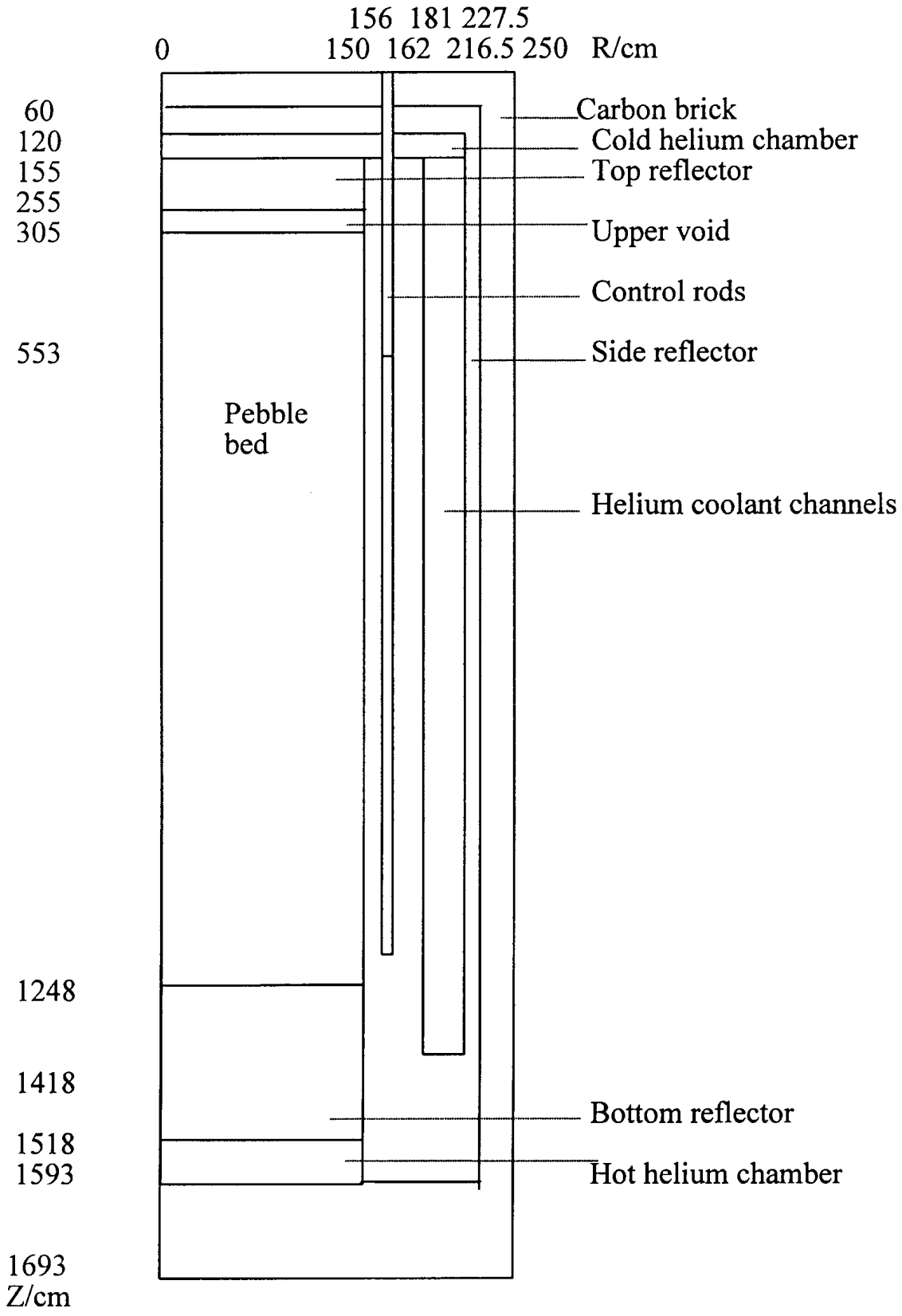

Figure1. The layout of HTR module.

Table I. Main design data of the HTR module

Reactor-Core	
Thermal power MW	200
Power density MW/m^3	3
Core height/diameter m	9.43/3.0
Heating of helium ºC	200→700
Helium pressure bar	60
Helium mass flow rate kg/s	85.4
Fuel element	
Diameter of pebble cm	6
Diameter of fuel zone cm	5
Density of graphite in the matrix and outer shell g/cm^3	1.75
Volumetric filling fraction of elements	0.61
Number of passes of spheres through the core	10
Coated particle	
Radius of the kernel cm	0.025
Fuel composition	PuO_2-ThO_2
Density of the kernel g/cm^3	10.5
Isotopic composition of plutonium %	Pu-239/Pu-240=94/6
Coating layers	C/C/SIC/C
Density g/cm^3	1.05/1.90/3.18/1.90
Thickness cm	0.009/0.004/0.0035/0.0035

Table II Temperature for spectrum calculation

Zone(cm)	Temperature(ºC)
0<R<150 305<Z<1428	Fuel:586 moderator:576
0<R<255 0<Z<255	203
0<R<150 255<Z<305	265
150<R<162 155<Z<1248	430
162<R<250 155<Z<1248	250
0<R<150 1248<Z<1518	748
0<R<150 1518<Z<1693	320
150<R<250 1248<Z<1693	320

The VSOP code [2] is used for calculation of HTR Module. The reactor is divided into eleven spectrum zones. The pebble bed is divided into five spectrum zones. The thorium absorption cross sections of resolved and unresolved resonances are generated by ZUT-DGL code basing on resonance data.

3. Calculation results

The equilibrium core is calculated for the case of the different heavy metal loadings and enrichments. The main results for a heavy metal loading of 7g/sphere are given in the Table III.

Table III. Main data of the equilibrium core under 7g(HM)/sphere

Enrichment(%)	8 9.6 11
K_{eff}	0.9430 1.0073 1.0402
Burnup MW·d/t_{HM}	97325 100925 105595
Fuel element residence time days	1258 1258 1258
Conversion ratio	0.575 0.518 0.493
Inventory of ^{233}U (kg/GW(th))	151.5 138.74 131.26
Inventory of ^{239}Pu (kg/GW(th))	97.65 214.79 321.54
Inventory of ^{232}Th (kg/GW(th))	11249.92 11047.86 10896.41
Supply of ^{239}Pu (kg/GW(th))	0.8271 0.9875 1.1322
Discharge of ^{239}Pu (kg/GW(th))	0.0002 0.0035 0.0132
Consumption of ^{239}Pu (kg/GW(th))	0.8269 0.9840 1.1190
Utilization of the loaded ^{239}Pu %	99.98 99.65 98.83

Table IV. Main performance for different heavy metal loading

Heavy metal loading	g/sphere	7	9	11	13
Enrichment	%	9.6	10	11	12
Burnup	MW·d/t_{HM}	100925	101828	102628	103783
Fuel element residence time	days	1258	1618	1977	2336
Conversion ratio		0.518	0.535	0.549	0.569
Power peaking max./avg.		4.44	3.54	2.86	2.61
Max. power per ball	kW /ball	2.47	1.97	1.59	1.45
Core leakage	%	10.04	8.97	8.21	7.84
Neutron flux	E+14/(cm^2×s)				
Avg. thermal flux	(<1.86ev)	0.4176	0.2745	0.1752	0.1277
Avg. fast flux	(>0.1Mev)	0.2402	0.2376	0.2357	0.2373
Avg. total flux		1.2060	1.0479	0.9345	0.8843
Temperature coefficient	(Δk/k/)				
Fuel	(10^{-5})	-1.59	-2.02	-2.35	-2.70
Moderator	(10^{-5})	3.68	0.883	-1.49	-2.48
Reflector	(10^{-6})	2.82	2.34	2.21	1.89

The equilibrium status is calculated for enrichment 8%, 9.6% and 11% respectively. With the enrichment increment, K_{eff} value increases, conversion ratio decreases, consumption of ^{239}Pu increases and utilization ratio of the loaded ^{239}Pu decreases.

The pebble bed reactor operates by continuative loading and discharging of fuel elements. Therefore little excess reactivity is required in normal operation. The case of enrichment 9.6% is also able to operate.

In order that the reactor has negative moderator temperature coefficient, heavy metal loading in sphere is increased gradually and enrichment is adjusted so that K_{eff} value of core is around 1.01. The results for heavy metal weight in sphere 7,9,11,13 g are given in Table IV and Table V.

Table IV shows that as heavy metal weight in sphere increases the enrichment should increase for retaining a close K_{eff} value, the conversion ratio increases, moderator temperature coefficient changes from positive to negative.

Table V. Inventory, supply and discharge of main isotope

Heavy metal loading g/ball	7	9	11	13
Inventory (kg/GW(th))				
U-233	138.74	194.5	254.82	322.48
Pu-239	214.79	359.60	664.28	1031.45
Pu-241	100.54	156.24	241.75	326.27
Th-232	11047.86	14115.14	17027.33	19835.31
Pu-240	133.19	161.14	203.33	243.55
Pu-242	48.29	56.18	56.32	57.81
Supply-discharge (kg/GWd(th))				
U-233	0-0.1903	0-0.2075	0-0.2269	0-0.2409
Pu-239	0.9875-0.0035	1.0279-0.0103	1.1300-0.0408	1.2315-0.0861
Pu-241	0-0.0400	0-0.0580	0-0.1015	0-0.1364
Th-232	8.9653-8.6071	8.9164-8.5441	8.8043-8.4241	8.6896-8.2976
Pu-240	0.0633-0.0219	0.0659-0.0219	0.0724-0.0319	0.0789-0.0399
Pu-242	0-0.0684	0-0.0646	0-0.0586	0-0.0532
Consumption of Pu-239 (kg/GWd(th))	0.9840	1.0176	1.0892	1.1454

Table V shows that with increment of heavy metal loading per sphere weight of U-233 and ^{239}Pu in equilibrium code increases, weight of ^{233}U and ^{239}Pu in discharged spheres increases, consumption of ^{239}Pu increases.

The thermal hydraulics evaluation in the case of a heavy metal loading per sphere of 13 g and an enrichment in the fresh element of 12%, is performed. The temperature distribution of the regular operation is calculated. The temperature of spectrum zones is listed Table VI. The temperature value in Table VI is close to that in Table II. The maximum temperature of fuel elements is 779ºC. For the Loss of Coolant Accident (LOCA) residual heat can be removed from the core to surrounding graphite structures and then to the surface coolers though natural process. The variance of the relative decay power (relative to full power) is outlined in Fig. 2. The temperature transients of the core and pressure vessel are indicated in Fig. 3. The maximum temperature of core reaches at maximum value 1448ºC at about 106 hours after shutdown.

Therefore a thorium fuel cycle, burning plutonium, in the HTR module is entirely feasible.

Table VI. Temperature of spectrum zones

Zone(cm)	Temperature(ºC)
0<R<150 305<Z<493.6	Fuel:340.90 moderator:331.21
0<R<150 493.6<Z<682.2	Fuel:456.41 moderator:442.44
0<R<150 688.2<Z<870.8	Fuel:569.58 moderator:554.95
0<R<150 870.8<Z<1059.4	Fuel:658.30 moderator:645.94
0<R<150 1059.4<Z<1248	Fuel:711.62 moderator:703.37
0<R<255 0<Z<255	192.28
0<R<150 255<Z<305	264.71
150<R<162 155<Z<1248	427.62
162<R<250 155<Z<1248	260.30
0<R<150 1248<Z<1518	696.24
0<R<150 1518<Z<1693	342.28
150<R<250 1248<Z<1693	342.28

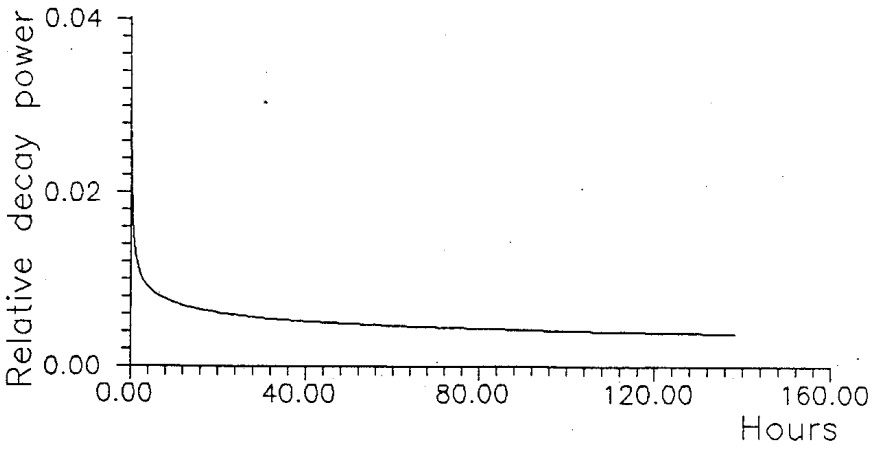

Figure 2. Variance of relative decay power.

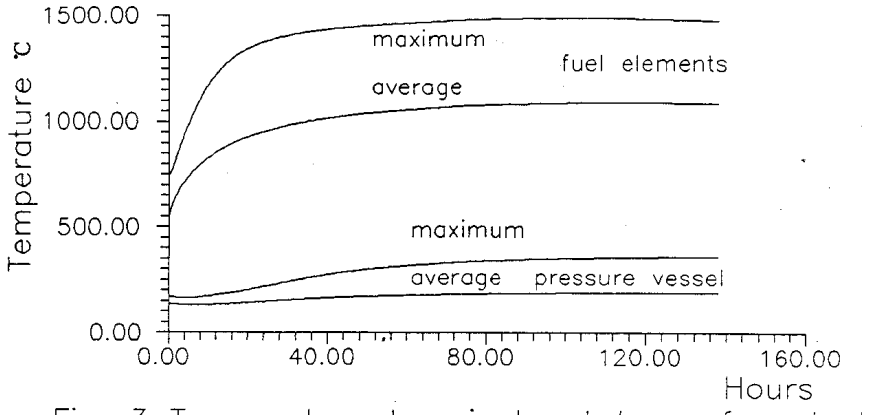

Figure 3. Temperature transients at loss of coolant.

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COMPLEX REACTOR PHYSICS INVESTIGATIONS OF THORIUM CONTAINING LIQUID FUEL*

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Abstract. The great progress in the development of new accelerators has substantially contributed to a new possibility of a practical solution of nuclear waste issue on the basis of transmutation. For an adoption of the given region of problems and a start of an own contribution to its solution in specific conditions of Czech nuclear power program, the first stage of investigation was performed in the Nuclear Research Institute Řež plc., the Nuclear Physics Institute of the Academy of Sciences of the Czech Republic and Research and Development Basis of the ŠKODA, Nuclear Machinery Ltd. with an assistance of the Czech Power Generating Board during the years 1994 -1996. Two special conferences were organized by their initiative in June 1994 and September 1995 which made an inventory of abilities and interests of Czech scientific, research and industrial institutions to take share in the solution of this problem. The construction of a nuclear power plant - transmuter, where the possibility to reach critical state is excluded and, simultaneously, the continuous separation of short-lived as well as of long-lived isotopes (including actinides) from the primary fuel-coolant circuit is made possible, is the goal. The key part of such a system will be, besides an accelerator and devices of a mechanical and chemical reprocessing, a subcritical reactor system containing a variety of fuel materials among them even Th containing components on which we have been focusing our work from the point of view of new proliferation resistant technologies. In the framework of the Czech national project of a transmuter called LA-0, there is a close collaboration assumed with a series of leading foreign laboratories with whom a very fruitful contacts have already been established.

1. THE CONCEPT OF LIQUID FUEL

1. Introduction

There are principle drawbacks of any kind of solid nuclear fuel listed and analyzed in the first part of this chapter. One of the primary results of the analyses performed shows that the solid fuel concept, which was to certain degree advantageous in the first periods of a nuclear reactor development and operation, has guided this branch of a utilization of atomic nucleus energy to a death end (not having been able to solve principle problems of the corresponding fuel cycle in an acceptable way). On the basis of this, the liquid fuel concept and its benefits are introduced and briefly described in the following part of the chapter.

As one of the first realistic attempts to utilize the advantages of liquid fuel, the reactor/blanket system with molten fluoride salts in the role of fuel and coolant simultaneously, as incorporated in the accelerator-driven transmutation technology (ADTT) being proposed in [1], will be studied both theoretically and experimentally. There is a preliminary design concept of an experimental assembly LA-0 briefly introduced in the following paragraph which is under preparation in the Czech Republic for such a project.

Finally, there will be another very promising concept [4,5] of a small low power ADTT system introduced which is characterized by a high level of safety and economical efficiency. This subcritical system with liquid fuel driven by a linear electron accelerator represents an additional element -nuclear incinerator- to the nuclear power complex (based upon thermal and fast critical power reactors) making the whole complex acceptable and simultaneously giving an alternative also very highly acceptable nuclear source of energy and even other

* 1998 meeting.

products (e.g. radionuclides, etc.). In the conclusion, the overall survey of principal benefits which may be expected by introducing liquid nuclear fuel in nuclear power and research reactor systems is given and critically analyzed. The other comparably important principles (e.g. the general subcriticality of reactor systems principle) are mentioned which being applied in the nearest future may form a basis for an absolutely new nuclear reactor concept and a new nuclear power era at all.

In spite of the fact that all what is following is well known it seems to be worth to remind it in the new circumstances of nuclear power at the end of the 20^{th} century while starting to search new nuclear energy systems and fuel cycle options for the 21^{st} century. Since the discovery of the reaction of atomic nucleus fission, the main goal of all efforts was to utilize it for an energy generation. As one of the most important conditions for an efficient achievement of this goal self-sustaining of fission chain reaction was demanded in an assembly containing fissionable nuclei of nuclear fuel without an external source of neutrons. If this was reached, the assembly was defined as being critical. Let us note that it was by definition (theoretically) critical on prompt neutrons released, immediately, from fission reactions only. Very early, it was observed experimentally that the assembly reaching criticality is in fact very slightly subcritical on prompt neutrons and that there is a not very strong natural source of delayed neutrons originated from radioactive decay of some of the fission products always added (which, fortunately, allowed easier control of the system).

At the early stages, reaching criticality was one of the most difficult tasks and all the effort and ideas had been devoted to this aim. The reason was that there were only small amounts of fissionable materials available in those times in the form of the low (0.7%) content of ^{235}U in natural uranium. Therefore, solid phase metallic uranium with highest as possible density was used and in the form of blocks with a specifically defined size arranged in a heterogeneous lattice filled in by a solid (graphite) or liquid (heavy water) moderator with a certain pitch determined by optimal neutronic conditions. This arrangement has remained nearly exclusive one being used even in latter systems with fuels enriched by ^{235}U content up to much higher levels than the content of natural composition of uranium. The reasons had been of different nature, however, the designs have mostly started from what became already an approved conventional principle - solid fuel blocks in a heterogeneous lattice - which has been kept even in the case of pure or high enriched fuel in a fast neutron system without moderator.

One of the next consequences of the adoption of the solid fuel concept has been a type of control system which has been mostly applied for a short term control of nuclear reactors - the concept of solid absorbers - and what is more the concept of a negative neutron source (neutron poison) at all. This, and a number of other consequences, can be traced to start all from the initial tension in neutron economy when the principle of a self-sustaining fission chain reaction and consequently the concept of a critical reactor have been adopted. They all begin to form a magic circle of convention in which the short term and finally even long term operational behavior of nuclear, namely power, reactors is being imprisoned and limited in its ability to give a positive and broadly acceptable development. Let us explain this thesis in some following more see-through examples.

The adoption of the solid fuel concept leads to the principal necessity to keep the fuel blocks at a certain position in the reactor core for a shorter or longer period of time. This in-core residential time is especially long in power systems where at least a quasi-continuous exchange of fuel would be very complicated and expensive. Therefore, the following very inconvenient consequence arises: the whole time, the block of solid fuel remains at a certain position in the reactor core, there are fission fragments and by neutron capture induced

radionuclides (let us call them altogether products) being accumulated in the volume of the fuel block. There are several secondary consequences caused by this fact which contribute to the above mentioned magic circle forming:

1) reactivity margin for a short term as well as long term negative influence of the increasingly accumulated products has to be applied which has to be compensated by another artificial negative source of neutrons. It has in principle a consequence in greater amount of fuel being present in the core than really necessary for a demanded power and then the more products including actinides is generated.
2) the original fuel is finally so heavily poisoned by the products that it cannot keep the self-sustained fission chain reaction any more and a further operation of the reactor under original conditions is impossible. There is a principle change in the operation and structure of the reactor unavoidable which means an outage and exchange of at least a part of the original fuel charge.
3) the most controversial problem what to do with spent solid fuel arises and a vicious circle has been closed or a solid fuel concept "trap" snapped.

The above briefly described solid fuel concept shows its most important and sensitive drawbacks:

1) continuous accumulation of products during the whole residential time of fuel blocks in the core,
2) following necessity to stop the operation, discharge spent fuel and store it for a necessary period of time (in order of magnitude of years until it reaches a desirably low level of radioactivity) in a specific storage,
3) the last and the most difficult drawback is the need of an optimal decision of the following destiny of spent fuel.

Up to now, the only two possible solutions were developed either to reprocess (chemically) it and to prepare next generation of solid fuel (it means with basically the same class of drawbacks) or to dispose it in a depository of a corresponding quality (which sometimes is called repository because a possible reuse of the disposed product is supposed). In the former case mostly chemical methods and processes are applied. In the latter, a lot of branches is involved, however, nearly all of them are of a classical (non-nuclear) nature. The only nuclear process which is employed is the natural radioactive decay.

This fact contains one very controversial principle or better say a violence of a basic principle which can be described as follows: The energy generation in nuclear reactors utilizes enforced nuclear process which are simultaneously producing products or nuclear waste (including secondary raw materials e.g. actinides). The treatment of the products needs to apply an adequate technology in an adequate scale. This principle has not been applied and fulfilled in those so far developed and designed systems for spent solid fuel management. There is an adequate technology which only one can utilize nuclear processes and which can transfer the high level and long-lived radionuclides towards short-lived or even stable nuclides-transmutation technology performed in a suitable nuclear reactor device and combined with a continuous separation of certain components of its core or reprocessing of the reactor fuel as to avoid the consequent induction of radioactivity by neutron irradiation of stable and short-lived nuclides. One of the principle concepts allowing to reach such a technology in an industrial scale is the concept of liquid nuclear fuel.

2. LIQUID FUEL CONCEPT FOR NEUTRON SOURCE-DRIVEN TRANSMUTATION TECHNOLOGY (NSDTT)

2.1. Molten fluoride salt fuel for neutron source-driven transmutation technology

The concept of a neutron source - driven subcritical blanket for a nuclear incineration of nuclear waste is well known for a several recent years [1]. Let us recall at least very briefly the main features of the last developed version of this concept and let us show a part of a proposed research program to approve its ability for an efficient realization in the industrial scale.

The fuel material is in the form of the fluoride salt AcF_4 dissolved in a molten salt carrier whose composition is a mixture of ^7LiF and $^9BeF_2$. The carrier's melting point and operating temperature are about 500^{o}C and 650^{o}C, respectively. The molten salt flows over either the outside of a close-packed set of cylindrical high-purity graphite blocks or inside cylindrical channels coaxially situated in e.g. hexagonal graphite blocks.

There has been an experimental research system designed by the author preconceptually in [3] which should be developed and realized in the Nuclear Research Institute Řež plc in the Czech Republic. The final purpose of the system would be an experimental testing of a given type of transmuter reactor/blanket core neutronics and possibly also other physical and technological characteristics and properties including time behavior. For the very first stage, the following scheme can be applied which will allow to reach the first results very cheaply and relatively soon. There can be an elementary, however, a sufficiently representative sample of the investigated reactor blanket lattice inserted into an existing experimental reactor core serving like a driver and the basic set of its characteristics can be experimentally measured and verified. The suitable experimental reactor can be e.g. the experimental reactors LR-0 (full-scale core modeling in Nuclear Research Institute Řež or VR-1 (training reactor at Czech Technical University Praha) which have been successfully operated for core analyses of thermal reactors since 1982 and 1990, respectively.

2.2. Low power ADTT system

The molten salt reactors (MSRs) with the continuous control of nuclide composition almost do not require an initial reactivity margin. In such reactors, subcriticality may be reduced up to the minimum value ß where ß is the effective delayed neutron fraction. However, with such a small subcriticality and in view of available uncertainties in nuclear data and nuclide concentrations, the difference between subcritical and critical MSR in a great extent disappears: in both cases the nuclear safety is ensured by the large negative temperature reactivity effect. The deeper subcriticality is of course substantiated by the fact that under such conditions we exclude the necessity to control a reactor - burner in a dynamic mode, that is a bit difficult and poorly known.

In this case, the e.g. accelerator - driven positive source performs only one of the usual functions - the function of a reactor control system without inertia, an alternative to, up to now usually used as reactor control organs, negative sources like e.g. absorbers or decreasing of the dimensions of the system, etc. The high level parameter proton accelerator with its all disadvantages (like e.g. the length ~ 1 km, the investments ~ U$ 1 billion, etc.) having been applied e.g. in the Los Alamos concept is not necessary more in the system and a low level parameter accelerator can be employed.

3. CHEMICAL PROBLEMS OF SPENT NUCLEAR FUEL TRANSMUTATION

3.1. Introduction

In the beginning of the 90s, there has been a transmutation process [6-10] proposed in the Los Alamos National Laboratory which enables burning of actinides contained in spent nuclear fuel and their decomposition to fission products in a subcritical reactor blanket driven by an external neutron source (to be sufficiently strong the spallation reaction initiated by a beam of highly accelerated charged particles e.g. protons by a high power linear accelerator being operated in LANL was suggested as the external source of neutrons). Thus, it would make possible to transform spent fuel from commercial reactors as well as from military production (including plutonium warheads) to short-lived fission products.

The principle of the chemical treatment applied in those projects has been based on the experience obtained in the sixties during the operation of nuclear reactors with molten fluorides in the ORNL [11-16]. Two types of reactors have been operated there: Molten Salt Reactor (MSR) and Molten Salt Breeder Reactor (MSBR). Both reactors have originally been designed for the thorium fuel cycle, where fissionable ^{233}U is formed from ^{232}Th. However, during the experimental verification ^{235}U and ^{239}Pu have also been used.

There was a mixture of BeF_2, LiF and ThF_4 used as a carrier for both the fissionable and breeding components. Molten salt has been circulated by a pump from the graphite-moderated reactor via a heat exchanger back to the reactor. An original process for continuous removal of the melt was worked-out. The melt was processed in order to obtain ^{233}U produced and to separate fission products. The purified molten salt together with ^{233}U obtained were returned to the reactor.

The chain fission reaction was initiated with ^{235}U, the use of ^{239}Pu has also been experimentally verified. Circulating a melt composed of LiF, BeF_2, ThF_4, UF_4 or PuF_3 has served as a basic load of the reactor where the fission reaction proceeded. At the same time, it served also as a heat-exchanging medium. The temperature of the salt in the reactor was 500 - 700^oC. In the heat exchanger, the molten salt was cooled down from 700^oC to 550 - 500^oC giving heat to the secondary circuit. A mixture of molten $NaBF_4$ and NaF was selected as a coolant circulating in the cooling circuit where the coolant is not exposed to radiation and neutron flux and, therefore, cheaper material of a lower melting point could be used.

Metallic parts of the reactor and of the equipment for the salt treatment being in contact with molten salts were produced from a hastelloy N type material. Its main component is nickel containing approximately 16% Mo, 7% Cr, 4% Fe and 0,05% C. This material proved fully satisfactory and did not show corrosion or radiation damage during three years of operation.

3.2. Chemical processes taking place during the isolation of uranium, palladium and fission products from the MSR and MSBR type reactors [16-20]

The operation of a reactor with the fluoride molten salt needs a continuous removal of the melt. The aim is to reprocess the melt, i.e., to obtain uranium and protactinium and to separate fission products.

For the primary separation of uranium elementary fluorine is used passing through the molten salt and escaping in the form of UF_6 together with some volatile fluorides of fission products, such as MoF_6 or TcF_6.

Metallic bismuth having a low melting point of 271°C is used for extraction and reducing extraction. It is immiscible with basic components of the molten halide mixtures containing fluorides, chlorides and bromides and has a negligible vapor tension in the range of the temperatures used. Further, bismuth dissolves some metals such as lithium, thorium, uranium, protactinium and rare earth elements. Dissolved lithium is able (under the given temperatures) to reduce fluorides to a metal according to the general equation $MF + nLi(Bi) = M(Bi) + LiF$.

In addition, is has been found that after the reduction of rare earths it is possible to extract them selectively from bismuth to LiCl or LiBr.

Molten salt composed of LiF 72%, BeF_2 16% and ThF_4 12% contains (at a continuous removal of 0.3 mole % of UF_4) approximately 0.0035 mole % of PaF_4. About 99% of uranium are separated from the salt by fluorination. Remaining uranium and protactinium in contact with liquid bismuth and dissolved lithium (when the salt is passing through the counter-current extractor) are going to the metal. Metals dissolved in bismuth are converted to nonvolatile fluorides by passing-through hydrogen fluoride. The fluorides formed are mechanically segregated. Technological scheme has been verified at a laboratory scale and recommended as a part of the 1000 MW(e) reactor.

3.3. Chemical processes connected with reprocessing of spent fuel in ATW systems

General scheme of the whole process supposed for the basic types of the processed material is given in Figure 1 The fuel is adjusted before the processing, i.e., all metallic parts of fuel elements and packings of plutonium warheads are separated. Fuel elements with Zr and Nb coating are dissolved in BeF_2 + LiF melt under a continuous bubbling-through of hydrogen fluoride. Hydrogen is released during the process getting off with the volatile components of the fuel, predominantly with Xe and Kr. Before entering the reactor, the melt containing dissolved fuel element undergoes electrolysis in order to separate some metallic components such as zirconium, uranium and some fission products. The melt is then pumped into the reactor where it surrounds the neutron source inducing the nuclear reaction desired.

After a certain reaction time, the melt is continuously taken off in order to separate fission products and remaining actinides by the method of reduction extraction with liquid bismuth and probably also by the centrifugation method. The basic fluoride melt (carrier medium) is purified, its composition is adjusted and then returned to the process.

At present, the chemical problems of the molten salt medium reactors are thoroughly studied in USA at the Oak Ridge and Los Alamos National Laboratories, in Russia at the Kurchatov Institute of Atomic Energy, Moscow and the Research Institute of Atomic Reactors, Dimitrovgrad, in Japan at JAERI, Tokai Mura and at several universities (there is a complementary Japanese programme [22-26] called "Fuji" which is directed mainly to the application of thorium cycle), in France at CEA, at the Belorussian Academy of Sciences in Minsk, at the Royal Institute of Technology, Stockholm, Sweden, and at the Nuclear Research Institute Řež in the Czech Republic.

The Department of Fluorine chemistry of the Nuclear Research Institute Řež has been engaged in the field of inorganic fluorides for more than 30 years. Experience thus obtained was used in nuclear chemistry, especially to the separation of a series of fluorides of uranium, plutonium, fission products and some transplutonium elements [27-30].

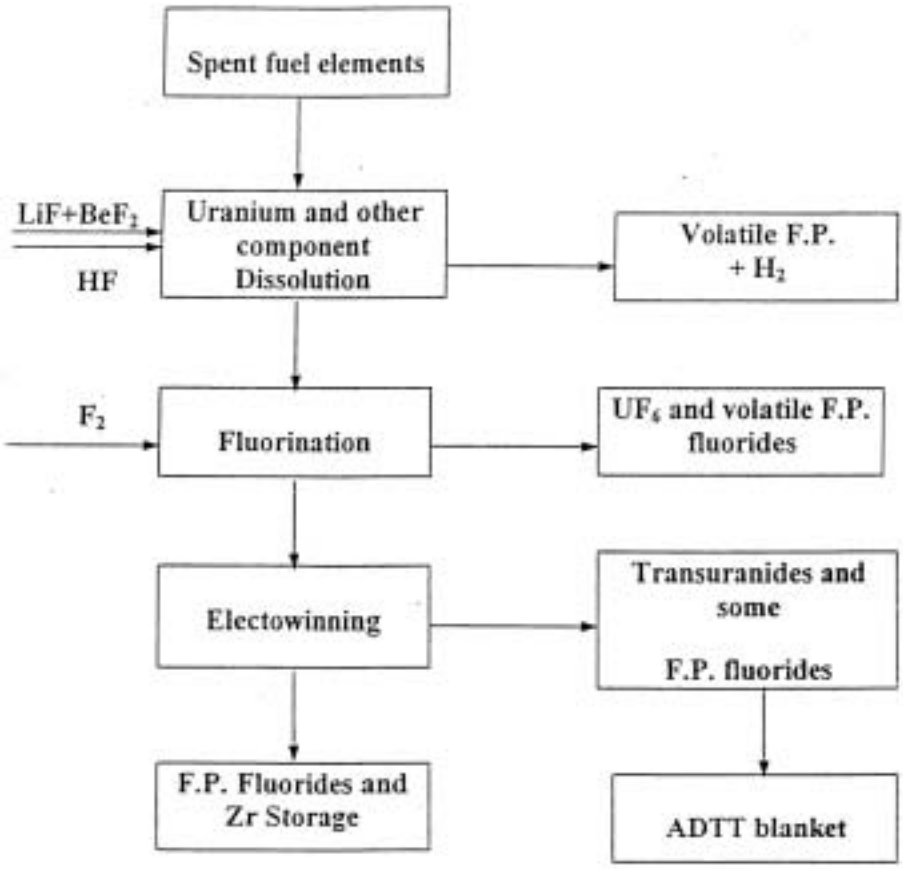


Figure 1. The scheme of an ADTT simplified chemical engineering process

3.4 Experience in fluoride volatilization and chemical problems associated with molten salt technology application in ADTT

A technological process has been worked-out for the separation of uranium and plutonium from the spent fuel by the so-called fluoride method. The whole process was upgraded to a pilot plant scale with a capacity of 1-3 kg of processed fuel/hour. There was a part of the technological equipment built and verified at the inactive scale at the Nuclear Research Institute, Řež. The whole technological process was then realized in the Institute of Atomic Reactors at Dimitrovgrad. All equipment including fittings, measuring instruments and accessories have been built in the former Czechoslovakia, the plutonium part of the pilot plant has been built in the former USSR.

A certain experience has also been obtained on the uranium isotopes separation by ultra-centrifugation. Solutions of UF_6 in perfluoroorganic compounds have been treated. The separation effect was determined by mass-spectrometry.

The experience gained in the course of the research is going to be applied in developing fluoride chemistry based separation processes for the use in the Accelerator Driven Transmutation Technology (ADTT).

3.4.1. Spent fuel reprocessing

The process consists in fluorination of the fuel, separation of plutonium, uranium, and fission product fluorides by partial thermal decomposition and uranium hexafluoride rectification in a distillation column.

Fluorination of the powdered oxides was performed in a flame fluorinator in a combination with a fluidized bed fluorinator (for the secondary fluorination of plutonium oxides). The fluorides formed passed through two types of condensers and an apparatus for thermal decomposition of plutonium hexafluoride, apparatus for uranium hexafluoride purification by rectification and columns packed with sodium fluoride and aluminum oxide pellets. The whole line called Fregat was installed in hot cells of the Institute of Atomic Reactors in Dimitrovgrad and operated with a capacity of 1 - 3 kg of spent uranium - plutonium oxide fuel per hour.

3.4.1.1. The spent fuel

The fuel to be reprocessed was spent fuel from the BOR-60 fast reactor operated in the Institute of Atomic Reactors in Dimitrovgrad. It was a pelletized UO_2 - PuO_2 mixture in stainless steel cladding enriched by 90 % of ^{235}U and 15 - 25 % of ^{239}Pu.

The burn-up of fuel was 10 -15 %, cooling time 15 - 30 days. Considering this high activity only inorganic compounds could be used in the reprocessing procedures.

Fuel for reprocessing was prepared in the Institute of Inorganic Materials in Moscow. The decladding of the fuel was realized by fusion of the cladding at 1550 ^{o}C. The pelletized UO_2 was converted by voloxidation to powdered U_3O_8 or by mechanic grinding to powdered UO_2 (PuO_2).

3.4.1.2. Process flowsheet

The technological flowsheet is shown in [6]. The process starts with the inlet of fluorine and nitrogen through protection columns loaded with pelletized sodium fluoride heated at 100 ^{o}C. The protection columns are acting against the blow-back of technological gases in the case of accident in the fluorinator.

The fluorinator has two functions, the first one is the fluorination of uranium oxides to UF_6 and of some fission product elements to volatile fluorides in the flame part of the apparatus. The second function is the fluorination of plutonium in the pseudo-fluidized-bed part of fluorinator proceeding at a big excess of fluorine. The bed is formed by pelletized corundum.

The fluorinator has a duplicator for heating and cooling. It is heated by air preheated in a calorifer to 200 - 250 ^{o}C. Fluorine gas is preheated in an electrical heater to 500 ^{o}C. Under these conditions, the reaction between uranium oxides and fluorine gas begins immediately after starting up the feeding.

There is a pre-condenser held at 80 ^{o}C intended for the condensation of the not very volatile products like NbF_5, SbF_5 and some other fluorides and oxide fluorides. Nearly 99 % of products are condensed in the first tube condenser (temperature around 0 ^{o}C) cooled by portions of liquid nitrogen. The last residues of products are trapped at - 40 ^{o}C in the second tube condenser packed with Rashig rings made of Monel metal.

The last traces of PuF_6 are retained in the protection column loaded by pelletized sodium fluoride and heated at 350 ^oC.

The reactor for thermal decomposition of PuF_6 operates at 350 ^oC either as a thermal decomposer or as a reducer (by using CO_2).

There is a reservoir intended for the collection of the unconsummated fluorine, nitrogen and more volatile fission products. Before operation the reservoir is evacuated. Fluorine and fluorides are trapped at the pelletized large-surface-area A12O3. Temperature of the column must be controlled as the reaction is exothermic.

For the secondary fluorination, a circuit comprising the fluorinator, condenser, sorption column and circulation compressor are used. Constant pressure is maintained in the system throughout the time of fluorination. Fluorine is refilled through the fluorine inlet to the fluorinator.

UF_6 and fission product fluorides are collected after thermal decomposition of PuF_6 in a condenser. The condenser is then heated to 90 ^oC and the liquid UF_6 is pumped to a distillation column. Pure UF_6 is filled after rectification into a container situated outside of the hot cell.

The exit gases are passing through absorption columns filled with $A1_2O_3$ and activated charcoal to the exhaust ventilation.

The hot cell where the reprocessing unit was situated had been designed specially for this installation. The cell was 12 m long and 3 m wide. Composition of the technological gases was determined by mass-spectrometry. Movement and settling of plutonium fluorides has been observed by neutron detectors.

The fluoride process technology was worked out in co-operation of two institutes, namely of the NRI Řež and Kurchatov Institute, Moscow in 1987 -1988.

The technological apparatus was designed in the Institute of Chemical Equipment at Brno, Czech Republic and produced in the NRI Řež. Equipment for thermal decomposition of PuF_6 was designed and made in Kurchatov Institute, Moscow.

Valves of special construction, manometers, differential manometers, and flowmeters have been designed and manufactured in the Institute of Nuclear Fuel, Prague - Zbraslav.

The line Fregat was subjected to functional and technological bench tests in the Nuclear Research Institute Řež. Depleted U_3O_8 was used as a fuel in these tests. After completion of the tests the entire technology was transported to the NIIAR institute in Dimitrovgrad, the former Soviet Union to make experiments in hot cells [7]. In both cases the function of fluorination, UF_6 condensation, UF_6 distillation, and waste gases absorption blocks was verified.

In the NIIAR the line was remotely controlled from the operator's panel. The flame fluorination, fluorination efficiency, fluorinator capacity and fluorine excess necessary for complete reaction of the dosed uranium oxides were studied. It was found that the fluorinator capacity is 1 - 3 kg of powdered UO_2 per hour, temperature in the combustion chamber does

not exceed 650 ^{o}C even at the maximum capacity and up to 100 % fluorine excess is necessary for complete conversion of uranium oxides to UF_6.

The program itself and the co-operation with the Russian institutions was discontinued after the Chernobyl accident in 1988, mainly due to the lack of financial means available for development of new technologies.

3.5. Involvement in the ADTT

The transmutation process enables the decomposition of transuranium elements into fission products in a subcritical reactor. Thus, it is possible to transform the spent fuel from nuclear reactors as well as the military plutonium and americium waste to the shorter-lived fission product nuclides.

Spent fuel from the LWR reactor contains about 95 % of uranium (UO_2), about 5 % of cladding metal (Zr, Nb), fission products and transuranium elements. Chemical processing in the molten salt fluorides is suitable for simple separation of uranium by fluorination. According to the experience of ORNL with the molten-salt breeder reactor, MSBR, it would be possible to obtain 99 % of uranium by fluorination from fluoride salt melt.

A program was proposed and is being carried out at present on the development of spent LWR reactor fuel reprocessing before its transmutation in the ADTT process. The main task of the reprocessing is the separation of uranium; transuranium elements and some fission products and their fixation in a fluoride glass. Technological scheme of the process was developed and some of the operations were verified experimentally at a laboratory scale. The scheme supposes the dissolution of fuel elements in molten alkaline fluorides in the presence of hydrogen fluoride, separation of uranium by reacting with elementary fluorine to form volatile uranium hexafluoride, separation of transuranium elements and some fission products by the use of physical or chemical procedures. The separated fission products are fixed in the form of fluoride glass by melting with calcium difluoride.

On the base of literature information and our own experience a technological flow sheet of the reactor fuel treatment before and after the transmutation was proposed. It comprises the following operations:

1. Dissolution of spent reactor fuel elements (pelletized UO_2) and cladding material (Zr) in fused fluoride salts in the presence of hydrogen fluoride. This reaction was studied at a laboratory scale last year. We have measured the kinetics of the dissolution of U, Al, Zr and UO_2. The molten fluoride salts composed of LiF, KF and NaF have a m. p. of 450 ^{o}C.

2. Fluorination is carried out by contacting dissolved uranium, zirconium, fission products and transuranium elements with elementary fluorine in fused fluoride salts. Uranium is released from the reactor in the form of gaseous UF_6 together with the volatile fission products, for example MoF_6, TcF_6, etc. Only nonvolatile fluorides, transuranium and fission product elements remain in the molten salt.

3. Separation of other components from molten salt. There is no experience on this subject in our laboratory, but according to information from Los Alamos, electrowinning or other physical methods are recommended for separation of Zr and some fission product elements.

The project "Experimental Molten Salt Loop for ADTT program" is carried out in the Škoda Works in Plzeň, Czech Republic. In the frame of this project technological loop for studying of molten fluoride salts characteristics was designed and fabricated and then installed in the NRI Řež.

3.6. Conclusions

In order to determine optimum flowsheets of chemical processes both for spent fuel processing before the introduction in the reactor and for the treatment of the fluoride molten salts taken-off the reactor, the following range of problems needs to be considered by research organizations:

1. Dissolution of spent fuel elements and materials containing plutonium and other transuranium elements in a molten salt medium by the reaction with anhydrous hydrogen fluoride.
2. Separation of certain metals before the irradiation fluorination (by electrolysis, extraction with metals).
3. To elaborate a simple technological flowsheet by utilizing the pieces of knowledge acquired in the work on the separation of fission products by molten salt processing (within the framework of MSBR) and the information on new processes based on physical methods of separation of elements.
4. To elaborate a process for the regeneration of the valuable LiF-BeF_2 mixture.
5. To verify experimentally the electrolytic precipitation of the individual elements or of whole groups of elements.
6. To verify experimentally the efficiency of fluorination with F_2 in the molten salt medium.

4. PROPOSAL FOR A COMPLEX REACTOR PHYSICS INVESTIGATION OF TH CONTAINING FUEL

4.1. The concept of an experimental blanket

There has been a convenient blanket concept for an efficient nuclear incineration of PWR spent fuel developed as a combination of those two ideas described in the paras above. The concept is illustrated by the Figures 2 and 3 where two zones are indicated, one under-moderated (red fuel) and thus better equipped for actinides burning and the second well-moderated (green fuel channel in a graphite block) and thus more convenient for fission products incineration. There have been a similar system proposed which will be utilized for a complex neutronic research of liquid Th containing fuel either by computational analyses or as an inserted core into the experimental reactors available (the LR-0 reactor will be used for full-scale core (driven by a suitable driver) static mostly and the VR-1 reactor for studies of time behavior of the coupled system of a subcritical core driven by an external neutron source.

The first estimations of basic neutronic characteristics of the complex system as shown in the Fig. 4 are illustrated by the Figs. 5 and 6.

4.2 The concept of complex technology testing

There has been a complex basis for engineering investigations of liquid fuel and coolants based on molten fluorides of different compositions under development in the frame of the national Consortium TRANSMUTATION in the Czech Republic. The first stage, i.e. a single

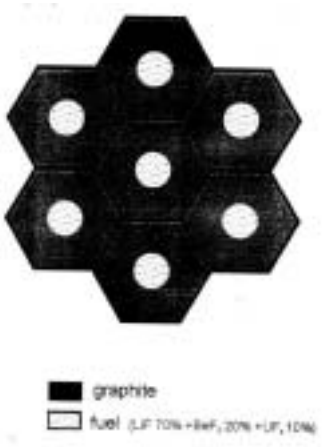


Figure 2. The experimental transmuter LA-0 one - zone blanket concept.

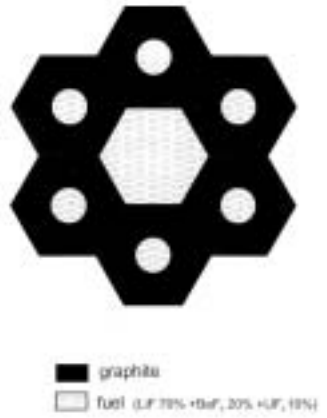


Figure 3. The experimental transmuter LA-0 two - zone blanket concept

Figure 4. The experimental transmuter LA-0 blanket concept for complex neutronic testing.

loop with a natural circulation of molten salts (it means without a pump) as shown on the Fig. 7, was realized by the SKODA Works, Nuclear Machinery Ltd., and a more matured one (with a pump) as shown on the Figure 8 is under construction in the NRI Řež plc. and will be put into operation very likely by the end of this year. Both those loops will be utilized for a complex technology testing of the Th containing liquid fuel, too.

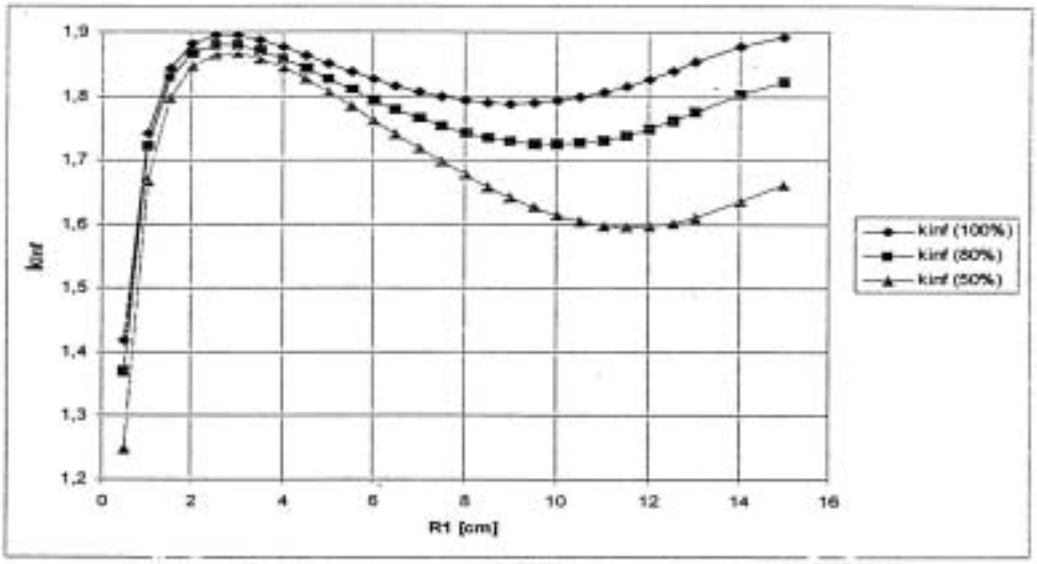
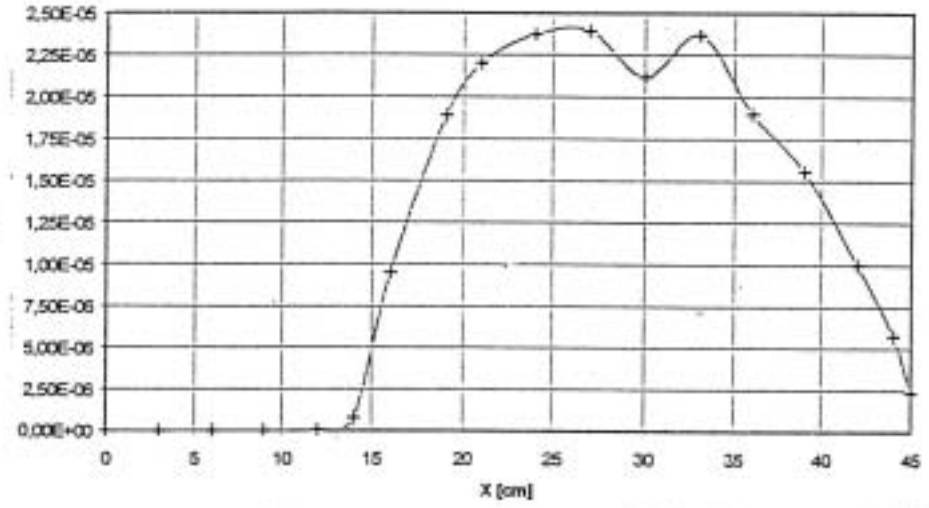


Figure 5. The blanket multiplying characteristics optimization.



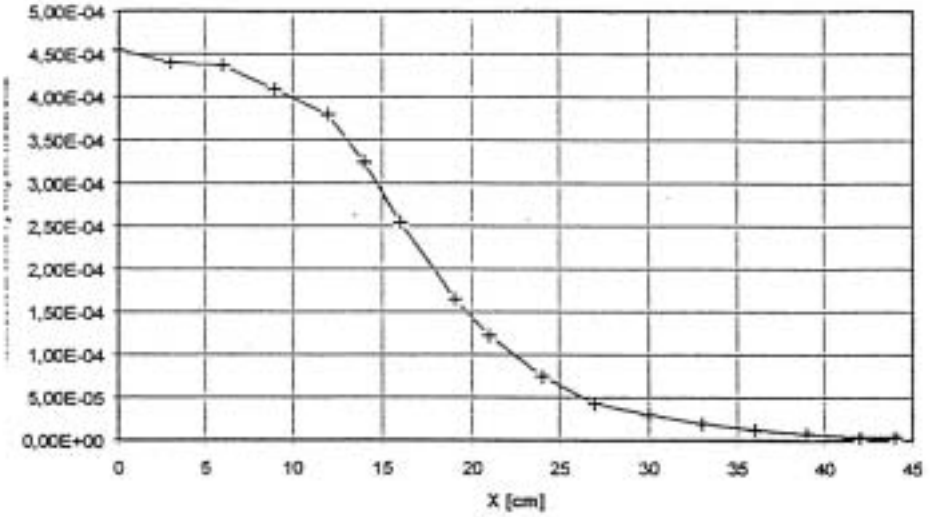


Figure 6. The thermal and fast neutron fluxes in the LA-0 blanket.

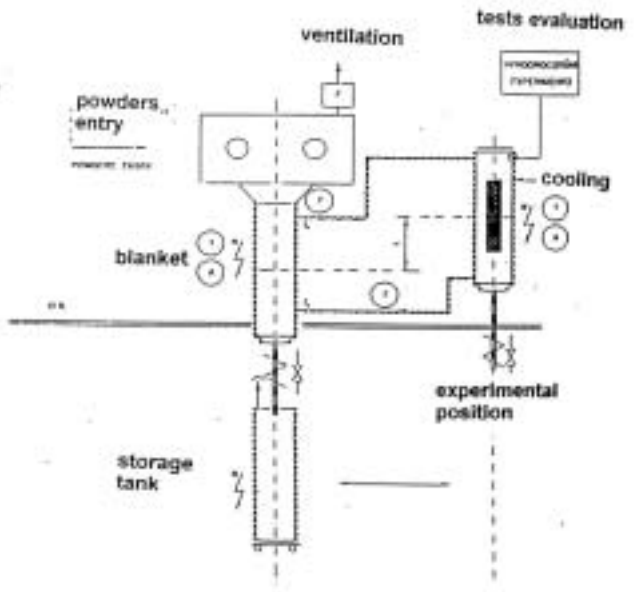


Figure 7. The single loop with a natural circulation of molten salts.

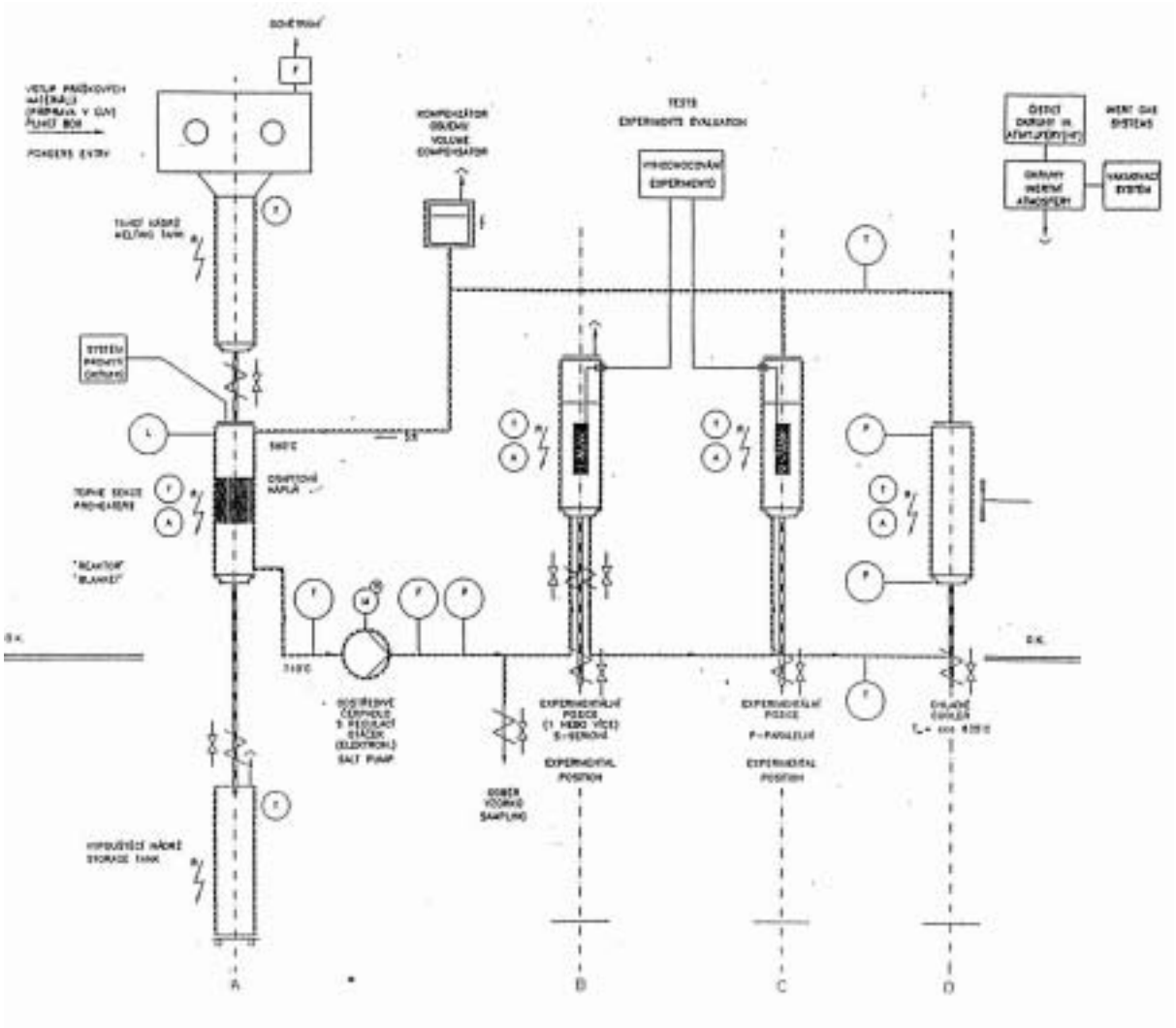


Figure 8. The loop with a forced circulation of molten salts.

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ASSESSMENT OF ^{232}TH NUCLEAR DATA THROUGH CRITICAL EXPERIMENTS USING THE KYOTO UNIVERSITY CRITICAL ASSEMBLY*

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Abstract. An assessment of ^{232}Th nuclear data was conducted through the analysis of critical experiments using the Kyoto University Critical Assembly (KUCA) of the thermal neutron system. The impact of the difference among evaluated nuclear data files was also examined for ^{232}Th and ^{233}U through the neutronics calculation. It was found that C/E values for cores containing ^{232}Th became larger with hardening neutron spectrum, whereas these values for cores without ^{232}Th were rather constant regardless of neutron spectrum. This fact indicates that there remains a certain problem in the evaluation of ^{232}Th capture cross sections, which would be desirable to be solved for the purpose of developing the Th-U fuel cycle. It was also found that the difference in ^{233}U fission and capture cross sections compiled in different nuclear data files could lead to a significant difference in reactivity. This result indicates that the assessment of ^{233}U nuclear data through the systematic critical experiments would be inevitable to develop the Th-U fuel cycle.

INTRODUCTION

It was recognized from the early days of nuclear energy development that Th would become a practical energy source, although the transmutation of ^{232}Th into ^{233}U is inevitable to realize the Th-U fuel cycle. Attractive points of the Th-U fuel cycle can be summarized as follows:

(1) The abundance of thorium resource is estimated to be triple of the uranium resource.
(2) The thermal breeding is feasible, whereas the fast breeding is necessary in the current U-Pu fuel cycle.
(3) The production of transuranium (TRU) elements is essentially lower compared with the U-Pu fuel cycle.
(4) Since high energy gamma rays are emitted from the Th-U fuel, it is considered to be beneficial for the nonproliferation policy of nuclear weapon materials, although it arises additional difficulties to handle the Th-U fuel.

Recently, in accordance with the delay in practical use of fast breeder reactors, the Th-U fuel cycle has internationally been paid attention again especially from a view point of the transmutation or incineration of TRU elements including both Pu and minor actinides (MAs).

In Kyoto University Research Reactor Institute (KURRI), an experimental study on the Th-U fuel cycle was initiated in 1977 by using a multi-core type critical assembly, Kyoto University Critical Assembly (KUCA). This facility was established in 1974 for the joint use program among Japanese universities aiming both the research and education of reactor physics. Thereafter, the critical experiments have been intensively carried out in a solid moderator core of the KUCA by using thorium metal plates [1-5]. In addition, basic experiments on fusion-fission hybrid reactors containing Th have been also carried out in combination with a Cockcroft-Walton type accelerator installed in the KUCA [6]. Furthermore, for the promotion of experimental study on the Th-U fuel cycle in the KUCA, a future plan is being considered

* 1999 meeting.

to perform critical experiments by purchasing the denatured ^{233}U fuel from United States [5,7].

The present paper describes the results of assessment on the ^{232}Th nuclear *data* obtained through the analysis of critical experiments in the KUCA. This paper also includes some results of neutronics calculations concerning the impact of the difference in evaluated ^{232}Th and ^{233}U cross sections to nuclear characteristics in the Th-U fuel cycle.

KUCA EXPERIMENTS ON THORIUM-URANIUM FUEL CYCLE

Brief Description of the KUCA

The KUCA is a multi-core type critical assembly of the thermal neutron system. It attained the first critical state in 1974. The horizontal cross section of the KUCA building is shown in Figure 1. The reactor room is divided into four sections. The A and B cores are solid moderator cores where either polyethylene or graphite can be employed as the moderator and reflector. The C core is a light water moderated and reflected core. A Cockcroft-Walton type accelerator can generate 14 MeV neutrons by D-T reactions and these neutrons can be injected into any core of the A, B and C cores. The B-core has been utilized for a series of critical experiments for the study on the Th-U fuel cycle.

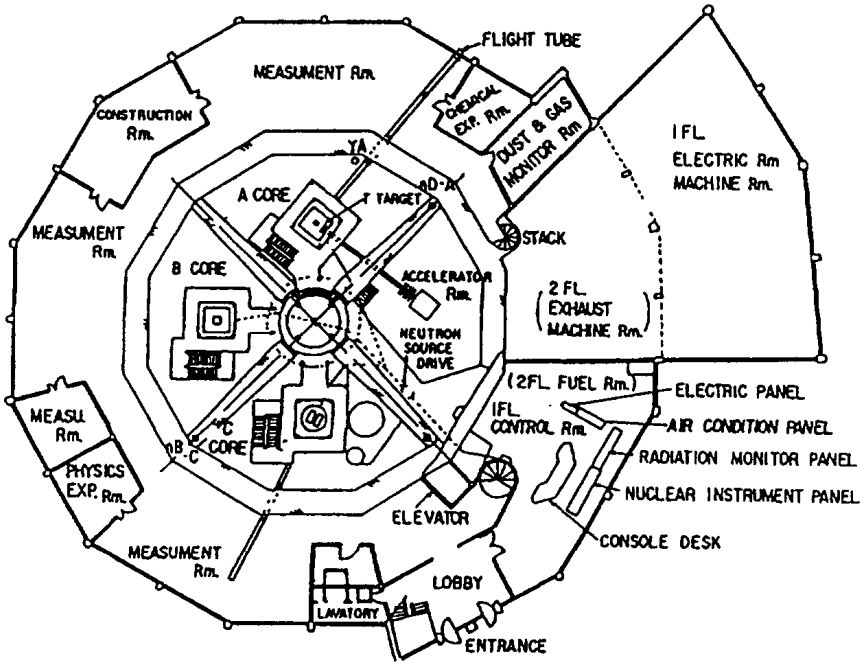

Figure 1. Horizontal cross section of the KUCA building

In the A and B cores, several material plates with a cross section of 5.08 cm (2") square can be used for the critical experiment These plates are made of 93 % enriched uranium-aluminum (U-A1) alloy (EU), natural uranium metal (NU), Th metal, polyethylene, graphite and so on. Various core elements can be formed by piling up these material plates in the Al square pipes of 1.5 m in length with a certain combination as illustrated in Fig. 2. Then, they are arranged by sticking onto the grid plate of the solid moderator core to construct the critical assembly.

KUCA Solid Moderated Core

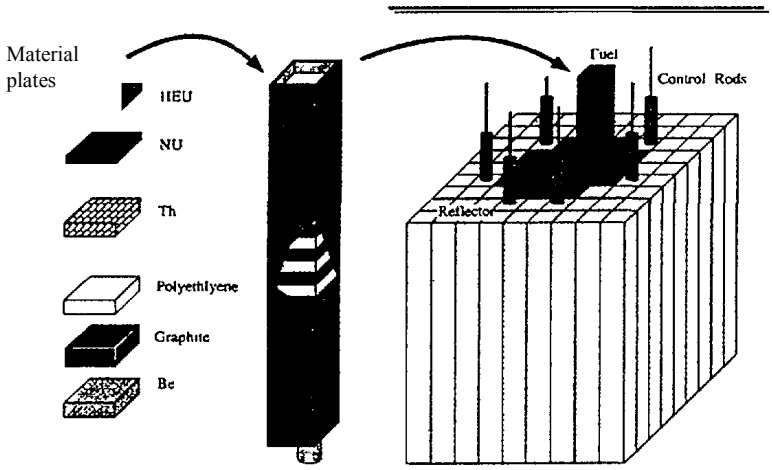

Fig. 2 Construction method of solid moderator cores in the KUCA

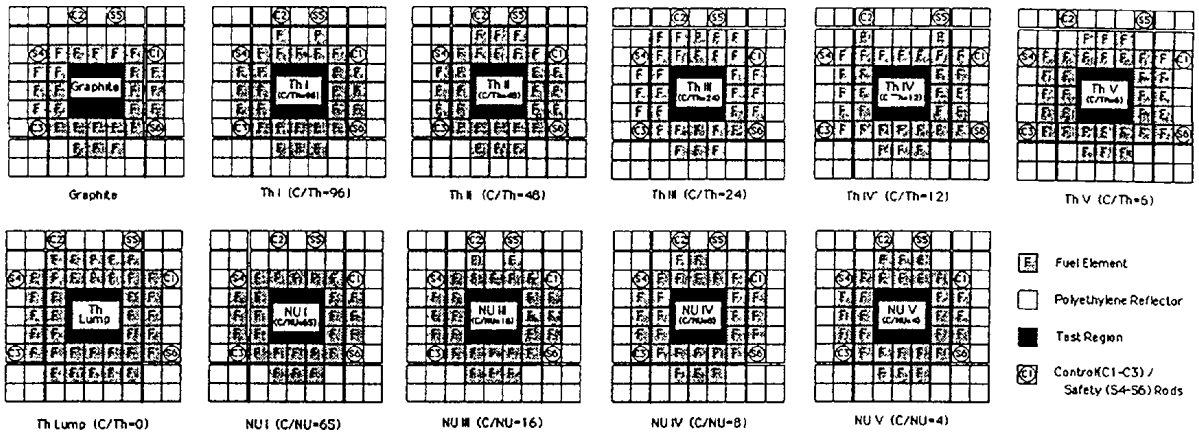

Figure 3. Configurations of zone-type cores

Polyethylene moderated and reflected cores have been employed in a series of critical experiments for the Th-U fuel cycle. The thickness of thorium metal plate employed was 3.18 mm. The EU plate of 1.59 mm in thickness was used to attain the criticality, and the NU plate of 1mm in thickness was also used as a reference material to examine the accuracy of Th nuclear data. The neutron spectrum in the core was systematically varied by changing a combination of EU and polyethylene plates in the fuel cell. Two types of cores have been utilized in the KUCA experiments: zone-type cores and single region cores with thorium in the fuel. Note here that these experiments have been conducted as the joint use program of the KUCA among the Japanese universities.

Critical Experiments by Using Zone-type Cores

In the early days of the experimental study on the Th-U fuel cycle in the KUCA, critical experiments of zone-type cores were intensively carried out for the purpose of grasping the nuclear characteristics of thorium in the nuclear reactor of thermal neutron system [1,2]. In this series of experiments, a central part of the polyethylene moderated and reflected cores

loaded with EU plates was substituted for a test zone containing thorium metal plates as shown in Figure 3. The H/^{235}U atomic ratio of a driver zone was 316. Critical experiments were performed by systematically varying the ratio of thorium and graphite in the test zone as shown in Figure 4. For reference, additional experiments were performed by substituting thorium for NU in the test zone to examine the accuracy of ^{232}Th nuclear data through the comparison between the two series of experiments using thorium and NU.

Th & NU Test Zones: mixture of Th or NU and graphite (C) plates
C/Th = 0 (no graphite), 6, 12, 24, 48, 96 : 6 cores
C/NU = 4,8, 16, 65: 4 cores

Th Test Zone Elements		NU Test Zone Elements	
C/Th	Th Structure	C/NU	Structure
0	Th Lump(1/8"Th*175)	4	[3mmNU+1/4"C]*58 cells
6	(1/8"Th+1/4"C]*58cells	8	[3mmNU+1/2"C]*36 cells
1	2 [1/8"Th+V2"C]*35 cells	16	[3mmNU +1 "C]*20 cells
2	4 [1/8"Th+1"C]*20 cells	65	(3mrmNU +4"C]*5 cells
4	8 [1/8"Th+2"C]*10 cells		
9	6 [1/8"Th+4"C]*5 cells		

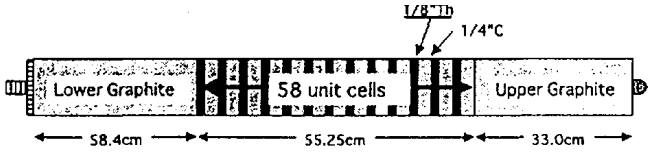

Fig. 4 Configurations of core elements employed in the zone-type core experiments

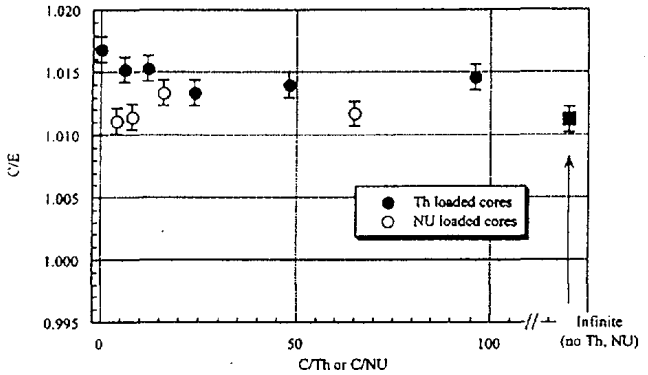

Figure 5. C/E values of zone-type cores containing thorium and NU in the test zone

By taking advantage of the recent progress in the continuous energy Monte Carlo code which is considered to be very suitable for the assessment of nuclear data, an analysis has recently been conducted by using the MVP code [8] and the JENDL-3.2 nuclear data file [4]. Through this analysis, it was found that there is a systematic difference in the C/E value of effective multiplication factor k_{eff} between the zone-type cores containing Th and NU as shown in Figure 5. In the MVP calculation, 1,000,000 neutron histories were traced to suppress the statistical error in k_{eff} less than 0.1%. From Figure 5, it is observed that C/E values are approximately constant for cores containing NU regardless of the difference in the ratio of

graphite/NU in the test zone. Note that C/E values suffer from a certain pedestal of approximately 1 % which is considered to be come from an inadequate evaluation of ^{235}U capture cross sections in JENDL-3.2. On the other hand, C/E values for cores containing Th show an increasing tendency with decreasing ratio of graphite/thorium in the test zone. This trend could be attributable to the Th capture cross sections compiled in JENDL-3.2.

Critical Experiments by Using Single Region Cores with Th in Fuel Cell

In the later experimental study on the Th-U fuel cycle in the KUCA, the Th plate has been introduced into the fuel cell which is essentially composed of the EU and polyethylene plates [3]. Therefore, cores employed in the experiments were changed from the zone-type core to the single region core. Using this kind of single region core, the systematic experiment could be performed by varying the ratio of not only polyethylene/EU but also Th/EU in the fuel cell. This means that effects caused by the change in neutron spectrum could be systematically examined through experiments, without suffering from the large heterogeneity due to the introduction of the test zone.

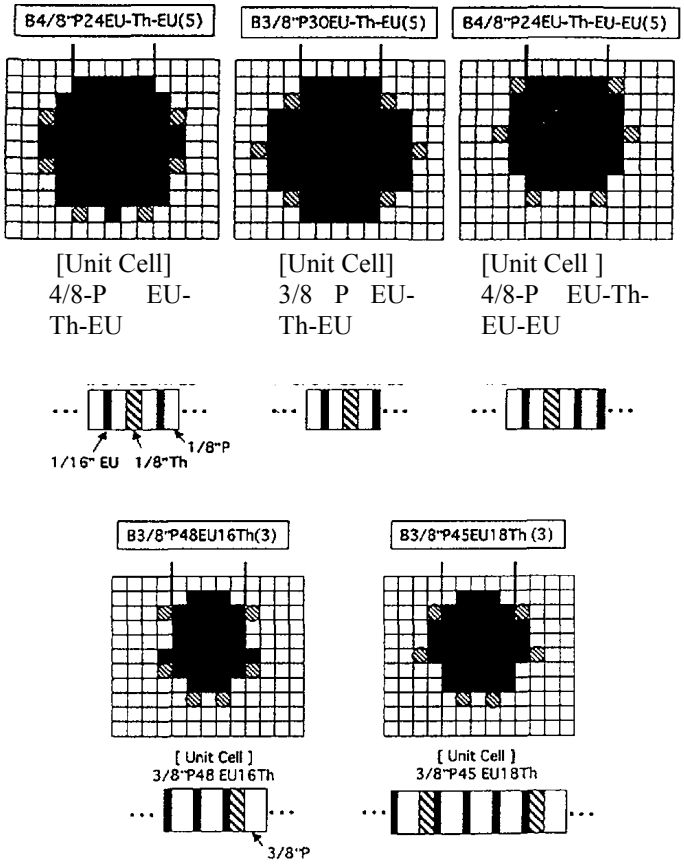

Figure 6. Configurations of single region cores with Th in the fuel cell

Figures 6 and 7 show core configurations of single region cores employed in the KUCA experiments and the cell averaged neutron spectra calculated by SRAC [9]. From Figure 7, one can notice that the neutron spectrum changes systematically with varying the ratio of Th/EU and polyethylene/EU. Besides the criticality of the core, the following quantities have been measured in this series of experiments; 1) the neutron flux distribution including the fine structure in the fuel cell, 2) the sample worth of various material including aluminum (Al),

99.5 % enriched ^{233}U-Al, 93.2 % enriched ^{235}U-Al and 99.8 % depleted ^{238}U-Al, 3) the reaction rate of several activation foils, 4) the prompt neutron decay constant α and 5) so on.

An analysis has recently been performed with the same method employed in the zone-type core analysis by using MVP and JENDL-3.2. The C/E values are shown in Figure 8. From this figure, one can clearly observe that the C/E value becomes larger with decreasing H/^{235}U atomic ratio and increasing the Th/^{235}U atomic ratio. This fact indicates that the C/E value becomes larger with hardening neutron spectrum in the core. This trend could be also attributable to the inaccuracy of the Th capture cross sections.

From the early stage of the KUCA experiments, all university researchers strongly hoped to conduct the critical experiments by using ^{233}U fuel, since ^{233}U is the main fissile element in the Th-U fuel cycle. For the promotion of experimental study on the Th-U fuel cycle in the KUCA, a future plan is being considered in real earnest to perform the critical experiment by purchasing the denatured ^{233}U fuel of 11.5 % from United States. In view of the future critical experiment using the ^{233}U fuel in the KUCA, preliminary neutronics calculations were executed by assuming the use of the denatured ^{233}U fuel plate in the solid moderator core as shown in Figure 9.

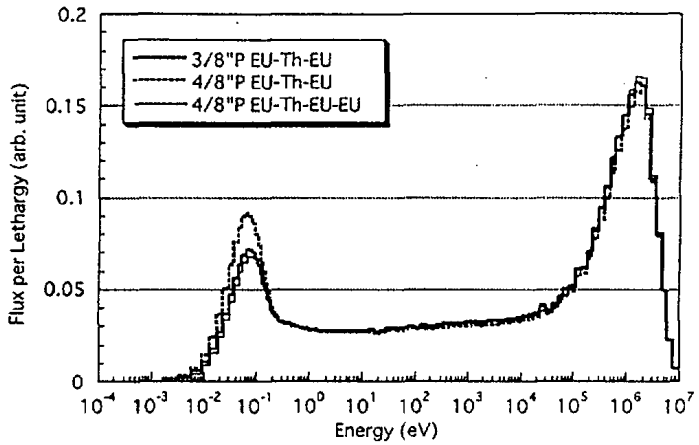

Figure 7. Cell averaged spectra of single region cores with Th in the fuel cell

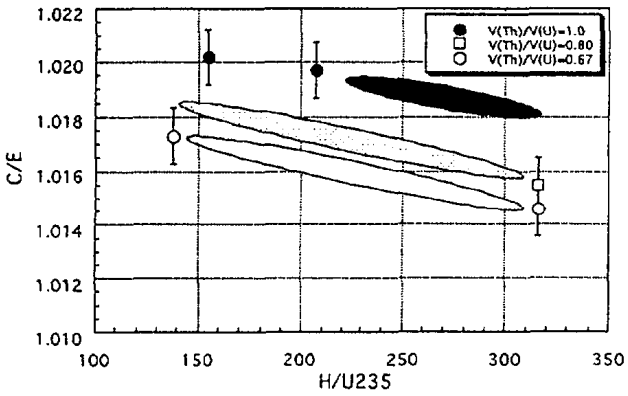

Figure 8. C/E values of single region cores with Th in the fuel cell

Future Plan of KUCA Experiments

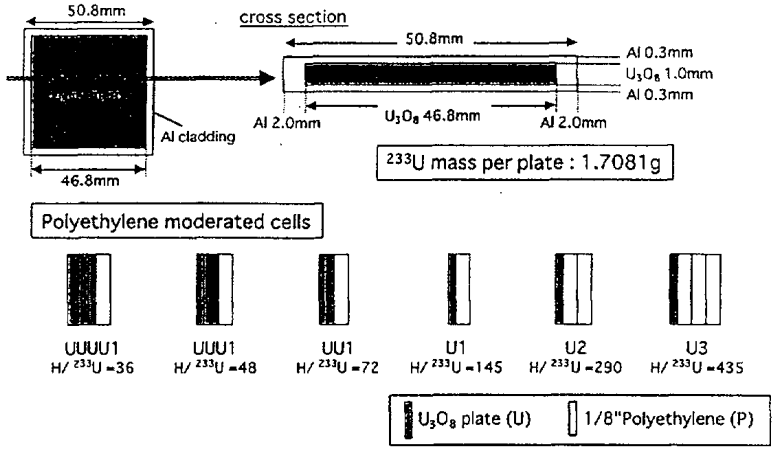

Figure 9. Structure of the fuel plate containing U_3O_8 powder and patterns of the fuel cell

The results of neutronics calculations executed by SRAC on the basis ofJENDL-3.2 are shown in Figures 10 through 13. Three kinds of fuel plates are considered here; 1) the U_3O_8 powder fuel as shown in Figure 9, 2) the uranium metal fuel with the same structure as shown in Figure 9 and 3) the U-A1 alloy fuel of the same size as shown in Figure 9 excluding Al canning. The infinite multiplication factor k_∞ is shown in Fig. 10 as a function of the H/^{233}U atomic ratio. From this figure, it is found that the k_∞ value reaches the maximum in the range from 80 to 100 in H/^{233}U. One can easily suppose that the critical mass of U-A1 fuel would become much larger than the others, since the k_∞, value of U-A1 fuel is much lower than the others. The ^{233}U mass needed to attain a critical state is shown in Figure 11 as a function of the H/^{233}U atomic ratio, m this calculation, the ANISN routine installed in SRAC was used by assuming a spherical core with the polyethylene reflector of 20 cm in thickness. From Figure 11, it is found that the critical mass becomes larger with decreasing H/^{233}U atomic ratio and more than 3 kg of ^{233}U is necessary to attain criticality by using U-A1 fuel. The critical radius calculated by ANISN in SRAC is shown in Figure 12 as a function of the H/^{233}U atomic ratio. The critical radius of core loaded with the U metal or U_3O_8 powder fuel is less than 20 cm for almost all cell patterns examined. Especially, the critical radius loaded with the uranium metal fuel becomes smaller with decreasing H/^{233}U atomic ratio, which indicates the difficulty in performing critical experiments. For reference, the cell averaged spectra in cores loaded with the U_3O_8 powder fuel are shown in Figure 13. From this figure, it is found that the neutron spectrum becomes harder with decreasing H/^{233}U atomic ratio.

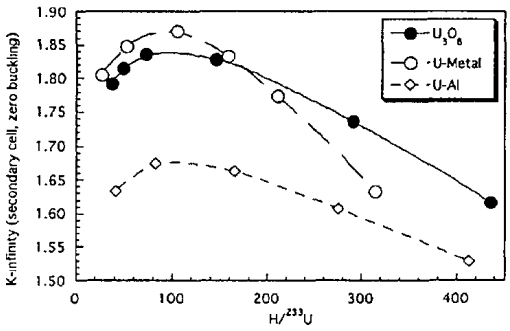

Fig. 10 Calculated k_∞ as a junction of the H/^{233}U atomic ratio

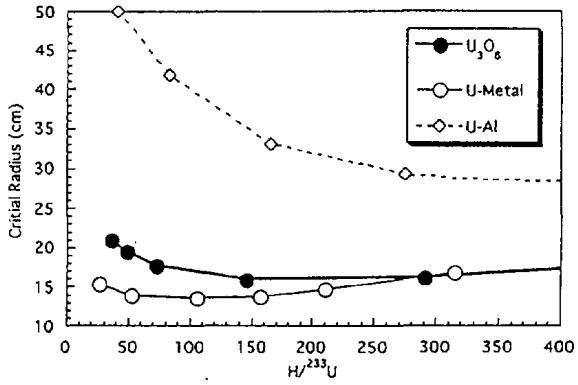

Fig. 11 Calculated ^{233}U critical mass as a function of the H/^{233}U atomic ratio

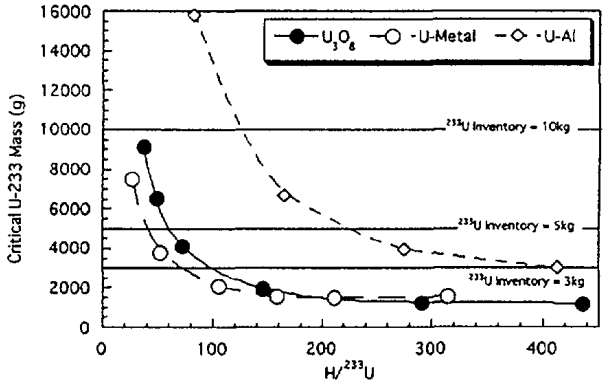

Figure 12. Calculated critical radius as a function of the H/^{233}U atomic ratio

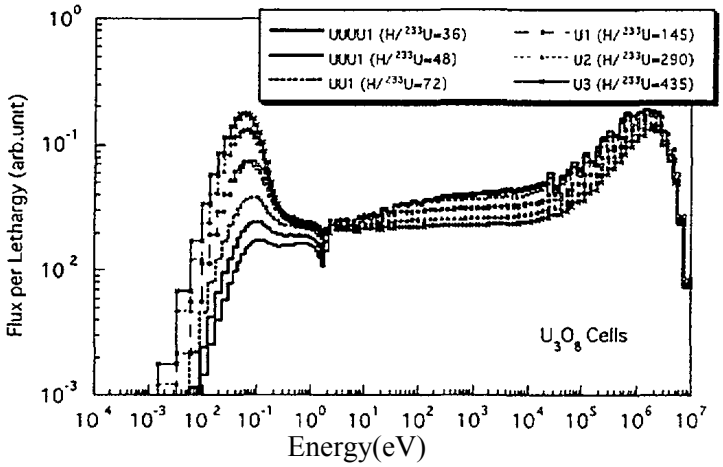

Figure 13. Cell averaged neutron spectra in cores loaded with the U_3O_8 powder fuel

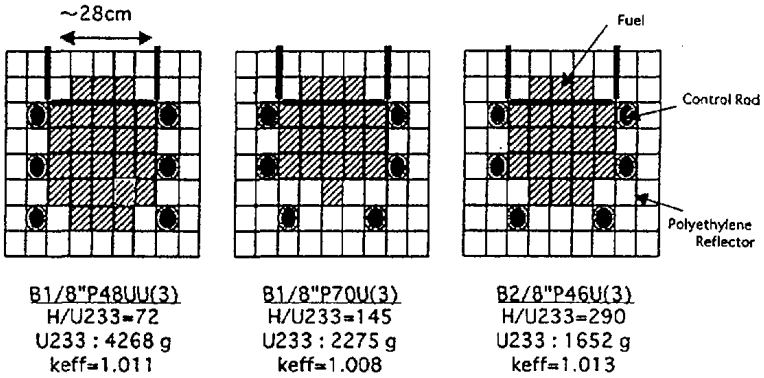

Figure 14. Realistic configuration of the critical core with the denatured U_3O_8 fuel in the KUCA

Since the critical radius obtained by the above calculation is rather small, more detailed calculations were executed by MVP on the basis ofJENDL-3.2 to get an image of a realistic core configuration loaded with the denatured U_3O_8 powder fuel. Configurations of critical cores are shown in Figure 14 together with the H/^{233}U atomic ratio, the ^{233}U mass, and the k_{eff} value when all the control and safety rods are fully withdrawn from the core. It was found that the size of the ^{233}U critical core is slightly larger than that of the smallest ^{235}U one constructed in the KUCA. Therefore, the critical experiments of ^{233}U fueled cores could be realized in the KUCA, if the denatured U_3O_8 powder fuel were successfully purchased from United States.

IMPACT OF DIFFERENCE IN EVALUATED NUCLEAR DATA LIBRARY TO NUCLEAR CHARACTERISTICS IN THORIUM FUEL CYCLE

Impact of Difference in ^{232}Th Cross Sections

The difference among the evaluated Th capture cross sections in JENDL-3.2, ENDF/B-VI and JEF2.2 is shown in Figure 15. From this figure, it is found that there exists very large difference at the valleys of cross sections adjacent to the resolved resonance peaks, although this difference is considered to affect the nuclear characteristics only slightly. The largest effect on the nuclear characteristics is caused by the difference in the unresolved resonance region.

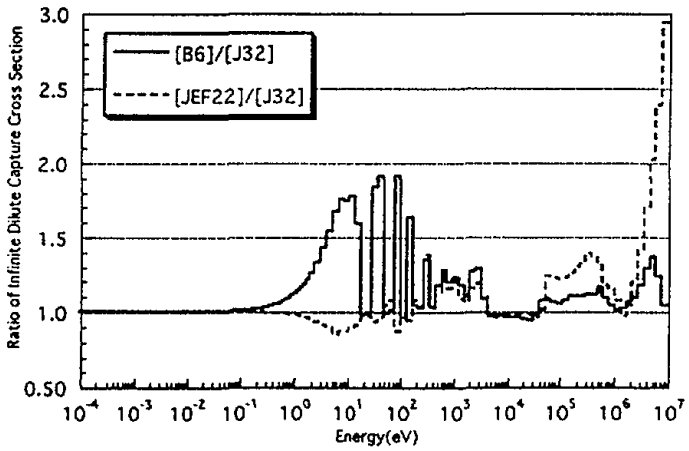

Figure 15. Difference among the thorium capture cross sections in JENDL-3.2, ENDF/B-IV and JEF2.2

The impact of the difference in thorium capture cross sections to the nuclear characteristics was examined through the cell calculation in a one-dimensional infinite plate geometry by using SRAC. An imaginary cell containing Th, EU and polyethylene was considered as shown in Figure 16. The E3 cell in this figure means that one 1/16" thick EU plate is sandwiched between 1/8" and 1/4" thick polyethylene plates. By substituting thorium cross sections of ENDF/B - VI for those of JENDL-3.2 in the k_∞ calculation, the reactivity difference becomes –1,565 pcm. The breakdown of contribution to this reactivity difference from each nuclide is also shown in Figure 16. From this breakdown, it is found that the difference in thorium cross sections in the energy range around 1 keV is dominant. The cancellation mainly between Th and ^{235}U can be seen in the lower energy range, which is considered to be caused by the spectrum change due to the difference in thorium cross sections in the unresolved resonance region around 1 keV. This result indicates that the systematic experimental study with varying neutron spectrum is quite useful to evaluate the thorium nuclear data.

Impact of Difference in ^{233}U Cross Sections

The difference among the evaluated ^{233}U cross sections in JENDL-3.2, ENDF/B-VI and JEF2.2 is shown in Figure 17. The difference is fairly large even in the lower energy range compared with that among the evaluated cross sections of ^{235}U, ^{238}U and ^{239}Pu which are major nuclides used in the current U-Pu fuel cycle. Therefore, it is considered to be inevitable to conduct the assessment of ^{233}U nuclear data through a series of experiments to promote the development of the Th-U fuel cycle.

The impact of the difference in ^{233}U cross sections to the nuclear characteristics was examined through the core calculation in a one-dimensional spherical geometry by using the CITATION routine installed in SRAC. It was assumed that a spherical core with a polyethylene reflector of 20 cm in thickness consists of the Ul cell shown in Figure 9. The Ul cell consists of one 1/16" thick denatured U_3O_8 powder fuel and one 1/8" thick polyethylene plates. The H/^{233}U atomic ratio of this cell is 145. A perturbation calculation was executed by substituting ^{233}U cross sections of ENDF/B-VI for those of JENDL-3.2, the reactivity difference becomes -0.5 %Δ/k which is considered to be more than -1 $ in the ^{233}U fueled system.

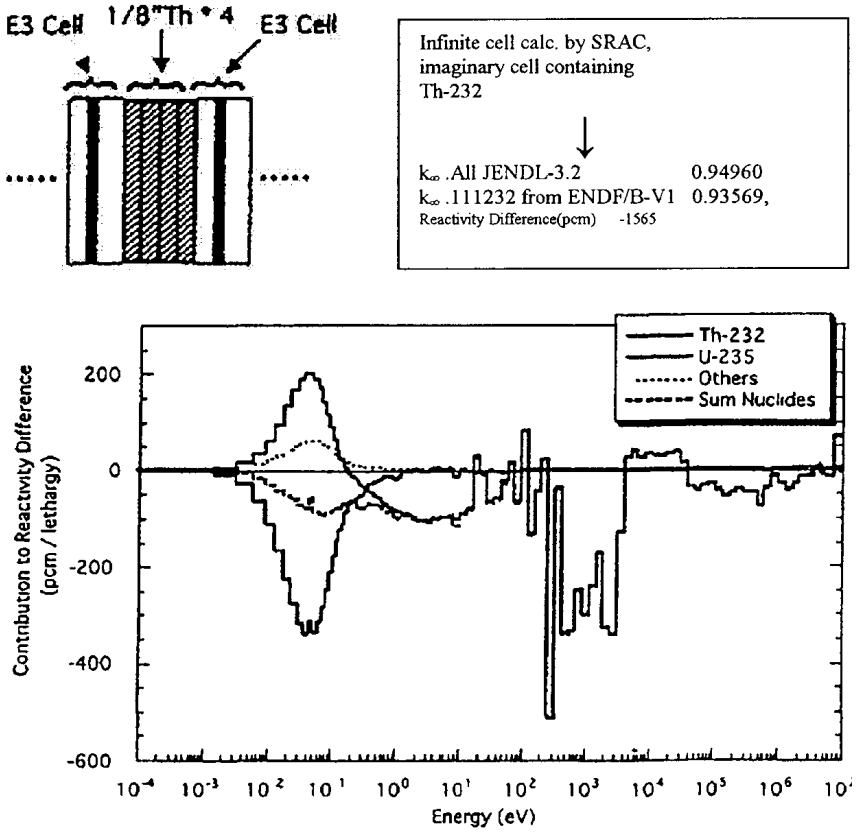

Figure 16. Impact of the difference in thorium cross sections to the nuclear characteristics

The breakdown of contribution to this reactivity difference from the fission, absorption, moderation and leakage terms is shown in Figure 18. From this breakdown, it is found that the fission term contributes to the negative reactivity with the largest magnitude, the absorption term to the positive reactivity with the second magnitude, the moderation term to the negative reactivity with the third magnitude, and the leakage term slightly to the negative reactivity.

The significant reactivity difference of -0.5 % Δk/k comes from the cancellation effect mentioned above. The negative contribution of the fission term is mainly caused by the difference in the energy range around 1 MeV. The positive contribution of the absorption term is mainly caused by the difference in the lower energy range. Since this magnitude of reactivity difference can be easily measured by the critical experiments, it is considered to be inevitable to perform a series of systematic critical experiments to promote the development of the Th-U fuel cycle.

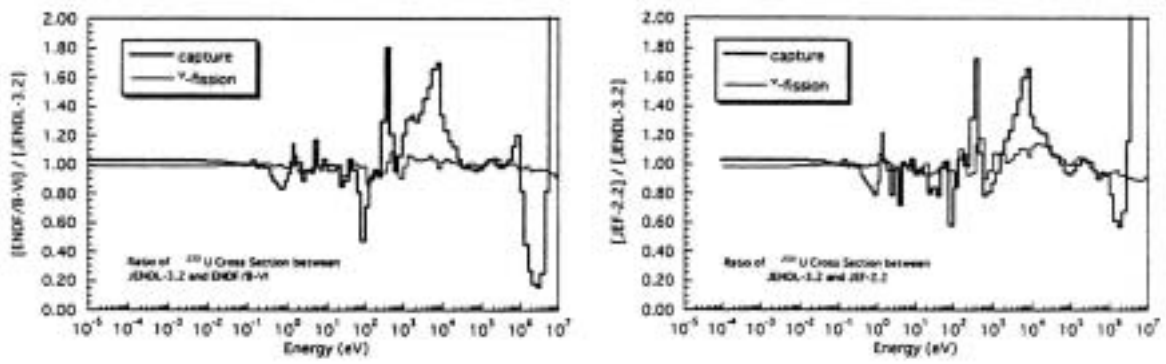

Figure 17. Difference among the ^{233}U capture and fission cross sections in JENDL-3.2., ENDF/B-IV and JEF2.2

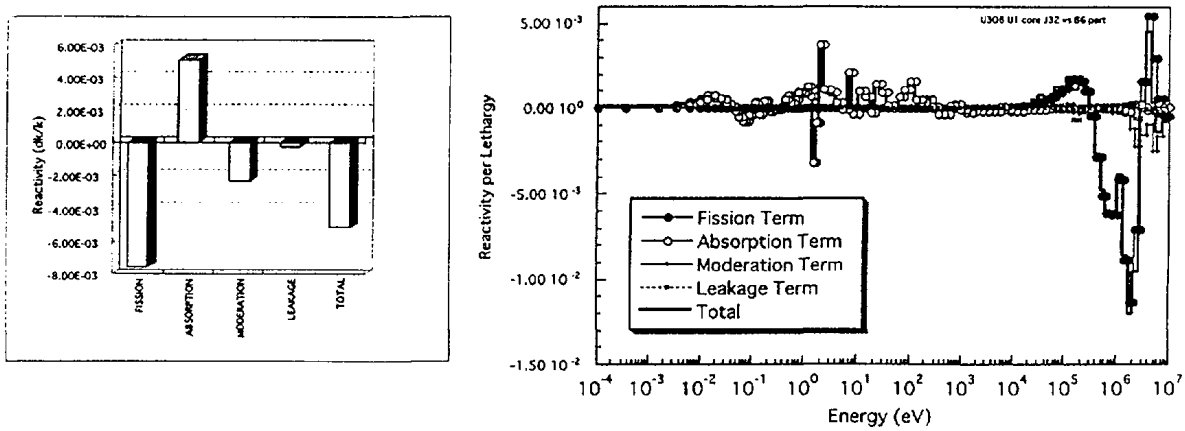

Figure 18. Impact of the difference in ^{233}U cross sections to the nuclear characteristics

CONCLUSION

From the assessment of thorium nuclear data through the KUCA critical experiments, it was found that the reevaluation of thorium capture cross section especially in the unresolved resonance region would be necessary to promote the development of the Th-U fuel cycle. The impact of the difference in thorium and ^{233}U cross sections compiled in the evaluated nuclear data file was examined through the neutronics calculation. It was indicated that the critical experiment using even the denatured ^{233}U fuel would be necessary to promote the development of the Th-U fuel cycle.

ACKNOWLEDGMENTS

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A. Hayashi of Rikkyo Univ. and their group members for their cooperation in the experimental study. They should thank to Prof. Keiji Kanda of KURRI for the generous support in the course of this study.

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IV. TECHNOLOGY AND DESIGN OPTIONS

THORIUM-BASED FUEL DESIGN FOR INCINERATION OF EXCESS WEAPON GRADE PLUTONIUM IN EXISTING PWRs*

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Abstract. The main objective of the present design is to use thorium based fuel for an efficient incineration of excess weapon grade plutonium. A heterogeneous, seed-blanket (SBU) fuel assembly design was adopted. The main design approach is to use plutonium as a seed fuel providing neutrons to a subcritical blanket loaded mainly with thorium. The seed subassembly fuel consists of Pu/Zr metal alloy and the blanket subassembly fuel consists of Th-Pu-U mixed oxide. The plutonium provides a fissile component, while natural uranium part is added to denature (dilute) the ^{233}U built-up in thorium. This design is usually designated as the Radkowsky Thorium Fuel (RTF). A simulation of an "equilibrium" cycle demonstrated the feasibility of the RTF design with an efficient plutonium incineration. Advantages of thorium-based SBU assembly design in compliance with the standard PWR control system requirements are also demonstrated.

Why thorium-based fuel and SBU geometry?

The efficiency of incinerating the excess weapon grade stockpiles by utilization of the mixed oxide fuel (MOX) is significantly reduced by the production of the "new" or the second generation plutonium. For the MOX fuel based on natural uranium, residual plutonium in discharged fuel amounts to 60-70% of the initial plutonium load. Thus, using the MOX fuel is equivalent to a transformation of the pure weapon grade plutonium into reactor grade plutonium contained within the discharged fuel. Replacing the uranium by thorium as a fertile material for plutonium incinerating cycle is investigated in this work in order to improve the efficiency of the plutonium incineration cycle.

A well known design problem associated with heavy plutonium loading required in the plutonium incinerating cycles is the reactivity control problem. The higher thermal absorption cross-section of plutonium, as compared with uranium, causes reduction of the reactivity worth of all LWR control mechanisms: control rods, burnable poisons and soluble poison, by approximately a factor of two. Several solutions were proposed and investigated, such as using enriched boron, gadolinium (Gd), or even additional control rods to compensate this effect.

An alternative approach is offered by a heterogeneous, SBU fuel assembly geometry. The SBU geometry allows separate lattice optimization for the seed and blanket parts. The seed region is well moderated (V_m/V_f =3.5) while the blanket part lattice is similar to a standard PWR (V_m/V_f = 1.7). In the present design the control rods and burnable poisons are concentrated in the seed region with a high moderator content. Thus, the reactivity worth of the control mechanisms is increased.

* 1998 meeting.

Fuel Management Scheme.

The fuel management scheme is based on two separate material flows for the seed and blanker fuels. The seed part of the core (consisting of all seed sub-assemblies) is managed in three batches, each residing 300 full power days (FPD's). Thus, the seed in-core residence time is 900 FPD's. The blanket is managed as a single batch residing for 6 seed cycles, i.e. 1800 days. This fuel management scheme is designed to assure an efficient utilization of thorium, in terms of natural uranium savings. In addition, a 3-batch seed reload scheme was chosen to provide an "optimal" balance between two different performance parametors: the plutonium incineration rate and the residual plutonium content in the discharged fuel. The first one should be maximized and the second one should be minimized. The Th-based fuel cycle proposed and investigated in this work was designated for a standard PWR core, similar to Westinghouse and/or EPR design. The design description is given below and in Figure 1:

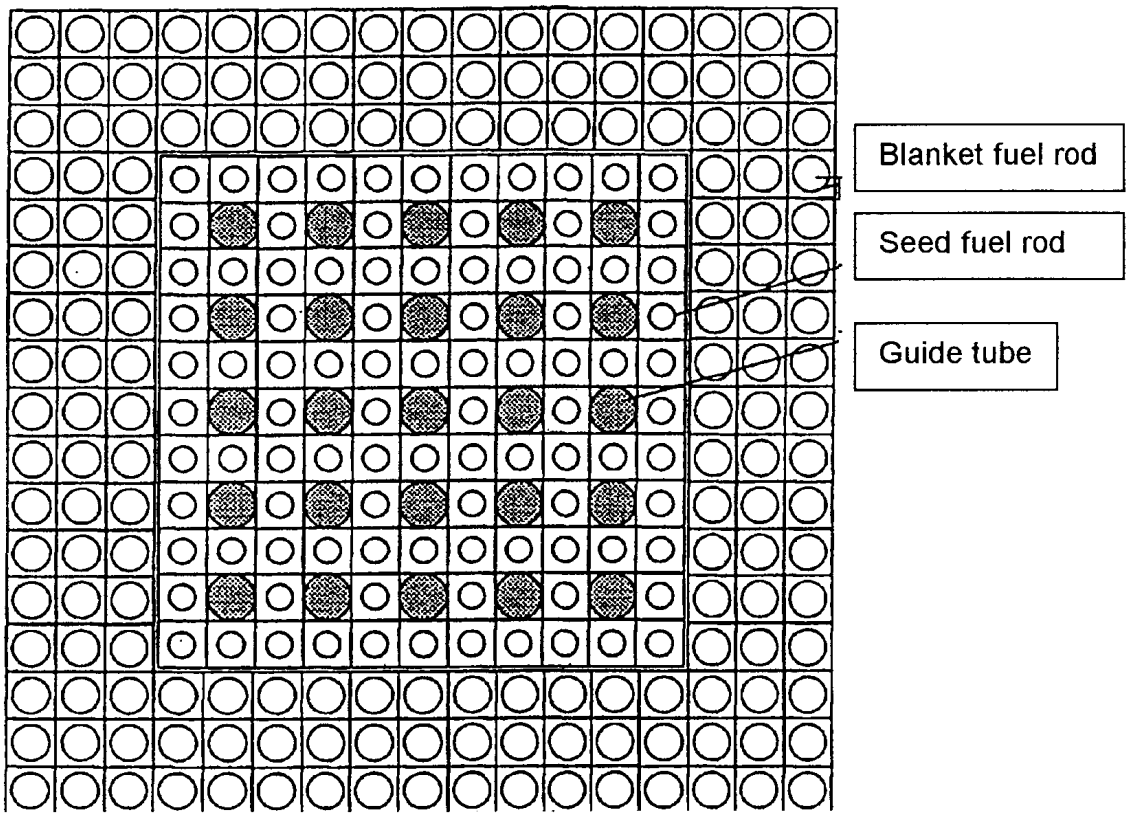

Figure 1. A Schematic View of a Seed-Blanket Unit (1/4 assembly)

Core Design Parameters

Power output (MW(th)) =	3,400
Number of fuel assemblies (SBU's) =	193
Average Power Density (w/c^3) =	104
Total coolant flow (kg/s) =	194X0

Seed Design Parameters:

Assembly Volume fraction (%) =	40.1
Composition:	7.0 weight % Weapon grade Pu 93.0% weight % Zr alloy.
Number of fuel rods =	96
Number of guide tubes =	24 (+ one central)
Moderator to Fuel Volume Ration =	3.535
Lattice (cell positions):	11x11
Cell Geometry: fuel pellet radius (cm) =	0.310
clad outside radius (cm) =	0.350 (no gap)
lattice pitch (cm) =	1.205
Average Fuel temperature (°C) =	470.0
Average Cladding temperature (°C) =	340.0
Average Moderator temperature (°C) =	306.0
Average Specific Power (MW/t) =	186.0

Blanket Design Parameters:

Assembly Volume fraction (%) =	59.9
Composition:	O.8% weapon grade Pu oxide + 8.2% Natural U oxide + 91.0% Th oxide.
Cell Geometry: fuel pellet radius; (cm) =	0.4095
clad outside radius (cm) =	0.475
lattice pitch (cm) =	1.258
Average Fuel temperature (°C) =	750.0
Average Cladding temperature (°C) =	340.0
Average Moderator temperature (°C) =	306.0
Number of fuel rods =	168
Number of guide tubes =	0
Moderator to fuel volume ratio =	1.659
Average Specific Power (MW/t) =	30.0

Results of calculations (equilibrium cycle).

A full simulation of the proposed cycle involves the calculation of a single blanket life-time, which is equivalent to 6 seed replacement cycles. In this work this full simulation is approximated by a calculation of the "equilibrium" cycle assuming that its performance parameters are representative of a complete simulation, i.e. 6 seed cycles.

The equilibrium cycle for a 3-batch fuel management scheme is represented by a core which includes three seed fuel types - fresh, once-burned, and twice-burned, and a single blanket fuel type with an averaged burnup value of 900 FPD's. Burnup dependent reactivity and power sharing between seed and blanket are summarized below. The hot channel power density and a summary of the mass flow for all important isotopes are in the following tables.

Reactivity, power sharing, and fuel temperature summary:

days	Keff	seed (MW)	blanket (MW)
0	1.06931	507.0	343.0
20	1.06460	503.4	346.6
100	1.04S01	490.6	359.4
160	1.03633	481.2	368.8
200	1.02S45	474.7	375.3
260	1.01613	464.6	3H5.4
300	1.00745	457.5	392.5
310	1.00522	455.6	394.3

Reactivity Control.

A series of assembly level calculations were carried out to evaluate the reactivity worth of the different control mechanisms and the moderator temperature coefficients. It should be noted that the values generated on the assembly level represent a "core averaged" evaluation and are applicable only for the comparison of different cycle options. Several U-Pu based cycle options were considered in order to obtain a clear picture of the reactivity controllability issue. The cycle options considered are designated as follows:

PWR - A standard slightly enriched U fuel,
MOX - A mixed oxide fuel (U-Pu oxide, reactor grade plutonium),
MOX239 - A mixed oxide fuel (U-Pu oxide, weapon grade plutonium),
TMOX - A thorium based homogeneous mixed oxide fuel (Th-Pu oxide reactor grade Pu),
TMOX239 - A thorium based homogeneous mixed oxide fuel (Th-Pu oxide weapon grade plutonium),
RTF239 - Radkowsky thorium fuel seed-blanket (weapon grade plutonium).

Summary of Performance Parameters.

A seed-blanket heterogeneous fuel assembly design combined with a Th-based fuel offers an attractive option for a plutonium-incineration cycle. Two main problems associated with plutonium incineration in LWR's: an overall efficiency of the plutonium destruction and the reactivity control issue are addressed adequately by the flexibility of the SBU geometry. A preliminary design analysis indicated that approximately 700 kg of plutonium per year may be incinerated and the residual plutonium "quality" is sufficiently deteriorated to reduce the danger of its diversion to weapon material. A set of assembly level calculations also shows that only 10% reduction in the reactivity worth of control rod and soluble boron reactivity control mechanisms for the RTF design, compared with approximately 50% reduction for all other fuel cycles options.

Hot Channel Power Density Summary:

Axial average power density: seed: 1240.3 (w/c^3 in fuel)
blanket: 247.6 (w/c^3 in fuel)
SBU: 126.46 (lattice)

Cycle Mass flow Summary (kg)

Core charge	Core inventory		Core discharge
seed fresh(64)	BoC seed fresh(64)	EoC seed once(64)	
Pu239 873.07 Pu240 55.73	Pu239 873.07 Pu240 55.73	Pu238 0.05- Pu239 546.37 Pu240 125.09 Pu241 32.50 Pu242 2.15	
	seed once(65)	seed twice(65)	
	Pu238 0.04 Pu239 565.82 Pu240 128.33 Pu241 29.10 Pu242 1.80	Pu238 0.25 Pu239 276.51 Pu240 165.34 Pu241 53.91 Pu242 9.26	
	seed twice(64) Pu238 0.21 Pu239 278.72 Pu240 163.40 Pu241 52.95 Pu242 8.60	seed out(64) Pu238 0.83 Pu239 91.54 Pu240 154.35 Pu241 57.67 Pu242 21.97	seed out(64) Pu238 0.83 Pu239 91.54 Pu240 154.35 Pu241 57.67 Pu242 21.97
Initial Load 47,484.0 - - - - 33 .4 4,664.8 - 475.0 30.3	blanket(193) Th232 46098.16 Pa231 3.95 U232 2.62 U233 633.70 U234 81.19 U235 20.32 U238 4376.22 Pu238 1.33 Pu239 63.09 Pu240 42.91 Pu24i 43.78 Pu242 35.12	 Th232 45629.82 Pa231 4.38 U232 3.73 U233 708.64 U234 115.20 U235 26.09 U238 4279.75 Pu238 2.10 Pu239 59.82 Pu240 29.95 Pu241 33.94 Pu242 40.78 1	

Weapon grade plutonium incineration.
Summary

	Total plutonium incinerated (kg/a)	Pu239 incinerated (kg/a)	Residual Fraction
seed	602	778	0.35
blanket	95	79	0.24
TOTAL	**697**	**857**	

Summary of Reactivity worth values.

Fuel Cycle Option	Soluble Boron (Δρ/ppm B)	Control rods worth (all rods inserted)
PWR	-6.50e-03	-0.3332
MOX	-2.97e-03	-0.2157
MOX239	-3.21e-03	-0.2217
TMOX	-3.05e-03	-0.23 IX
TMOX239	-2.97e-03	-0.223
RTF239	-5.X2e-03	-0.2936

Conclusion: The Radkowsky thorium fuel seed-blanket design demonstrated an efficient weapon grade plutonium incineration: high destruction rate and relatively low residual content. In addition, the reactivity control system of existing PWR cores seems adequate for the Radkowsky thorium fuel (RTF) plutonium -incinerator design.

THE INDIAN PERSPECTIVE ON THORIUM FUEL CYCLES*

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Abstract. India is a country with limited deposits of uranium and large deposits of thorium. This fact ensures that India's nuclear power program cannot be totally uranium based the way it today is in most countries that have gone for nuclear power. Therefore, almost from the beginning of India nuclear power program, some effort has always been expended towards developing the technology of the thorium cycle. This has included fuel cycle studies, technology development, inpile irradiations and health physics aspects. A complete study of thorium cycles in various reactor types led to the conclusion that heavy water reactors were second only to molten salt reactors in this respect. HWR then, was the natural option for India. Currently India is working on the design of an advanced heavy water reactor (AHWR), specially designed with thorium in mind. In this paper, the results of a few interesting studies involving the recycling of plutonium with thorium in PHWR are presented.

1. INTRODUCTION

Of all countries that have ongoing nuclear power programmes, India is unique in being a country with limited deposits of uranium, but vast deposits of thorium. Unlike other countries therefore, India could not set thorium aside and settle for the uranium cycle. India followed a steady, even though low key, programme on thorium fuel cycle studies, technology development, and inpile irradiation. A complete study of thorium cycles in various reactor types led to the conclusion that heavy water reactors were second only to molten salt reactors in this respect. HWR then, was the natural option for India. Consequent to the decision to go for thorium cycles in heavy water reactors, the following things were done:

(1) 500 kg of thorium in the form of fuel bundles has been loaded in each of the two units of the Kakrapar Atomic Power Station (KAPS-1 & 2) for the purpose of initial power flattening. This scheme will be followed for all future PHWRs as well.
(2) There has been a fairly continuous programme of (irradiating thorium rods in the reflector of the research reactor CIRUS.
(3) A ^{233}U fuelled experimental reactor PURNIMA-II was commissioned at Trombay in 1984. Another system, called PURNIMA-III was made critical in 1990. This made use of the same fuel as was used in KAMINI at a later date.
(4) A research reactor KAMINI fuelled by ^{233}U -A1 alloy was commissioned in 1996. This was designed at BARC and built at Kalpakkam. It will be primarily used for neutron radiography on active components.
(5) Capability for reprocessing thorium to extract ^{233}U has been developed here and the ^{233}U used in PURNIMA and KAMINI was extracted here.
(6) On the fuel fabrication side work has been done on automatisation and remotisation which is needed for the highly active ^{233}U fuel. Already completed work includes the fabrication of thorium bundles for PHWRs, and the fabrication of thorium-plutonium mixed oxide fuel clusters.
(7) (Th, Pu) oxide fuel has been irradiated in the pressurised water loop of the CIRUS reactor to a burnup of 18000 MWD/T, and performed well without failure.
(8) Currently India is working on the design of an advanced heavy water reactor (AHWR), specially designed with thorium in mind.

* 1997 meeting.

2. FUEL CYCLE STUDIES

Thorium cycles were analysed in all the extant thermal reactor types. These include:
a. Light water reactor;
b. High temperature gas cooled graphite reactor;
c. Molten salt breeder reactor;
d. Aqueous suspension heavy water reactor;
e. Open lattice pressure vessel heavy water reactor;
f. Pressurised heavy water reactor with pressure tubes;
g. Advanced heavy water reactor;
h. Source driven reactor.

All reactor types have their own special advantages, but the accent in our studies was on fuel utilization. From this perspective, it turns out that the best system (barring the source driven system) is the molten salt breeder reactor, with the heavy water reactors coming in second best. The molten salt technology being very different from what India is accustomed to, and the heavy water reactor being Indians chosen reactor type, the logical sequel was to continue the examination of thorium cycles in heavy water reactors. We shall return to the fuel cycles later on in this paper.

3. WORK DONE IN INDIA TOWARDS THORIUM UTILISATION

(i) Thorium has been used for initial power flattening in the PHWR for the first time in the Kakrapar Atomic Power Station (KAPS). This has been done in both units KAPS-1 & 2. About 500 kg of thorium in the form of 35 fuel bundles was used for this purpose in each unit. This also involved solving a somewhat intricate reactor physics problem [1] whereby these 35 thorium bundles could be distributed in the core in such a fashion as to achieve power flattening without adversely affecting the reactivity worth of the two shutdown systems in the reactor. These bundles have performed satisfactorily in the reactor. In unit-1, all thorium bundles have been discharged. It has been decided to follow this scheme in all future PHWRs.

(ii) A program of irradiating thorium rods in the reflector of the CIRUS reactor was started almost along with the commissioning of the reactor. This is being continued in a fairly sustained manner.

(iii) In 1984, an experimental reactor using ^{233}U as fuel commissioned in Trombay. This reactor was named PURNIMA-II [2]. It was a solution reactor in which ^{233}U in the form of uranyl nitrate was dissolved in water, and this solution was surrounded by a reflector of beryllium oxide. In 1990, a zero energy reactor using ^{233}U -A1 alloy plates as fuel was commissioned. It had nine fuel subassemblies and was named PURNIMA-III [3].

(iv) The 30 kW research reactor KAMINI [4] was built at Indira Gandhi Centre for Atomic Research (IGCAR), for neutron radiography, activation analysis, and radiation physics research. This used ^{233}U -A1 alloy plates as fuel. It had nine fuel subassemblies having a total of 72 U-A1 alloy fuel plates. This reactor was made critical for the first time on October 29, 1996.

(v) Two pilot scale facilities have been operated for the recovery of ^{233}U from thorium rods irradiated at CIRUS using a modified version of thorex process employing a 5% TBP extraction flowsheet.

The ^{233}U recovered from the above operations have been used to meet the fuel inventory requirements of Purnima- II, Purnime-III and Kamini. Efficient recovery and recycling of ^{233}U during the reactor experiments and from alloy scraps have played a crucial role in economic utilization of the ^{233}U resource. Periodic purification of ^{233}U to remove the daughter products of ^{233}U that emit high energy gamma radiations also forms a part of this effort. The expertise gained in all these domains will go a long way in the implementation of scaled up operations.

For the future, reprocessing of zircaloy clad thorium rods from PHWRs, which require chop-leach dissolution treatment, and the processing of thorium, uranium and Plutonium bearing experimental fuels are some of the tasks that need attention. In the long range, better dissolution techniques for thorium oxide, and an assessment of the impact of the presence of ^{231}Pa in thorex HLW and ^{228}Th in the separated thorium product would greatly enhance the viability of the back end processes.

(vi) Fabrication of high density sintered ThO_2 pellets for the ThO_2 bundles used for flux flattening of the initial core of PHWR is carried out by the conventional powder metallurgy technique of cold-compaction and high temperature sintering either under reducing or oxidizing atmosphere. ThO_2, being a perfectly stoichiometric compound, with a high melting point (-3400 °C) needs a sintering temperature of > 1800 °C for obtaining high density (>96%T.D.). Addition of 500-600 ppm of MgO can lower this temperature to 1650-1680 °C.

The following techniques have been tried for thorium based fuels: (a) Cold pressing of powder mixture of (Th, Pu)O_2 or (Th/^{233}U)O_2 followed by high temperature sintering, (b) vacuum impregnation of partially sintered low density (~ 70 - 80 % T.D.) ThO_2 pellets with uranyl nitrate or plutonium nitrate solution followed by drying and final sinterning, (c) Sol-gel derived microsphere pelletisation followed by sintering. The sol-gel microsphere pelletization process (SGMP), utilizes sol-gel derived dust-free and free flowing soft microspheres of (Th, U)O_2 in the size range of 100-600 microns, which are cold compacted and sintered to high density pellets just the way powder pellets are fabricated.

(vii) Fuel irradiation program is carried out in Pressurised Water Loop (PWL) of the 40 MW(th) research reactor, CIRUS. To study the behaviour of (Pu. Th) oxide fuel under high power and high burnup conditions, a six-pin assembly of (Pu, Th) oxide pins was installed in the PWL. The pressure and temperature conditions in the PWL were similar to those in the PHWR power reactor. The cluster was irradiated to a burnup of 18,500 MW·d/t, and its performance was satisfactory [5].

Subsequently, another cluster, which is a mixed cluster of (Th, Pu) oxide pins and (U, Pu) MOX pins has been loaded into the PWL. This has already seen about 10,000 MW·d/t burnup. At the time of writing, it is still inside the loop.

(viii) Currently, India is working on the design of an Advanced Heavy Water Reactor (AHWR). This reactor is a pressure tube kind of reactor [6], which is specially designed with the thorium cycle in mind. ^{233}U enrichment in thorium has been

adjusted to be at the self-sustaining level. A discharge burnup of 20,000 MW·d/t is attained by using a certain amount of plutonium makeup. The plutonium is not mixed with the thorium, but is used in the form of (U, Pu) oxide pins. This has three advantages: (a) the plutonium pins can be placed wherever the spectrum is most advantageous to plutonium, (b) the discharge burnup of thorium pins can be adjusted independent of reactivity considerations, and (c) the thorium fuel remains uncontaminated by the long-lived actinides produced by the uranium cycle.

Since the thermal absorption of thorium is high as compared to uranium, the deleterious effects of parasitic absorption are less prominent in thorium systems. Thus it is possible to consider light water as coolant. The AHWR is cooled by boiling light water. The pressure tubes are vertical, and it is possible to have 100% heat removal by natural circulation, thus ensuring passive safety. The reactor has been designed to have negative void coefficient of reactivity.

4. FUEL CYCLE STUDIES IN THE PHWR

The most important thorium cycle is the self-sustaining equilibrium cycle (SSET). The salient features of our findings on this cycle were reported in the previous AGM and have been included in the report which is being presented to this AGM as working material [7]. As such, we do not repeat it here.

Next in importance is the thorium cycle without reprocessing. Figure 1 shows the findings from some studies carried out for a cycle in which thorium in combination with plutonium was burnt in a reactor to very high discharge irradiations. The plutonium considered was of an isotopic purity corresponding to the fuel discharged from the PHWR, and was about 75% fissile. Shown in this figure as a function of the discharge burnup of the fuel are two parameters. One is the fissile inventory ratio, which is the ratio between the fissile contents of the discharged fuel and the initial fuel. The other one we have called the fissile plutonium ratio, and it is the ratio of the fissile plutonium contained in the discharged fuel to that of the initial fuel. What is noteworthy here is the practically complete burning of the plutonium for even moderately high discharge burnups.

Figure 2 depicts a similar cycle with higher grade plutonium. This is almost pure ^{239}Pu, 96% fissile. Though we are not in a position to adopt this cycle, it has been analysed because of the current international interest in disposing of the plutonium that has become available from dismantled nuclear weapons. The independent variable is once again the discharge burnup of the thorium-plutonium fuel. Three quantities are plotted in this figure. One is the fissile plutonium in the initial fuel. Second is the fissile plutonium in the spent fuel, and the third is the fissile uranium, mainly ^{233}U, in the spent fuel. Once again the excellent efficacy of the cycle in burning plutonium is clear. A modest amount of ^{233}U gets built up.

Mixing plutonium with thorium contaminates the thorium with the long lived actinides of the uranium cycle, and thus damages one of the attractive features of the thorium cycle. In this context, Milgram's [8] once through thorium (OTT) cycle is of interest, although Milgram originally proposed it as an incentive for reluctant thorium users. This cycle keeps the uranium and thorium fuels apart, offers a measure of flexibility in that one can decide to start or discontinue the loading of thorium fuels at almost any time, permits the discharged thorium to be stored pending decisions about reprocessing or disposal, and does all this without any penalty in terms of either cost or fuel utilization in the event a decision is taken not to reprocess the thorium to recover ^{233}U. If extracted, the ^{233}U is thus obtained as a bonus.

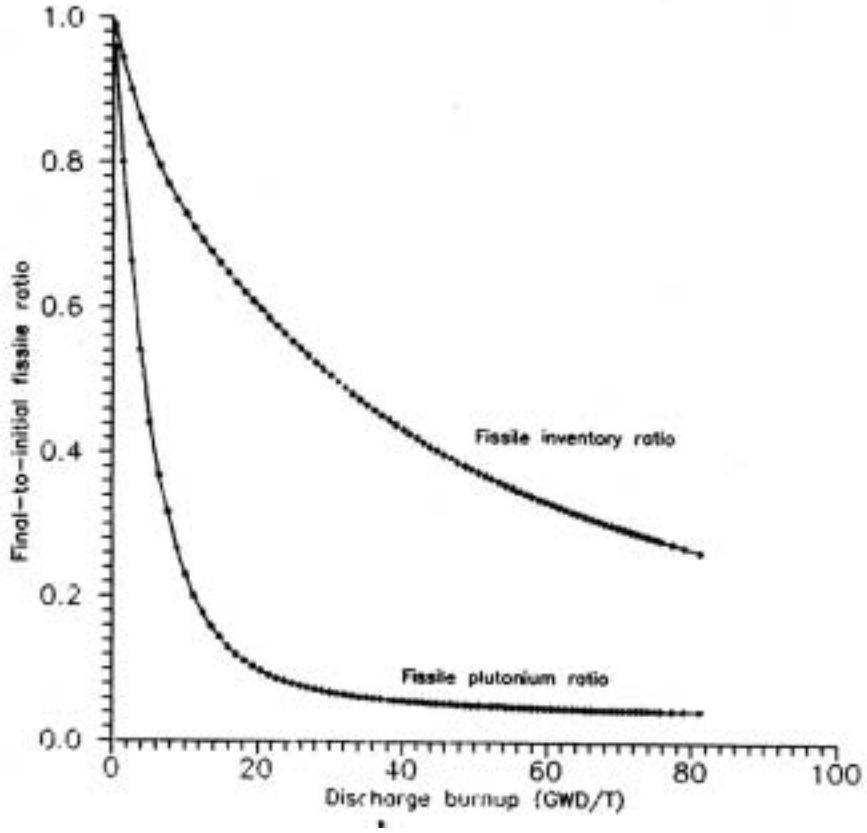

Figure 1. Thorium-plutonium in PHWR without reprocessing.

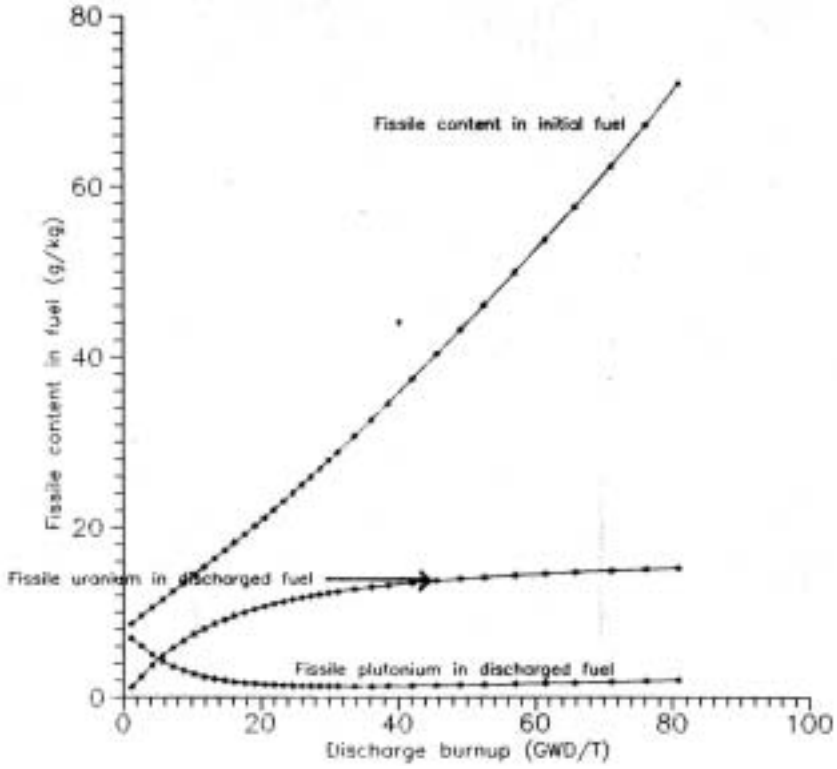

Figure 2. Fissile inventories in the thorium-plutonium cycle without reprocessing.

Figure 3 shows the essential feature of the OTT cycle. The central idea is to fuel the core with a combined loading of uranium fuel and plain thorium fuel. The thorium will act as a load on the uranium and decrease its discharge burnup. But as the thorium resides in the neutron flux, it will build up ^{233}U and begin to produce energy by ^{233}U fission. As the residence time of thorium increases, so will the penalty suffered by the burnup of uranium. However, if we express the energy obtained from a unit of uranium mined by taking credit for the power produced in thorium as well, we get the curve of Figure 3. As the residence time of the thorium increases, the energy from uranium mined decreases at first, but with growing production of energy from ^{233}U fission, the curve turns upward and climbs until it overtakes the value corresponding to the uranium fuel without thorium. It continues to climb, but finally turns downward again due to the accumulated fission products acting as a load. The figure also shows the discharge burnup of the uranium fuel, which decreases all the way. The gain in this cycle also depends upon the level of enrichment of the uranium fuel, the feed ratio of thorium to uranium fuel, and the flux level in the system.

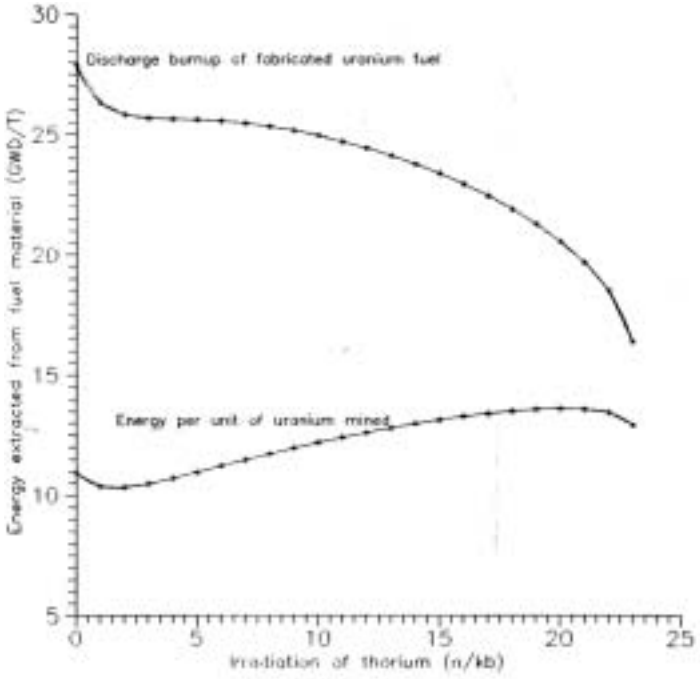

Figure 3. Energy from uranium mined in the OTT cycle.

Figure 4 shows a plot of the energy from mined uranium as a function of the enrichment of the uranium fuel. The cycle works best for low enrichments. Figure 5 shows the same quantity as a function of feed ratio of thorium fuel to uranium fuel. It would appear that a feed ratio in the vicinity of about 10% is most suitable. Figure 6 shows the sensitivity of the energy from uranium mined to the flux level at which the thorium fuel is operated.

An interesting extension of the OTT would be to follow the same kind of segregated loading as the OTT does, but at the same time, reprocess the discharged thorium to recover ^{233}U and feed the thorium fuel in the form of thorium-^{233}U fuel, keeping an enrichment level that would be self-sustaining in ^{233}U. Figure 7 shows the energy extracted as a function of the thorium-to-uranium feed ratio. This quantity increases as the feed ratio increases. For high feed ratios, the major part of the energy is actually coming from the thorium fuel.

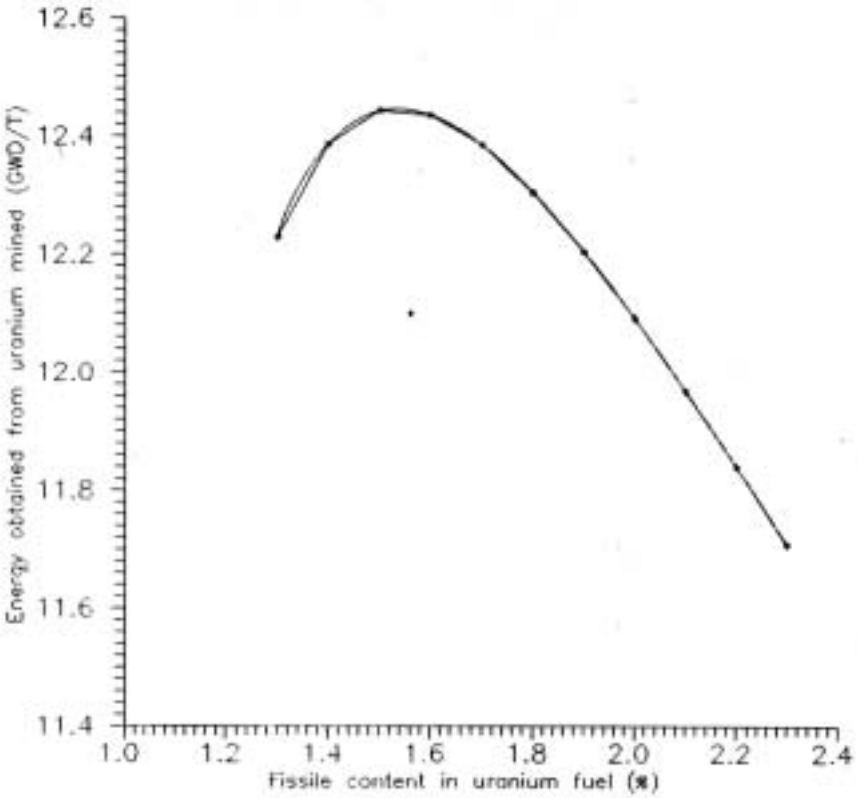

Figure 4. Effect of enrichment on the efficiency of the OTT cycle.

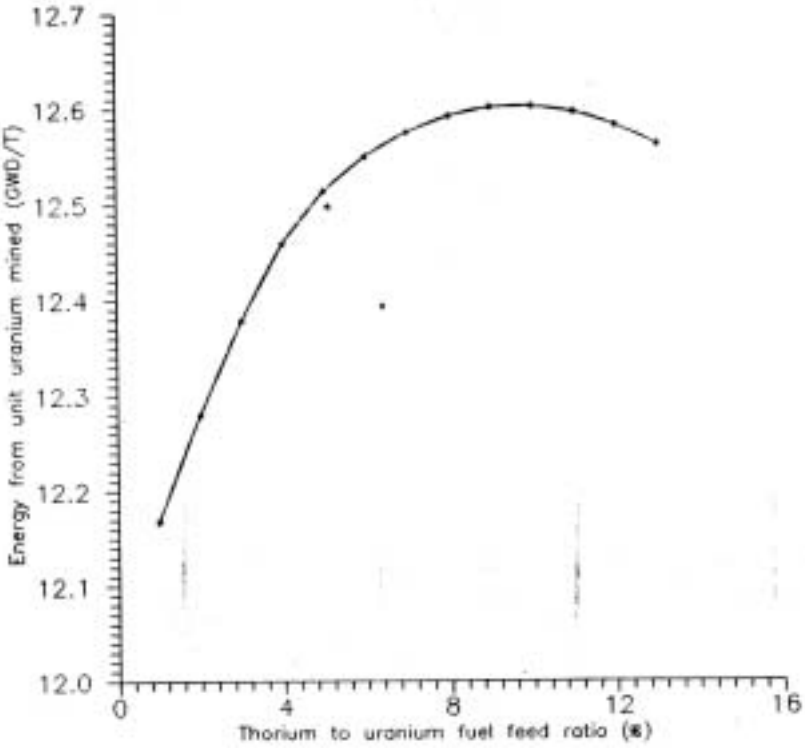

Figure 5. Dependence of fuel utilization on thorium-to-uranium feed ratio in the OTT cycle.

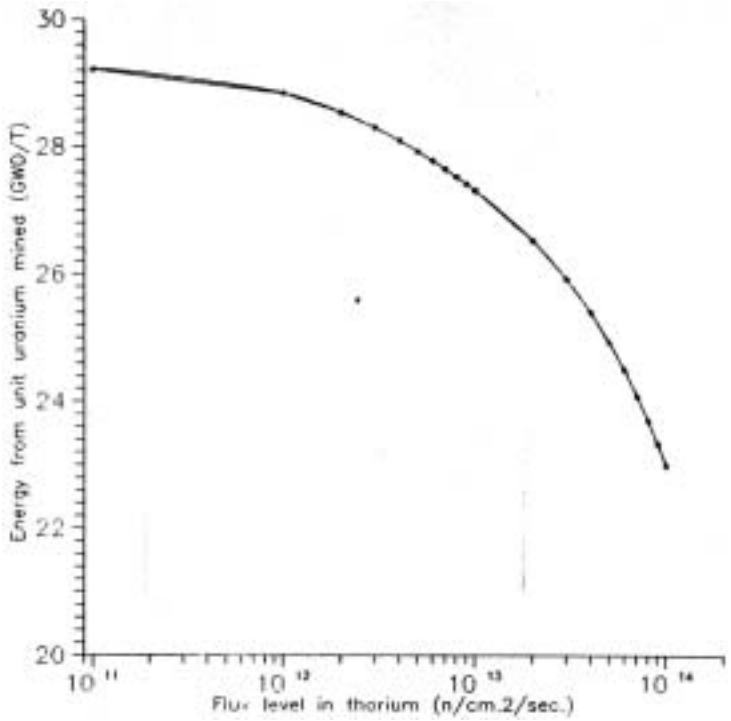

Figure 6. Influence of flux level in thorium on the fuel utilization in the OTT cycle.

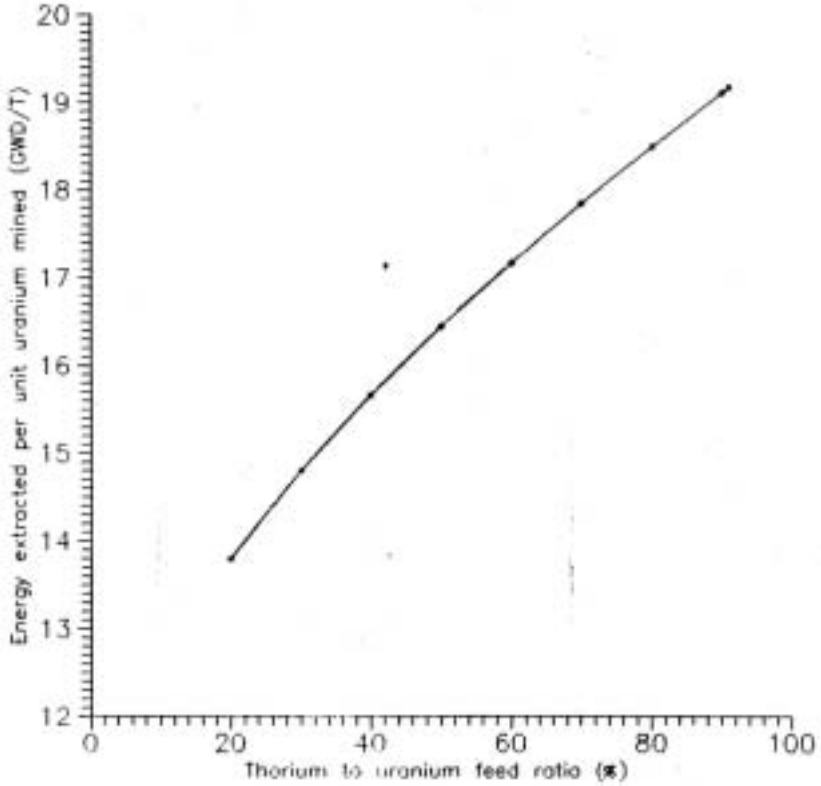

Figure 7. Segregated loading of uranium and thorium fuels with reprocessing of thorium and recycling of ^{233}U.

5. CONCLUSIONS

The versatile nature of thorium makes the thorium cycle very suitable for retrofitting into existing reactor designs. The most attractive feature of thorium lies in the nuclear properties of ^{233}U. Unlike ^{235}U and ^{239}Pu, the reactor physics performance characteristic of ^{233}U is almost spectrum independent. This property of being able to perform well in any spectrum gives thorium fuels great flexibility in the context of reactor systems designed for the uranium cycle.

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THORIUM FUEL FOR LWRs AND INNOVATIVE CONCEPTS: STATE OF THE ART AT THE CEA/DRN*

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Abstract. This paper briefly summarizes the state of the art on the study of thorium at the CEA and its projected use in pressurized water reactors. The advantages and drawbacks of thorium for the nuclear cycle are examined. Studies carried out in France are reviewed, major findings are recalled and possible future orientations for inert matrix fuels are presented.

1. Introduction

Viewed within the context of a fast-growing world population, which will have progressed from 2.8 billion individuals in 1970 to a projected 9 billion in the year 2020 (×3), primary energy needs are expected to increase at a steady rate, from 5 billion epts in 1970 to more than 16 billion epts in the year 2000 (estimate) (×3.4). The share of electricity in this demand is great. Thus, in France, electricity consumption rose from 120 to 410 TW·h (480 TW·h production) (×3.4) in 1995, while primary energy needs increased from 150 to 230 Mepts (×1.5). Limited fossil fuel resources [1] (proven retrievable reserves amount to 68 years for gas, 45 years for oil, and 250 years for coal), the need to reduce carbon dioxide emissions (23 million tons in 1996), the need for both stable and varied sources of supply as well as an efficient, qualified, and reliable industry, together with profitable investments, all plead in favour of fission energy.

However, in the once-through cycle, reserves may very quickly come close to exhaustion, contrary to the widespread notion that cheap and unlimited uranium supplies will always exist. A certain number of solutions may be put forward to attempt to prolong the life span of fission energy :

– The reprocessing and recycling of material in existing reactors to favour the use of plutonium, reprocessed uranium, depleted uranium, and actinides. Certain solutions aiming at optimal consumption of civilian and military plutonium are already well underway [2–6]. In this way, the closed cycle would lead to an increase in the utilization of natural uranium by a factor of about 10 ;

– With our present understanding of the deficiencies of early reactors, efficient and reliable fast reactors would lead to an increase in the utilization of natural uranium by a factor of 50;

* 1998 meeting.

– The thorium cycles represent a formidable asset by increasing the use of natural resources for the purpose of prolonging the life span of nuclear energy;
– Optimal appropriate management of the various reactor types and fuels should permit increasing the intrinsic efficiency of nuclear fuel.

2. THE ADVANTAGES OF THORIUM

The half life of this element is 14 billion years, that is to say three times that of uranium-238. It is likely to exist in much greater abundance than uranium. The fissile isotope used in this cycle (^{233}U) has the best production/neutron absorption rate in the thermal spectrum (2.30 compared to 2.077 for ^{235}U). Minor actinide production is more limited than in the uranium cycle, which, in the short term, results in lower radiotoxicity, which does not appear to be a determining factor after 1000 years.

The melting temperature of the ThO_2 oxide is very high (3300°C in comparison to 2700°C for UO_2). This permits much higher powers and burnups given the values of the thermal conductivity which are very close to those of UO_2 (Table 1). An in-reactor qualification of the ThO_2 matrix exists and does not pose any particular problem. The resistance to proliferation seems slightly better than in the UO_2 cycle, for it is above all linked to the difficulty of reprocessing and to the activity of ^{232}U and its decay products. Finally, it should be pointed out that recently there has been renewed interest in thorium for hybrid systems.

Table I. Parameters of fissile and fertile nuclei.

Table Ia. Physical properties of the elements U, Pu and Th (metal and oxide).

.Properties	U	UO_2	Pu	PuO_2	Th	ThO_2
Melting point (°C)	1130	2760	632	2400	1750	3300
Phase change (°C)	660				1400	
Theoretical density (g/cm3)	18.9	10.96	19.8	11.50	11.7	10.00
Thermal conductivity (600°C) W/cm/°C	0.42	0.0452			0.45	0.044

Table Ib. Mean fissile nuclei parameters in a thermal neutron spectrum.

	^{233}U	^{235}U	^{239}Pu	^{241}Pu
σ capture (barns)	46	101	271	368
σ fission (barns)	525	577	742	1007
$\alpha = \sigma c/\sigma f$	0.088	0.175	0.365	0.365
$\eta = \nu\sigma f/\sigma a$	2.300	2.077	2.109	2.151
Eff. β fact. (pcm*)	270	650	210	490

* per cent of milli K (10^{-5} Δk/k).

Table Ic. Parameters of fertile nuclei.

	^{232}Th	^{238}U
σ capture (barns)	7.40	2.73
I.R.* capture (barns)	85	272
Fission cutoff (MeV)	1.5	0.8
Effective β factor (fast fission)	2030	1480

* I.R. : integral resonance in infinite dilution (0.625 eV at 20 MeV).

3. THE DRAWBACKS OF THORIUM

Thorium possesses no fissile isotopes and it is therefore necessary to trigger the cycle by using classic uranium or plutonium isotopes. Conversion by reaction (n, 2n) giving ^{232}U and by decay very energetic gamma emitters. However, it should be noted that this negative point becomes a positive one when considered from the « resistance-to-proliferation » standpoint. The proportion of delayed neutrons is low for ^{233}U : 270 pcm as against 670 for ^{235}U (Table I), which places this fuel on the level of the Pu factor for this type of parameter, and thus results in a certain penalty for accidents of the rapid reactivity insertion type. The accumulation of absorbent ^{233}Pa, parent of ^{233}U, leads to shutdown control problems, requiring over sizing of anti-reactive systems. Neutron capture of ^{232}Th is three times higher than that of ^{238}U, which ensures a faster conversion than in the ^{238}U-^{239}U cycle, but necessitates higher fissile igniters to trigger the first cycle. Reprocessing is delicate (but this is a positive point with respect to resistance to proliferation), ThO_2 is insoluble in nitric acid and the THOREX process uses highly corrosive products.

4. THORIUM RESOURCES

Outside the nuclear industry, a few uses for alloys in the field of aeronautics and for refractory materials have led to the production of a few hundred tons of thorium per year. However, this figure can be pushed up to about 2000 t/year as the ore (monazite) is available. The resources provided reasonably, outside of Russia and China, at a price lower than 70 US dollars are about 1.7 million tons and estimated additional resources are as much as 3 million tons. The principle ore, monazite, a thorium, lanthanide, and cerium phosphate, contains 3 to 9% Th. The main producer is Australia with about 14500 t per year.

5. RESEARCH AND DEVELOPMENT IN FRANCE

From 1969 to 1980, theoretical neutron studies were carried out in collaboration with EDF, followed by integral experiments performed to establish basic nuclear data. Then, in collaboration with NOVATOME, RHTF2 project studies on a Fort St. Vrain type HTR reactor were made using thorium carbide particles. This project was abandoned in 1978. Some studies on dissolved salt reactors, using thorium dissolved salt, were performed in collaboration with Pechiney. 2000 tons were produced in the pilot factory of Le Bouchet using monazite from Madagascar. This production was sold to the United States.

During the nineties, various studies were performed in the framework of waste elimination strategy:

- PWR and FBR analyses with an aim to replace ^{238}U by ^{232}Th so as reduce the production of minor actinides;
- Studies in the framework of the fourth ERDP (European R&D Program) on thorium use for the CAPRA project [7,8], aiming for better actinide incineration;
- PWR research with a view to assess the potential of thorium fuels, in strategies of natural resource protection, more efficient use of plutonium, and better waste management.

Interest in thorium grew with the appearance of hybrid reactor concepts as well as of new requirements as regards the validation of basic data (U, Th, Pa, etc.). Work has been initiated in order to perform a certain number of sub-critical experiments in the MASURCA reactor, in which 1.2 tons of ThO_2 in the form of rods will be used in the MUSE-4 tests during 1998. Finally, a last point, which will be further discussed below, concerns the role of thorium in the

use of fuels in inert matrixes (IMF), with the aim, in the case of uranium free plutonium, of improving the kinetic coefficients of the array.

6. PWR RESEARCH

Previous studies [9,10] concerned a 900 MW(e) PWR with third-core reload fuel management in an annual cycle of 287 EFPD with an average burnup of 33 GW·d/t. Five types of fuel were analysed :

^{232}Th + ^{235}U highly enriched uranium (HEU),

^{232}Th + commercial first generation plutonium,

^{232}Th + uranium with a high ^{233}U content (Ut), ^{233}U comes from the reprocessing of the fuel mentioned above,

^{232}Th + ^{235}U medium enriched uranium (MEU) ; denatured cycle, non-proliferating (^{235}U < 20 %)

^{232}Th + weapon grade plutonium.

Table II summarizes the characteristic differences between these fuels.

Table II. Some properties of studied fuel unloaded at 33 GW·d/t.

Fuel type	HEU/Th	RG Pu/Th	UT/Th	MEU/Th	WG Pu/Th	MOX standard	UO_2
Initial content fertile material	96.1 (Th)	93.5 (Th)	96.93 (Th)	81.0 (Th)	95.0 (Th)	94.7 (U)	
Initial content fissile material	3.9 (U)	6.5 (Pu+241Am)	3.07 (U)	19.0 (U)	5.0 (Pu)	5.3 (Pu+^{241}Am)	100 (U)
Initial content fissile nuclides	3.63 (^{235}U)	4.55 (^{239}Pu+^{241}Pa)	2.93 (^{233}U+^{235}U)	3.80 (^{235}U)	4.73 (^{239}Pu+^{241}Pu)	3.71 (^{239}Pu+^{241}Pu)	3.25 (^{235}U)
Fissile material consumption							
(Kg/TW·h(e))	30.3	113.7	18.5	62.2	123.7	52.5	90
(%)	20.7	47.1	12.2	8.7	65.8	26.1	73
Minor Actinides production							
(Kg/TW·h(e))	1.90	7.13	0.50	2.14	2.98	13.96	3.34
(% of the burned material)	6.3	6.3	2.7	3.4	2.4	26.6	3.7
Global convers. factor	0.621	0.631	0.759	0.680	0.497	0.590	0.475

RG Reactor Grade
WG Weapon Grade

Tables III-VII present the mass balances for beginning of life, 33 GW·d/t and 60 GW·d/t. Burnable poisons are not required insofar as the cycles envisaged (12 months, third-core reloading, and 33 GW·d/t) are short. Table II shows that the Ut/Th fuel represents the lowest consumption of fissile material (18.4 %), while producing the smallest amount of minor actinides, and its conversion factor is the highest of all the fuels analysed. These excellent results, however, are counterbalanced by the fact that this fuel is derived from the reprocessing of Pu/Th fuel, which is the greatest consumer of fissile material (advantageous in a strategy of plutonium stocks reduction), as well as a not negligible producer of minor actinides. This cycle is particularly interesting from the standpoint of natural resource protection as a result of its considerable production of uranium with a high (91%) 233U content.

Table III. HEU/Th fuel. Heavy nuclides inventory at BOL and EOL for a 900 MW(e) PWR (equilibrium 52 assemblies).

BU (GW·d/t)	0		33			60	
Nucleus	Masses (kg)	Isotopes (%)	Masses (kg)	Isotopes (%)	Balance (kg)	Masses (kg)	Isotopes (%)
228Th	0		0.0067			0.0203	
230Th	0		8.84×10-5			3.20×10-4	
232Th	22 858	96.10	22 206	93.36	–652.0	21 506	
Total Th	22 858	96.10	22 206	93.36	–652.0	21 506.020	90.41
231Pa	0		1.720			2.032	
233Pa	0		31.155	0.13		34.603	0.14
Total Pa	0		32.875	0.14	+32.875	36.635	0.16
232U	0		1.181	0.		2.490	0.
233U	0		292.500	1.23		343.007	1.44
234U	0		39.832	0.17		85.591	0.36
235U	863	3.63	211.680	0.89		63.881	0.27
236U	0		105.120	0.44		114.286	0.48
237U	0		0.266	0.		0.307	0.
238U	65	0.27	54.420	0.23		45.788	0.19
Total U	928	3.90	704.999	2.96	–223	655.350	2.76
237Np	0		10.156			18.136	
238Np	0		0.036			0.072	
239Np	0		0.041			0.039	
Total Np	0		10.233	0.04	+10.233	18.247	0.08
236Pu	0		0	0.		1.7×10-4	
238Pu	0		2.972	0.		9.651	0.04
239Pu	0		2.311	0.		3.139	0.01
240Pu	0		0.686	0.		0.985	
241Pu	0		0.778	0.		1.086	
242Pu	0		0.350	0.		0.934	
Total Pu	0		7.097	0.03	+7.097	15.795	0.07
241Am	0		0.019			0.027	
242Am	0		3.56×10-4			5.23×10-4	
243Am	0		0.067			0.324	
Total Am	0		0.086	0	+0.086	0.351	0
242Cm	0		0.008			0.018	0
243Cm	0		1.75×10-4			6.91×10-4	
244Cm	0		0.002			0.195	
245Cm	0		9.3×10-4			0.014	
Total Cm	0		0.010	0	+0.010	0.227	0
TOTAL	23 786	100	22 961	96.53	–825	22 233	93.47

Table IV. - UT/Th (Rmod = 1.3) fuel. Heavy nuclides inventory at BOL and EOL for a 900 MW(e) PWR (equilibrium 52 assemblies).

BU (GW·d/t)	0		33			60	
Nucleus	Masses (kg)	Isotopes (%)	Masses (kg)	Isotopes (%)	Balance (kg)	Masses (kg)	Isotopes (%)
228Th	0		0.0029			0.0408	
230Th	0		5.76×10-4			0.0010	
232Th	30 347		29 383			28 482	
Total Th	30 347	96.93	29 383	93.85	–964	28 482	90.97
231Pa	0		2.615	0.01		3.022	0.01
233Pa	0		44.901	0.14		46.889	0.15
Total Pa	0		47.516	0.15	+47.516	49.911	0.16
232U	3.070	0.01	3.175	0.01		4.306	0.01
233U	880.530	2.81	591.036	1.89		529.124	1.69
234U	67.570	0.21	158.917	0.51		200.999	0.64
235U	9.990	0.03	39.799	0.13		57.988	0.18
236U	0		6.324	0.02		16.592	0.05
237U	0		0.022	0.		0.058	0.
238U	0		0	0.		0.	0.
Total U	961.160	3.07	799.273	2.55	–161.887	809.067	2.58
237Np	0		0.551	0.		2.153	0.01
238Np	0		0.002	0.		0.009	0.
239Np	0		0	0.		0.	0.
Total Np	0		0.553	0.	+0.553	2.162	0.01
236Pu	0		3.04×10-6	0.		0.	0.
238Pu	0		0.142	0.		0.939	0.
239Pu	0		0.014	0.		0.121	0.
240Pu	0		0.002	0.		0.026	0.
241Pu	0		0.002	0.		0.028	0.
242Pu	0		3.05×10-4	0.		0.009	0.
Total Pu	0		0.161	0.	+0.161	1.123	0
241Am	0		2.17×10-5	-		4.87×10-4	0.
242Am	0		4.00×10-7	-		1.01×10-5	0.
243Am	0		3.76×10-5	-		0.0021	0.
Total Am	0		5.98×10-5	-		0.0026	0.
242Cm	0		5.53×10-5	-		2.06×10-4	0.
243Cm	0		8.20×10-8	-		5.33×10-6	0.
244Cm	0		6.60×10-6	-		7.64×10-4	0.
245Cm	0		3.15×10-7	-		5.79×10-5	0.
Total Cm	0		1.25×10-5	-		0.001	0.
TOTAL	31 308	100	30 230	96.56	–1 078	29 344	93.73

Table V. - Pu/Th fuel. Heavy nuclides inventory at BOL and EOL for a 900 MW(e) PWR (equilibrium 52 assemblies).

BU (GW·d/t)	0		33			60	
Nucleus	Masses (kg)	Isotopes (%)	Masses (kg)	Isotopes (%)	Balance (kg)	Masses (kg)	Isotopes (%)
228Th	0		0.0057			0.022	
230Th	0		5.05×10-5			2.05×10-4	
232Th	22 277		21 828			21 308.088	
Total Th	22 277	93.50	21 828.006	91.61	–449.	21 308.110	89.43
231Pa	0		2.698	0.01		3.299	0.01
233Pa	0		20.829	0.09		25.603	0.11
Total Pa	0		23.527	0.10	+23.527	28.902	0.12
232U	0		1.032	0.		2.718	0.01
233U	0		271.700	1.14		370.254	1.55
234U	0		22.440	0.09		56.221	0.23
235U	0		3.327	0.01		12.394	0.05
236U	0		0.216	0.		1.350	0.
237U	0		5.34×10-4	0.		0.003	0.
238U	0		3.53×10-4	0.		7.06×10-4	0.
Total U	0		298.714	1.25	+298.714	442.941	1.86
237Np	0		0.065	0.		0.160	0.
238Np	0		1.50×10-4	0.		4.68×10-4	0.
239Np	0		2.31×10-5	0.		3.69×10-5	0.
Total Np	0		0.065	0.	+0.065	0.160	0.
236Pu	0		0	0.		0.	0.
238Pu	20.754	0.09	25.954	0.11		26.651	0.11
239Pu	888.552	3.73	202.738	0.85		24.914	0.10
240Pu	351.114	1.47	287.383	1.21		129.951	0.54
241Pu	195.305	0.82	183.870	0.77		98.864	0.41
242Pu	73.259	0.31	109.153	0.46		139.336	0.58
Total Pu	1528.984	6.42	808.098	3.39	–720.884	419.716	1.76
241Am	19.670	0.08	16.701	0.07		8.451	0.03
242Am	0		0.461	0.		0.206	0.
243Am	0		26.613	0.11		42.175	0.18
Total Am	19.670	0.08	43.775	0.18	+24.105	50.832	0.21
242Cm	0		4.887	0.02		4.537	0.02
243Cm	0		0.187	0.		0.258	0.
244Cm	0		12.056	0.05		32.382	0.13
245Cm	0		1.189	0.		3.784	0.01
Total Cm	0		18.319	0.08	+18.319	40.961	0.17
TOTAL	23 826	100	23.020	96.62	–806	22 292	93.56

Table VI. - MEU/Th fuel. Heavy nuclides inventory at BOL and EOL for a 900 MW(e) PWR (equilibrium 52 assemblies).

BU (GW·d/t)	0		33			60	
Nucleus	Masses (kg)	Isotopes (%)	Masses (kg)	Isotopes (%)	Balance (kg)	Masses (kg)	Isotopes (%)
228Th	0		0.0056			0.0182	
230Th	0		6.87×10-5			2.50×10-4	
232Th	19 387		18 865			18 328	
Total Th	19 387	81.0	18 865	78.81	–522	18 328	76.57
231Pa	0		1.667	0.01		2.137	0.01
233Pa	0		23.615	0.10		25.548	0.11
Total Pa	0		25.282	0.10	+25.282	27.685	0.11
232U	0		0.968	0.		2.173	0.01
233U	0		259.334	1.08		321.114	1.34
234U	0		30.039	0.12		64.735	0.27
235U	909.538	3.80	281.962	1.18		104.254	0.43
236U			103.792	0.43		119.459	0.50
237U			0.241	0.		0.288	0.
238U	3 638.141	15.20	3 453.334	14.43		3 276.796	13.69
Total U	4 547.141	19.0	4 129.671	17.25	–417.470	3 888.819	16.25
237Np	0		9.763	0.04		18.572	0.08
238Np	0		0.029	0.		0.061	0.
239Np	0		0.725	0.		0.767	0.
Total Np	0		10.578	0.04	+10.518	19.400	0.08
236Pu	0		6.23×10-5	0.		2.00×10-4	0.
238Pu	0		2.685	0.01		9.515	0.04
239Pu	0		44.138	0.18		44.243	0.18
240Pu	0		14.616	0.06		18.037	0.07
241Pu	0		12.509	0.05		16.476	0.07
242Pu	0		4.169	0.02		11.723	0.05
Total Pu	0		78.117	0.33	+78.117	99.994	0.42
241Am	0		0.327	0.		0.563	0.
242Am	0		0.006	0.		0.011	0.
243Am	0		0.727	0.		3.627	0.01
Total Am	0		1.060	0.	+1.060	4.201	0.02
242Cm	0		0.107	0.		0.286	0.
243Cm	0		0.002	0.		0.010	0.
244Cm	0		0.186	0.		1.931	0.01
245Cm	0		0.010	0.		0.151	0.
Total Cm	0		0.305	0.	+0.305	2.378	0.01
TOTAL	23 935	100	23 110	96.55	–825	22 370	93.46

Table VII. - Weapon grade Pu fuel/Th. Heavy nuclides inventory at BOL and EOL for a 900 MW(e) PWR (equilibrium 52 assemblies).

BU (GW·d/t)	0		33			60	
Nucleus	Masses (kg)	Isotopes (%)	Masses (kg)	Isotopes (%)	Balance (kg)	Masses (kg)	Isotopes (%)
228Th							
230Th							
232Th	22 649						
Total Th	22 649	95.0	22 233	93.25	–416	21 623	90.70
231Pa			2.78	0.01		2.65	0.01
233Pa			24.68	0.10		27.24	0.11
Total Pa	0.		27.46	0.11	+27.46	29.89	0.12
232U			1.48	0.		2.74	0.01
233U			310.33	1.30		359.51	1.51
234U			33.52	0.14		63.40	0.26
235U			5.82	0.02		13.95	0.06
236U			0.	0.			
237U			0.	0.			
238U			0.	0.			
Total U	0.		349.95	1.46	+349.95	439.12	1.84
237Np							
238Np							
239Np							
Total Np	0.		0.02	0.		0.13	0.
236Pu				0.			
238Pu			1.76	0.01		4.17	0.02
239Pu	1 121	4.70	122.50	0.51		11.95	0.05
240Pu	64	0.27	153.42	0.64		60.53	0.25
241Pu	7	0.03	96.95	0.41		56.05	0.23
242Pu			33.40	0.14		56.65	0.24
Total Pu	1 192	5.0	408.03	1.71	–783.97	189.35	0.79
241Am			4.46	0.02		3.13	0.1
242Am			0.10	0.		0.07	0.
243Am			7.29	0.03		16.74	0.07
Total Am	0.		11.85	0.05	+11.85	19.94	0.08
242Cm			1.41	0.		1.95	0.01
243Cm			0.04	0.		0.08	0.
244Cm			2.58	0.01		9.69	0.04
245Cm			0.20	0.		0.87	0.
Total Cm	0.		4.23	0.06	+4.23	12.59	0.05
TOTAL	23 841	100	23 034	96.62	–807	22 314	93.59

Finally, it should be noted that thorium matrix fuel is a good candidate for using weapon grade Pu, in view of its significant burnup of 124 kg/TW·h(e), and that, at the end of the cycle, MEU/Th fuel has a high fissile nuclei content, which justifies reprocessing of very long cycles with an aim to saving resources. In addition, a large bibliographical study on the thorium cycle in a PWR has been performed [11].

7. INERT MATRIX FUEL

Table VIII shows the different neutron coefficient values for the five fuels of interest compared to UO2, in a core composed of homogeneous fuel assemblies.

The effect of the resonant thorium can be observed on coefficients sensitive to the neutron spectrum. Thorium thus appears to be a stabilizing factor for the kinetic coefficients, even in the case of Pu/Th in which the Pu isotopes strongly influence the values. Thorium appears as a tendency moderator, with, however, some highly disadvantageous factors, such as the very negative moderator coefficient, which makes steam line break accidents very deleterious, and a very weak effective beta, which makes the consequences of rod drop accidents very difficult to control.

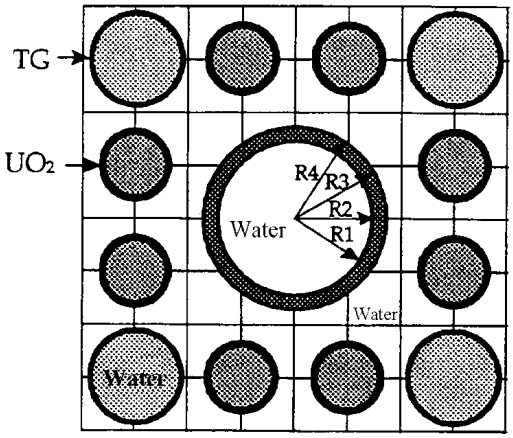
Table VIII. - Kinetic parameters of the fuel assembly. Beginning of lifetime, 600 ppm, Pn.

Fuel type	HEU/Th	Pu/Th	UT/Th (3)		MEU/Th	UO2
	(1)	(2)	a) mod.R=2	b) mod.R=1.3*	(4)	(3.7% 235U)
Soluble boron (pht/ppm)	8.84	3.55	8.90	5.17	9.43	8.9
Doppler (pht/°C) (650°-305°C)	–3.48	–3.29	–3.46	–4.40	–4.92	–2.6
Moderator coeff. (305°-285°C) pht/°C	–6.0	–22.20	+5.2	–5.7	–12.2	–15.4
Global draining (pht) (0-100% vacuum)	–105480	–68600	–97240	–95530	–97140	–70000
Effective beta factor						
0 GW·d/t	671	279	299	307	678	595
33 GW·d/t (pht)	435	357	319	327	444	522 (core)
Control rod worth Ag-In-Cd (pht)	36 980	23 790	33 430	33 210	36 340	34 000

higher fuel mass (thick fuel rods).

This is why, with a view to the use of inert matrixes to fabricate uranium free plutonium (IMF, or Inert Matrix Fuel), the solutions devised to use 100 % IMF homogeneous assemblies very quickly come up against the weakness of the Doppler coefficient, the risk of a positive draining coefficient, and, in general, highly deteriorated kinetic parameters [12, 14]. Solutions intended to use APA-type assemblies [2, 3, 4]. Figure 1 have demonstrated that the association of the uranium and plutonium fuel in an inert matrix, especially if the local moderating ratio near the Pu is well adjusted through the use of judicious geometry, is perfectly well-suited to an efficient use of plutonium in pressurized water reactors with acceptable kinetic coefficients vis-à-vis the usual safety criteria and correct core management.

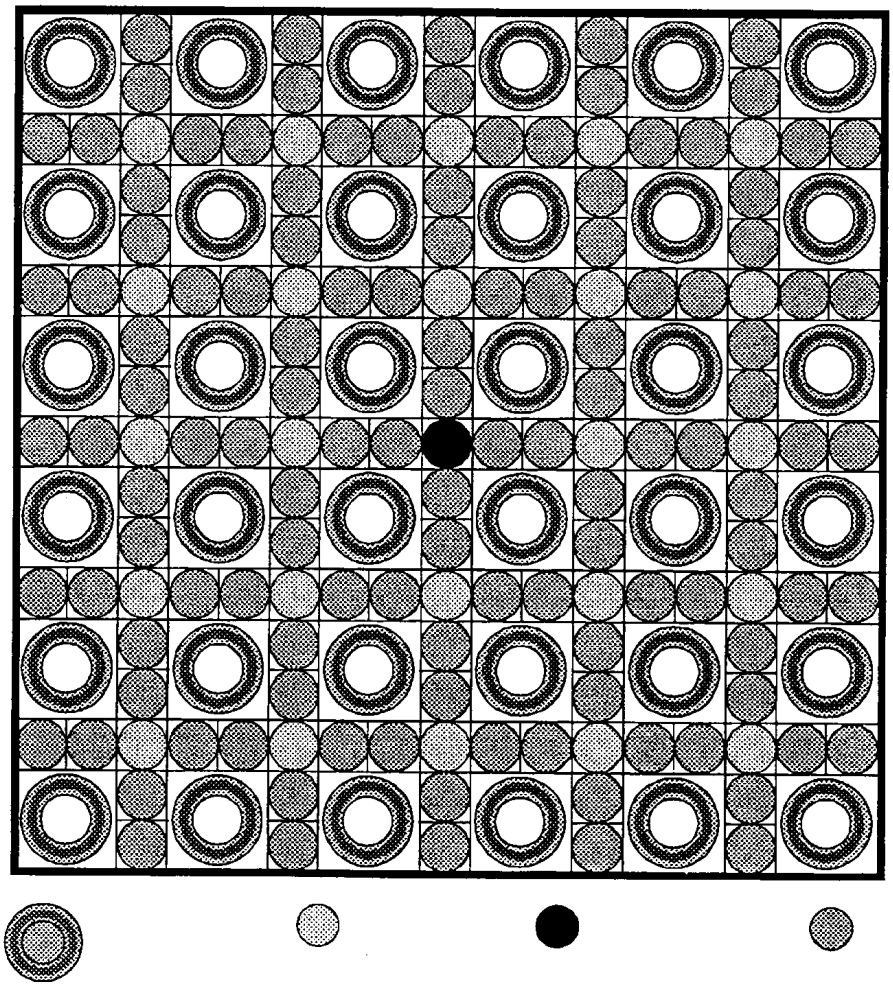
Geometric parameters



R4 (mm) = 11.5
R3 (mm) = 11.0
R2 (mm) = 9.739
R1 (mm) = 9.239
} 1.261 mm

VpuO2/Vcomb = 0.23

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



36 annular pins
24 guide tubes
1 instrumentation tube
120 regular UO2 pins

Figure 1. - Advanced Pu assembly.

Studies performed by Italian and Swiss partners [15, 16, 17] in the framework of the IMF workshop working group have also contributed to demonstrating that assemblies with a homogeneous standard geometry, but using 50 % UO2 rods and 50 % Pu/Th rods, would result in controllable cores in terms of both potential reactivity and kinetic coefficients. This has opened up a vast field of investigation into the optimization of the geometry and components of this type of fuel to improve, in turn, the various parameters which permit following a specific strategy: once-through, multiple recycling, reprocessing or no reprocessing, etc.

8. FABRICATION

Like uranium and plutonium fuels, thorium fuels, and more generally the thoria parts, are manufactured using dry process powder metallurgy techniques which permit obtaining dense parts with precise dimensions.

Natural ThO2 powder is first ground so as to increase its reactivity in sintering, and is then compacted and granulated to improve its flowability.

The blanks are then shaped, either by single-axis pressing for the pellets, or by isostatic cold compacting for the other more complex parts. The blanks are consolidated by sintering under thermodynamic conditions identical to those of standard UO2 fuel (four hours at 1700°C under hydrogen sweeping). The material obtained is dense and can reach 98% of theoretical density after this stage. Like uranium fuel intended for specific applications (advanced microstructures), the final material can be deliberately microcraked and its open porosity artificially increased by 30 % by the addition of organic products which will be eliminated during the sintering operation. The parts can be machined and ground using classic diamond grinding tools.

9. CONCLUSIONS

The study of the five types of fuel, together with the various strategies for the utilization of thorium as a plutonium support matrix with a view to the use of mixed UO2/Th+Pu assemblies clearly shows the interest of thorium for the improvement of core control, kinetic coefficients, and the use of natural resources. The various solutions proposed for both the fuels and the assemblies are very well-suited to diverse strategic alternatives, from the protection of natural resources to the use of 233U (Ut/Th) or plutonium (Pu/th). Thorium could be an excellent candidate to help define fuels able to mobilize weapons grade Pu, or to balance existing stocks, through the introduction of APA or IMF assemblies. All the operations of the fuel cycle - fabrication, reactor service life, reprocessing - call for a certain number of technologies that are known and have been tested in the laboratory. Industrial implementation would obviously require adaptation and optimization, but these would be based on knowledge and experience. In a reprocessing strategy, pilot plants using the Thorex process were able to operate, but were on the whole unwieldy. They could constitute a weak point resulting in the consideration of once-through cycles only, without reprocessing, but with very long cycles.

A certain number of basic data has to be updated to allow refining the calculations, particularly with respect to material balances after long cycles, and more detailed calculations must be made of transients and accidents linked to the nature of weak parameters, such as the effective beta.

A programme could be defined covering these fields of study with an aim to:

- Acquire or update basic data on certain Th and Pa nuclides;
- Validate kinetic coefficients and control efficiency through critical experiments;
- Perform technological irradiation and basic data (evolution) validation experiments for high burnups;
- Define appropriate arrays and heterogeneous assemblies for the various strategies of industrial interest envisaged to solve the diverse national problems that will be posed.

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SOME PECULIARITIES OF THORIUM DIOXIDE-BASE FUEL PRODUCTION AT NSC KIPT*

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Abstract. The paper describes the status of work on pyrocarbon-binder spherical fuel elements with thorium dioxide-base fuel for HTGR. Basic flow charts of manufacturing fuel microspheres, coated particles and spherical fuel elements are described. Results of investigations into the main characteristics of fuel elements and their constituents, including their operation under reactor irradiation conditions, are discussed. Some special features of the technology of pyrocarbon-bound $(U,Th)O_2$ - base spherical FE are presented and reports the results from studies of their main characteristics.

INTRODUCTION

Over a few decades, the development of high-temperature gas-cooled reactors (HTGR) has been an active field of research activities in nuclear power. These reactors are advantageously distinguished from reactors of other types by their ability to generate simultaneously both electrical and thermal energy, and also by high safety, an economical fuel cycle, a high efficiency (~ 40%), etc. The HTGR can be operated with both uranium and mixed U-Pu or U-Th fuel cycles or their combinations, and the uranium-thorium cycle appears to be most economical. The decisive argument in favour of the thorium cycle is the possibility of long-term providing the nuclear energetics on the whole with necessary breeding material. The combined use of uranium and thorium cycles assures the provision of fuel resources at moderate and stable costs.

In HTGR designs developed in the former Soviet Union and FRG, the use was made of the principle of pebble-bed core with graphite fuel elements, 60 mm in diameter, in the central part of which (kernel) the fuel is dispersed as coated particles (CP), while the periphery region (element jacket) is free of the fuel. The CP are the spherical particles of thorium and uranium carbides or oxides coated with protective layers of dense pyrocarbon and silicon carbide. The advantages of such fuel elements (FE) lie in their high radiation resistance and good fission product retention that enable the attainment of high nuclear fuel burnup levels (up to 100 000 MW¬24h/t and over) at a high operating temperature.

In the technology of spherical uranium-graphite fuel element production one can recognize the following three basic processes: (i) fuel microsphere (FM) production, (ii) coated fuel particle production, (iii) spherical FE production.

In the world practice there are three institutions (NUKEM - Germany, NSC KIPT - Ukraine [1], R&PA "Luch"- Russia [2]) known as the main developers of the spherical uranium-graphite FE processes. The first two processes are based on pressing the product billets by the known graphite-production electrode methods. The NSC KIPT technology has no foreign analogs as, instead of pressing, it makes use of the procedure of product billet molding followed by product densification with pyrocarbon deposited from gaseous hydrocarbons on

* 1998 meeting.

heated surfaces [1]. The NSC KIPT spherical FE have different design modifications, where fuel of different types (UO_2, $(U,Th)O_2$, UC, UCN, etc.) can be used.

FUEL MICROSPHERES

By now, a list of basic requirements on FM for spherical HTGR FE has been elaborated [3]. In particular, the FM must be uniform in size (500 - 50 mm), and their sphericity coefficient (d_{max}/d_{min}) should not exceed 1.05. The total free volume in both the FM and the buffer CP layer is in many respects responsible for the admissible fuel burnup and operating temperature of the fuel. It has been established by theory and experiment that the total porosity must be about 2 to 4 % per 1 % of heavy nuclei burnup at temperatures between 1300 and 1500 ^{o}C [4]. To provide the volume for collecting gaseous fission products (GFP) and solid fission products (SFP) in the FM, it appears more preferable, in our opinion, to follow the way of reducing the FM density rather than increasing the buffer CP layer thickness. Therefore, the FM density has been chosen to be about 85 % of theoretical density (TD), this is ensured at stages of "raw" billet manufacture and heat treatment of FM.

To make FM, the NSC KIPT team has developed the method of mechanical spheroidization of fuel billets made from plastified masses [5]. Though this method ranks below the sol-gel process in productivity, yet it is distinguished for its adaptability at the stage of experimental development of FM with different fuel compositions as the basis, and the process of FM production (to the sintering stage), as such, is wasteless.

The essence of the method consists in manufacturing and spinning cylindrical fuel billets from plastified masses until perfect microspheres are produced, which are then sintered at high temperature in vacuum.

Powders of uranium dioxide (enriched in uranium-235 up to 21, 36, 90 %) and thorium dioxide (natural) were used in experiments. The thorium-to-uranium ratios were 3:1 and 9:1.
The required powder mixture was prepared in the planetary-type centrifugal mill. Specially made metallic thorium balls, 12 mm in diameter, were used as mixing and milling elements.
The prepared powder mixture was mixed with 12 wt. % plasticizer based on paraffinum and petrolatum (65:35) at 70 ^{o}C in a mixer.

A special device was used to make from the plastified mass cylindrically-shaped sized billets with the H/D ratio of about 1.0 to 1.2. These billets were spinned in the "Spheroidizer" facility until spherical particles were produced. The nonsphericity of "raw" particles was about 1.02, the variation in size was less than 50 mm.

The heat treatment of the obtained fuel kernels was performed in two stages. At the first stage, the plasticizer was distilled off in vacuum. To retain the shape, the "raw" fuel kernels were heated in the aluminum oxide powder filling to a temperature of about 400 ^{o}C. At the second stage, the fuel kernels, separated from the filling, were sintered in the rotating container in a vacuum furnace at a temperature of 2000 ^{o}C. The density of microspherical fuel kernels after sintering was found to be 75-85 % TD. The fuel kernel $(Th,U)O_2$ structure is shown in Fig. 1. During the process of sintering, the chemical composition of mixed $(Th,U)O_2$ kernels remained the same.

Table I gives some characteristics of pilot batches of microspherical $(Th,U)O_2$ particles.

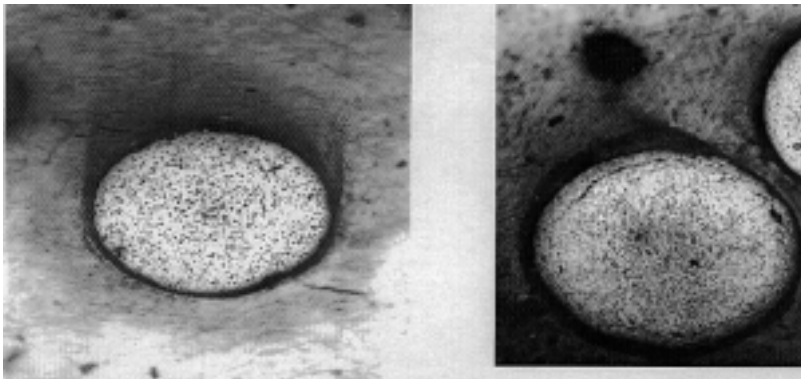

Figure. 1 Structure of fuel kernels (Th,U)O_2 (x 100).

Table I. Some characteristics of pilot batches of microspherical (Th,U)O_2 particles.

[1]	Batch number	Sintering conditions		Diameter, mkm	Density		Oxygen Coefficient
		Temper, K	Time, h		g/cm^3	% TD	
1.	69-36-1-84	2280	0,7	450-650	9,72	85	1,99
2.	70-21-1-87	2280	0,5	450-650	7,55	75	-
3.	01-90-1-87	2273	0,5	580-640	7,8	77	-
4.	02-90-1-87	2273	0,5	800-850	8,0	80	1,99
5.	03-90-1-87	2273	0,5	620-680	7,86	78	2,00

COATED FUEL PARTICLES

To deposit several protective coatings onto FM, the well-known "boiling layer" method is used at NSC KIPT. The technology devised to produce CP differs from analogs abroad by the type of gases used and by the conditions of protective layer deposition [6]. In particular, instead of inner and outer dense PyC layers, here the combined (PyC+SiC) coatings of ~2.4g/cm^3 are used. This substitution has allowed us to reduce the GFP yield from CP irradiated in the free-fill state by factors of 5 to 10.

As the process of CP production was developed, various CP designs were created:

type I - PyC-SiC-PyC,
type II - (PyC+SiC)-SiC-PyC,
type III - (PyC+SiC)-SiC-(PyC+SiC),
type II* (pilot) - (PyC+SiC)-SiC.

SPHERICAL FUEL ELEMENTS

To perform a combination of reactor tests of (Th,U)O_2 spherical FE, the last ones were fabricated by the process of volumetric gas-phase densification of porous bodies with pyrocarbon in the form of model spherical fuel elements (45 mm in diameter) [1].

Table II. Reactor test results for uranium-thorium fuel

№	CP batch number	Kernel material	Enrichment in ^{235}U	Kernel density g/cm^3	Kernel diameter, mkm	Coating material	Coating density, g/cm^3	Coating thickness (mkm)	Reactor, channel	Irradiation temperature (^oC)	Neutrons fluence E>0,1 MeV, x10^{20} cm^{-2}	Burnup, % fima	R/B in ^{88}Kr	R/B in ^{133}Xe
1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
1.	21-10-X-87	(3Th,1U)O_2	21	7,55	550± 100	PyC PyC PyC+SiC SiC	1,1 1,5 2,4 3,16	42 10 56 70	SM-2 №15	1250 1500	0,8 1,4	0 -2,1 2,1-3,5	8,1.10^{-6} 1,0.10^{-4}	1,1.10^{-5} 1,0.10^{-4}
2.	36-12-X-84	(3Th,1U)O_2	36	9,72	550± 100	PyC PyC PyC+SiC SiC PyC+SiC	1,1 1,5 2,4 3,18 2,4	35 7 70 140 120	MR-2 Karat 6	1100-1200 1350-1600 1400-1600	1,45 1,27 1,22	13,9 12,0 13,4		
3.	36-13-X-84	(3Th,1U)O_2	36	9,72	550± 100	PyC PyC PyC SiC PyC	1,1 1,5 1,8 3,18 1,8	35 7 60 100 70	RBT-6 № 2	1250	1,1	3,0	8,7.10^{-6}	1,42.10^{-5}
4.	* 21-10-X-87	(3Th,1U)O_2	21	7,55	550± 100	PyC PyC PyC+SiC SiC	1,1 1,5 2,4 3,16	42 10 56 70	SM-2 BKS-1	1250	2,1	0 - 4,0 5,0-8,0 8,0-9,8	1,2.10^{-6} 1,7.10^{-6} 1,0.10^{-4}	2,1.10^{-6} 3,3.10^{-6} 1,0.10^{-4}
5.	* 90T-1-X-88	(9Th,1U)O_2	90	7,8	610± 30	PyC PyC PyC+SiC SiC PyC+SiC	1,1 1,5 2,4 3,16 2,4	58 18 63 67 50	SM-2 № 13	1250		>3,0	0,9.10^{-6}	-

* model spherical fuel elements

REACTOR TEST RESULTS

The reactor test program for CP based on mixed $(Th,U)O_2$ fuel was a part of a wide-scale program of experimental development of uranium-graphite pyrocarbon-bound FE for HTGR. On performing reactor tests of CP and spherical FE with $(Th,U)O_2$ fuel, the primary consideration was given to the following issues:

- effect of the CP design on the GFP yield;
- effect of irradiation temperature on the serviceability of CP;
- behaviour of CP during failure of the outer protective layer under irradiation both in the free-fill state and as constituents of the graphite spherical FE matrix.

To simulate the failure of the outer protective coating, a pilot batch of CP (21-10-X-87) was specially prepared without the mentioned coating. To investigate the radiation resistance of microspherical $(Th,U)O_2$ fuel, a few batches of CP enriched in uranium-235 to 21, 36 and 90 % were manufactured. The torium-to-uranium ratios were 3:1 and 9:1. When making CP, various designs were checked: type I (batch 36-13-X-84), type II* (batch 21-10-X-87), type III (batches 36-12-X-84 and 90T-1-X-88).

As mentioned above, the CP were reactor-tested in the free-fill state and as constituents of model spherical FE. The CP were generally irradiated at a temperature of 1250 ^{o}C. However, several radiation-resistance tests of the $(Th,U)O_2$ fuel under development were also performed at temperatures between 1500 and 1600 ^{o}C.

The reactor test values for CP with this fuel are listed in Table II. It is seen from the table that the rate of GFP from CP of the mentioned batches is at the same level as that shown by similar CP designs but with the uranium dioxide fuel [6]. In free-fill tests of CP (batch 21-10-X-87) without an outer pyrocarbon layer, irradiation was performed at an elevated temperature of 1250 ^{o}C to 2.1 % fima, and then, with a jump-like rise in temperature up to 1500 ^{o}C, - to 3.5 % fima. Immediately after the rise in temperature the rate of GFP release increased by nearly an order of magnitude (from $8.1.10^{-6}$ to $1.0.10^{-4}$ (Kr-88) and remained the same until the experiment completion. This behaviour of type II* design CP has led us to the conclusion about impossibility of using such CP at elevated irradiation temperatures ($T \geq 1500$ ^{o}C). Yet, these CP can be used under standard irradiation conditions when they enter into the composition of pyrocarbon-bound FE, since the 20-30 mm thick pyrocarbon films deposited as a result of pyrodensification can partly fulfil the functions of the outer protective pyrocarbon layer.

The model spherical FE manufactured on the basis of batch 21-1-X-87 CP were irradiated at 1250 ^{o}C to 9.8 % fima (fast neutron fluence of 2.1×10^{20} cm^{-2}). The CP retained high service ability up to the design burnup value (for type VGM- and VG-400-type reactors), but after the excess of which some part of CP lost their hermiticity.

Studies were made of free-fill CP for the vitality of uranium-thorium oxide microspherical fuel (CP of batch 36-12-X-84) under conditions of design abnormal rise in temperature (up to 1600 ^{o}C) for operating conditions of VGM- and VG-400-type reactors. On achieving the fuel burnups of (12.0-13.4) % fima (T_{irr} = 1350-1600 ^{o}C) and 13.9 % fima (T_{irr} = 1100-1200 ^{o}C), the CP retained their serviceability. The undertaken metallographic examinations have confirmed the integrity of protective coatings.

So, the behaviour under irradiation of the uranium-thorium microspherical fuel considered here is virtually the same as the behaviour of UO_2 fuel under the same reactor test conditions. It has been established by experiments that the $(Th,U)O_2$ fuel entering into the composition of CP retains its normal operation up to a temperature of 1600 ^oC, and the pyrocarbon binder of the matrix GSP-graphite further increases the CP hermiticity.

CONCLUSION

The present results of prereactor and reactor tests give evidence for high radiation resistance and performance characteristics of CP and FE comprising the $(Th,U)O_2$ fuel and manufactured by the NSC KIPT technology.

At present, the joint LLNL (USA) - NSC KIPT (Ukraine) project is under way to develop the concept of underground nuclear reactor with a nondischarged core throughout the service life (TIW-reactor). The NSC KIPT was commissioned to analyse and select different-purpose materials (fuel and structural) as candidate materials of the TIW reactor core.

On elaborating the underground nuclear reactor concept, the important scientific and technical challenge is to develop thorium-base fuel materials serviceable as a nuclear fuel. In this connection, when making choice for the TIW reactor we shall take into account the successful experience of experimental development of microspherical $(Th,U)O_2$ fuel for HTGR.

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AN ULTIMATE FISSION ENERGY SYSTEM OPTION FOR THE NEXT GENERATION - THE AMBIDEXTER NUCLEAR COMPLEX*

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Abstract. Regarding to long term perspectives of nuclear power, it is well known that a number of compulsory requirements have to be fulfilled. Those are ultimate safety, non-proliferation, HLW management, public acceptance, resource security and economics. An AMBIDEXTER (Advanced Molten-salt Break-even Inherently-safe Dual-missioning Experimental and Test Reactor) nuclear complex, a kind of closed nuclear system utilizing the standard Th-^{233}U fuel cycle was proposed, that promises to greatly remove obstacles associated with those. The AMBIDEXTER complex essentially comprises two mutually independent circuits of the heat/energy conversion and the radiation/material transport, centered at the reactor assembly. The entire reactor system including the core, heat exchangers and recirculation jet pumps is integrated in the reactor vessel. A three-loop-cycle configuration incorporated with the intermediate heat transport system is adapted for the heat/energy conversion circuit. For the radiation/material transport circuit, small bore piping connections on the reactor vessel periphery provide the bypass flow to a series of pyrochemical units where purify and recondition the irradiated fuel salt continuously. The composition of ^{7}LiF-BeF_2-ThF_4-$^{233}UF_4$ fuel material is optimized to ensure the criticality and the conversion ratio requirements for the self-sustainability. Consequently neither fissile material flows beyond the system boundary nor excessive fissile inventory inside the reactor system is required for operation. The integral reactor configuration makes it possible to divide the reactor vessel internal into compartments of reactor system components and to eliminate any active valves or piping connections between them. On-power purification of the fuel salt through the radiation/material transport circuit removes, partitions and isolates fission products from the reactor system as much as designed. In safety point of view, this significantly reduces both releasable radioactive materials and decay heat sources. In order to investigate the integral performance of the concept, the conceptual design studies of the 250 MW(th) prototype AMBIDEXTER system are in progress. Some of the important design data are given in this paper for discussion.

1. INTRODUCTION

Regarding to long-term perspectives of nuclear power, it is well known that a number of compulsory requirements have to be fulfilled. Those are, especially associated with the water-cooled and low enriched uranium-fuelled power reactor technology, the ultimate safety, non-proliferation and high-level radwaste management. Their secondary effects, such as public acceptability and economical competitiveness, also become growing fast to threatening the future of nuclear power. Without removing these obstacles, their advantageous performance of the environmental cleanness would be in the crucible.

As emphasized in the recommendation report [1] from the U.S. President's Committee of Advisors on Science and Technology (PCAST) issued on November, 1997, nuclear power, however, can be neither ignored nor easily replaced without significant environmental and economic costs, particularly in an energy future marked by unpredictable uncertainties and potential instabilities. They also recommend a strong nuclear energy R&D agenda to continue past efforts on those issues confronting nuclear power today by utilizing all the knowledge gained over the past several decades but not being bound by the designs available to date.

Nevertheless, because of the heuristic nature of present LWR-based nuclear technology evolution, it may be impossible to single out one issue from others and to resolve it independently. One of the potentially promising approaches to accomplish this goal is developing an innovative reactor concept with the 'clean slate' evaluation of the present nuclear power technology. Resulted from various studies, followings are identified as the basic requirements for the life cycle design considerations:

* 1999 meeting.

- a closed and independent system for fuel cycle;
- an isolated and integral system for energy source;
- an partitioning and recycle system for radwaste management.

This paper proposes an innovative nuclear energy system concept, the AMBIDEXTER(Advanced Molten-salt Break-even Inherently safe Dual-missioning Experimental and Test Reactor) nuclear complex and discusses its technical feasibility. The complex is a kind of closed nuclear system utilizing the standard Th-^{233}U fuel cycle that should contribute to solving the safety, proliferation and radwaste problems inevitable in LWR technologies. It also has many positive prospects in securing long-term nuclear resource supply and improving public feeling on nuclear energy.

2. AMBIDEXTER SYSTEM ATTRIBUTES

Nuclear safety

During the molten fluoride salt fuel flows upward through the simple hollow-cylindrical lattices made of solid graphite moderator blocks in the reactor core, the fission reaction and heat transport simultaneously take placed within the same material. Thus, the AMBIDEXTER complex essentially comprises two mutually independent circuits of the radiation/material transport and the heat/energy conversion, centered at the integrated reactor assembly inside where the bulk of fuel salt inventory circulates.

Apart from the defensive design approaches for safety, based upon the defense-in-depth principle for conventional nuclear power reactors, the safety design of the AMBIDEXTER follows the offensive approaches by taking advantages of online purification of fluid fuel contents. By means of continuous separation and reconstitution processes, in-reactor inventory of fission products as sources of decay heat and releasable radioactivity can be kept as low as reasonably achievable. Moreover, when postulate a severe accident such as a large failure of reactor vessel, the total amount of radioactive materials to be released into air should be ignorable, because volatile fission products generated in the circulating fuel salt can be completely filtered out by the salt purification processes. [2]

Other inherent safety features of the AMBIDEXTER are the strong negative reactivity coefficient of the fuel temperature mainly due to the large thermal expansion coefficient of liquid fuel and the small excess reactivity maintained in the core by means of continuous ^{233}U loading during power operation, which ensure extremely low probability of a uncontrollable power excursion at any credible reactivity accidents.

Assuming a loss of heat sink accident and the fuel temperature increased beyond the boiling point of the salt material, the peak fuel pressure, because of its negligible vapor pressure, should not be built enough up to cause a significant damage to the reactor system pressure boundary. And as the integral reactor concept confining the entire primary heat transport system in the reactor vessel and there is no major piping routings outside, a large break LOCA should be incredible. In addition, when consider a leak through a small size crack or a loosely coupled joint, the lost liquid containing radioactive materials becomes immediately vitrified as soon as exposed at ambient conditions.

Regarding to the safety aspect of configuration design baselines, instead of the multi-layer pressure boundary technique adopted for designing conventional integral reactors, the AMBIDEXTER utilizes a genuine passive system concept. The reactor system resides in a

thin and large reactor vessel whose internal is divided into number of functional compartments corresponding with the core, chimney, heat exchangers, downcomer and inlet plenum, and thus there is neither connection pipings nor active valves in-between the compartments. One other advantage of this simple compartment system is that total friction loss of system pressure can be kept low enough to reap natural circulation effects and to save pumping power demands.

Proliferation resistance

In spite of that the civilian nuclear power technology does not have any close link to the nuclear proliferation-sensitive areas, international concerns on diversion of nuclear technology and material steadily grow as more nuclear power plants come on the line. This is because not only of its technological attributes originally developed from military purposes, but of its uranium-based fuel cycle which requires the uranium enrichment processes and facilities, and leaves large amount of fissile plutonium isotopes in spent fuel.

The standard design of the AMBIDEXTER references with the Th/^{233}U fuel cycle instead of the ^{235}U/Pu cycle of LWRs'. Thorium in 100% natural abundance of ^{232}Th is processed to fuel material by ordinary chemical treatments, transmuted into ^{233}Pa by a neutron capture in the reactor and converted to the fissile, ^{233}U after the β-decay with the 27 days half-life. Therefore, neither uranium enrichment nor a plutonium separation process is required.

Because of the highest conversion ratio attainable by thermal neutrons due to large η value of ^{233}U, the thorium fuel cycle is known as only the appropriate one for designing thermal breeders. The AMBIDEXTER core geometry was optimized to facilitate the conversion ratio to be near break-even during lifetime operation, so that it would not be so difficult to find a self-sustaining fuel management scheme without unnecessary surplus or scarcity of the fissile inventory. Therefore, should one intend to produce excess ^{233}U using the AMBIDEXTER, he or she has to add equivalent amount of expensive and restricted ^{235}U or ^{239}Pu fissile to the fuel inlet stream.

In respects of safegurdability, the kinetics of ^{232}U concentration in fuel salt, born by the ^{233}U(n,2n)^{232}U reaction and lost either by the α-decay with the long half-life of 68.9 years or by the neutron capture with high probability of 141.5 barns, is very important. This is because that, as ^{208}Tl, a daughter element in the ^{232}U decay chain emits high energy (2.6 MeV) γ-ray, technical problems associated with storing and handling of a highly pure ^{233}U bulks but contaminated with ^{232}U become extremely difficult ever after a long cooling period.

Moreover, the AMBIDEXTER can easily adopt a denatured fuel cycle by adding a small amount of SEU denaturant. This leads the reprocess technology to become more difficult and thus significantly reduces the chance of fissile diversion risk.

HLW management

As for many countries operating conventional nuclear power plants, it is a general practice not to hurry up to have any solid decisions on the permanent destination of spent fuels, the temporary storage for voluminous spent fuels should be effective for considerable periods. On the other hand, for countries adopting the reprocess policy, the HLW containing more than 99% of nonvolatile fission products and about 0.5% of uranium and plutonium isotopes is generated after spent fuel reprocessing. From the nuclear safety point of view, in either cases, the uncertainties in predicting long-term behaviors of the α-emitting transuranium isotopes

with the half-life of more than 5 years and of the very long half-lived fission products such as ^{99}Tc, Cs, ^{129}I and ^{79}Se, remain too large to be implemented in the engineering design verification.

Nuclear characteristics of the Th-^{233}U composite under the base-salt material, ^{7}LiF-BeF_2 environment restraint the transmutation probability of Th to transuranium actinides by multiple capture of neutrons, compared to that of the ^{235}U-Pu fuel system. Concerns on the long-term safety of HLW storages, thus, become remarkably lightened in the AMBIDEXTER fuel cycle. Among many beneficial functions of the AMBIDEXTER on-line fuel purification system, separation and partitioning of nonvolatile fission products via multistage electrochemical processes can significantly reduce the volume and weight of the HLW to be easily managed and stored.

Radioactive materials in the separated HLW stream contain more than 60 isotopes distributed over broad ranges of the mass numbers and of the radiation energies, from which precious radioisotopes and radiation sources can be selectively produced. Besides, it is technically feasible to design a compact radwaste storage canister filled with chemically stabilized HLWs as the bulk source of large scale γ-irradiators for many industrial applications.

Miscellaneous

As indicated, paradigm changes in safety, proliferation and HLW should provide some positive impact on enhancing the public understanding of nuclear energy and technology. People can be more familiarized with benefits of radiation and radioisotopes. Non-power applications of nuclear heat, such as seawater desalination, heat and steam supply, etc., could be extensively considered. In economics aspects, liquid fuel system not furnished with cladding and structural materials drastically saves fuel fabrication cost. Modular design and shop fabrication of major systems including the integrated reactor assembly help to reduce construction period and cost, too. High temperature heat transport system with large thermal margin promises high efficiency of the thermodynamic cycle.

3. AMBIDEXTER SYSTEM ELEMENTS

As illustrated in Figure 1, the integral AMBIDEXTER nuclear complex embodies the design principle that the heat and radioactive materials generated by nuclear reactions should be separated as early as possible from their origin and be transported as effectively as possible to their terminals in the plant. This enables the reactor system to be operated in clean and cool condition desirable for economics and safety of the plant.

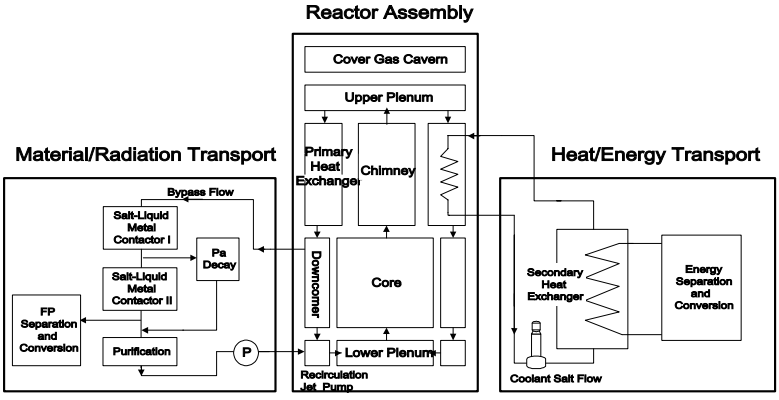

Figure 1. Schema of the AMBIDEXTER Nuclear Energy Complex.

Regarding system configuration, the AMBIDEXTER combines the passive reactor concept, equipped with few active parts or components, and the integrated reactor concept, networked without piping connection between equipment. Since the entire system is placed in the reactor vessel, divided into compartments of the reactor core, chimney, primary heat exchangers, downcomer and inlet plenum, the heat and friction pressure losses during circulation of the fuel salt in the system can be kept very low. Table I summarize the preliminary results of important design parameters derived for the standard 250 MW(th) AMBIDEXTER prototype module currently under development.

Integrated Reactor Assembly

For the fuel salt material, $^{7}LiF\text{-}BeF_2$ was selected as the base composition because of its proven performance under high temperature and radiation fields with respect to neutron economics, chemical stability, thermalhydraulic efficiency, solubility of heavy elements and fission products compounds, econotechnical competitiveness of its production, reconditioning and reprocessing processes, and so on. [3] In order to ensure the desired fluid conditions in terms of viscosity and homogeneity and to permit maximum loading of thorium and ^{233}U, limit on the fractional content of $^{7}LiF\text{-}BeF_2$ is evaluated to be no more than 87 mole %.

The concentration ratio of $^{233}UF_4$ to ThF_4 should be determined to satisfy the self-sustainability requirements, i. e., simultaneous fulfillment of the minimum excess reactivity and the break-even conversion ratio conditions, by continuously feeding appropriate quantity of ^{233}U fissile. In this aspect, for the present 250 MW(th) single fluid AMBIDEXTER fuel salt, the estimated contents of ThF_4 and $^{233}UF_4$ are of 12.0 mole % and 0.375 mole %, respectively.

The cylindrical reactor core compartment, dimensions of 282 cm dia. and 245 cm height, is placed at the lower center of the reactor vessel and surrounded by the reactor barrel. It is essentially simple and uniform arrangement of hexagonal graphite cylinders having the 10 cm face-to-face pitch and 250 cm height. The effective channel radii of the core and the blanket lattices are 1.856 and 4.696 cm, respectively through that the fuel salt upwardly flows and heats up. Reflector modules have the same dimension but no fuel channels are provided.

Table I. Characteristics of the 250 MW(th) AMBIDEXTER

Parameter	Data	Parameter	Data
-Gross power, MW(th)	250.0	-Reactor physics	
-Reactor vessel:		k_{eff} (HFP)	1.0056
Material/shape	Hastelloy/cylind.	C. R.	1.005
Dimension, cm	$420.0^{ID}\times 650.0^{H}$	Max. Flux, $\times 10^{14}$ nv	
-Core lattice:		φ_{th} ($E\leq$ 1.0eV)	7.43
Material/shape	Graphite/hex.	φ_f	14.42
Pitch, cm	10.0	F. tmp. coe., δ k/k°C	-4.62×10^{-5}
Eff. channel rad., cm	1.856/4.696	-Thermalhydraulics	
Eff. core rad., cm	141.0	Pk. pwr. den. w/c^3	127.9(salt)
-Fuel salt	$LiF\text{-}BeF_2\text{-}ThF_4\text{-}UF_4$	Core inlt/outlt tmp., °C	621.0/704.0
Composition	71.625-16.-12.-.375	Pwr peaking factor	$2.79^{R}\times 1.59^{H}$
Fraction, mole%	3.309	-Prmy. heat exchgnr.	
Avg. density, g/c^3	17.6	Type	Shell & tube

Since the density of the fuel salt, 3.309 g/c^3 is greater than that of the graphite fabricated, 1.9 g/c^3, the reactor core is supported by the upper and the lower grid plate structures enough to survive against the buoyancy force and other flow-induced dynamic loads. Both grid plates are anchored to the core barrel, so that the entire core assembly mechanically behaves so much as a single structure.

All the internal structures and the reactor vessel of the AMBIDEXTER are made of the Ni-based Hastelloy whose mechanical and corrosion resistance properties against molten fluorides have been verified. Although the chemical stability of the graphite is well proven even at extremely high temperature, large degradation in its mechanical strength has been observed when irradiated with high fast neutron fluences. Provisions, thus, for repairing or replacing damaged graphite modules may be necessary as one of the AMBIDEXTER lifetime maintenance considerations.

Heat/Energy Transport Systems

Fuel salt heated up to the core outlet temperature, 704 °C flows upwardly through the inner annulus chimney above the core top and dumps out its fission energy to the primary heat exchangers symmetrically arranged at the outer annulus surrounding the top of the chimney. Four units of the straight tube-and-shell type heat exchanger with dimensions of 82.2 cm O.D. and 231.6 cm length should have sufficient capacity to remove 250 MW(th) heat while small enough to be fit into the available annular space in the reactor vessel.

About 10 % of the total heat exchanger outlet flow, after cooled down to the core inlet temperature, 621ºC, is bypassed to the fuel salt purification system where the salt contaminated with fission products and ^{233}Pa is reprocessed. The purified bypass flow, then, returns to the recirculation pumps suction lines. The pumps, mounted on the bottom of the downcomer annulus outside the core barrel, feed the mixture of the bypass and the main recirculation flows into the core inlet plenum.

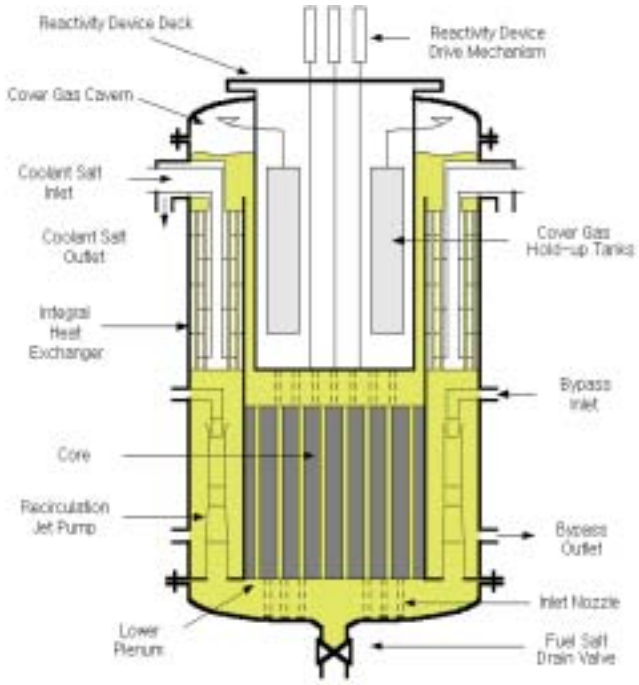

Figure 2. Cut View of Integral Reactor Assembly.

The compartment-type system concept limits the total system pressure drop not to largely departing from the total frictional pressure drop in the heat exchangers and in the fuel channels. Due to its negligible vapor pressure, temperature rising beyond the salt boiling point should not accompany any unacceptably high pressure loads to the reactor vessel. On top of this, the natural circulation enhancement due to large thermal expansion coefficient of and thermal margin of the molten fuel salt precludes the maximum system pressure and pump power from becoming severe design constraints. Preliminarily estimation predicts that the 250 MW(th) reactor system pressure should be less than 5 bars.

The AMBIDEXTER accommodates the intermediate heat transport (IHT) loop between the reactor system and the energy conversion system, which provides additional barrier against potential release of radioactive materials. Retaining good thermodynamic and chemical compatibilities with the fuel salt, the coolant salt in the IHT system will remove the rated power with operating temperature range of 455ºC to 565ºC . Also it can be said that, subject to various dynamic reactivity perturbations, as the stability of the reactor power is so high, the reactor power regulation solely by changing thermodynamic conditions of the IHT system should be sufficiently reliable. This inherent stability nature of the AMBIDEXTER extensively promotes its load-following capability.

Materials/Radiation Transport Systems

The 10% heat exchanger outlet flow fed into the small bore bypass pipe suction nozzles penetrating reactor vessel wall close to bottom of the downcomer, stays at the holdup tanks before being sent to process units of the on-line fuel purification system. In these tanks, a cooling means is provided to bring down the fuel salt temperature suitable for purification processes and to remove decay heat from the very short half-life fission products.

The multistage electrochemical process units having been developed for the single fluid MSBR program at ORNL in 1970's are assumed to be available for the system design basis. Studies on evaluating their process time constants and associated economic impacts are undertaken. The system units comprise the fluorination process for separating Th and ^{233}U, the reductive extraction process for partitioning ^{233}Pa and rare earth FPs, the adsorption process for stabilizing noble gases such as Xe and Kr.

Among these process units, the noble gas separation is to be accomplished by sparging tiny helium bubbles into the reactor system and collecting them at the cover gas plenum inside the upper head of the reactor vessel. The cover gas system takes functions of controlling the reactor system pressure and stripping off the noble gas fission products effectively.

The protactinium separation process, in public viewpoints, may impose a burden on non-proliferation superiority of the AMBIDEXTER, although the reactor could not operate for a considerable time when the fissile balance governed by the break-even conversion ratio is broken. Alternative design options of the system of separating fission products but not isolating protactinium from the bypass stream remain as one of interesting subjects for future study.

Before feeding back to the reactor inlet, reprocessed salt with significantly reduced neutron poison and decay heat sources, needs reconditioning for compensating the burnup loss of ^{233}U and the neutron capture loss of Th in order to guarantee the criticality condition for steady full power operation.

4. CONCLUSIONS AND REMARKS

Nuclear experts in the world do not much differ in views over the future of nuclear power, that the present technology stemed from the LWR development would not guarantee ultimate resolutions for vital issues of the nuclear safety, proliferation and HLW management. Bearing in mind this cloudy perspective, this paper suggests development of the AMBIDEXTER concept, a new integral and closed nuclear energy system for a long-term solution.

The AMBIDEXTER reactor system adopts most of generic features of the molten salt reactors, for examples, the standard Th-^{233}U fuel cycle, graphite-moderated core, Hastelloy reactor material and on-line fuel reprocessing. The figure of the integrated compartment concept embodies its distinctive attribute that the heat and radioactive materials generated by fission reactions can be separated as early as possible from their origin and transported as effectively as possible to their terminals in the plant. And the self-sustainability requirement is realized by optimum design of the core lattice to achieve the break-even conversion ratio under minimum loading of fissile.

In nuclear safety aspect, this new configuration design incorporates the genuine passive system by eliminating pipings and active valves from the equipment routing and the offensive safety by keeping the in-reactor inventory of decay heat and releasable radioactivity sources as low as reasonably achievable. Inherent features of very low excess reactivity and strong negative temperature coefficient of the fuel salt also suppress the power runaway at loss of regulation events.

The Th-^{233}U fuel cycle subject to self-sustainability enhances safeguard transparency as well as resource security when consider its flexible fuel management applications. Nonvolatile HLWs separated and partitioned via multistage electrochemical processes should have largely reduced volume and weight, so that the storage and management of HLW become much easier.

To demonstrate the validity of the AMBIDEXTER concept, a conceptual design study of the 250 MW(th) prototype module with emphasis on investigating its integrated performance, is in progress.

ACKNOWLEDGEMENT

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PERSPECTIVE OF THORIUM RESEARCH AND BORON CARBIDE COATING OF URANIA-GADOLINIA FUEL IN TURKEY*

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Abstract. Turkey has thorium resources amounting to about 380,000 tonnes as observed until today. Thorium content in complex ore reserves is roughly 0.2% in average. An extensive survey including determination of tenor for complex ore is expected to be undertaken till the end of year 2000. Other main constituents are composed of rare earth elements including yttria (^{203}Y). These constituents and regional thorium contents are tabulated and mapped in this presentation in detail. The research and development (R&D) institutions in Turkey consider R&D studies on thorium fuel technology as a key element for future reactor technologies. Recently, some other R&D investigations on advanced fuel production were undertaken at the Middle East Technical University (Ankara) in collaboration with the Turkish Atomic Energy Authority. As an example; pure UO_2 and urania-gadolinia (5% and 10% gadolinia) fuels were then coated with boron carbide by chemical vapor deposition (CVD). Boron carbide was produced from the reaction of carbon tetrachloride and boron trichloride under excess hydrogen in a thermal CVD furnace at 1000°C - ll75°C. The morphology and the thickness of the coating and preliminary burnup calculations have been investigated.

1. INTRODUCTION

In the 21st century, the conventional fission energy issue may not be practical to solve the global energy problem. Not only severe global environmental problems, but also economical energy supply, poverty and desertification of the world will not be inevitable.

However, the future nuclear energy technology will undertake the main global difficulties in the medium and long terms in the next 21st century. Therefore, increase in energy demand in the next century will lead to a considerable increase in nuclear energy contribution when climate change problems are considered [1].

In order to solve these global energy and environmental problems, a new philosophy is required. One of this philosophy can be thorium utilisation, as thorium is more abundant and widely localized resource in the world. In addition, thorium molten salt technology has main benefits, such as utilisation of fissile materials; ^{233}U, ^{235}U and plutonium, and this utilisation is achieved without reactor-core design modifications [2]. Additionally, molten salt reactors carry some advantages in terms of cost and safety.

On the other hand, fluid fuel reactors possess attractive waste facilities due to continuous processing and immediate separation of the residual fuel from waste, and simple waste handling applications.

* 1999 meeting.

There are three possible general fuel types to burn plutonium in light water reactors (LWR):

i) uranium-based mixed oxides $(U,Pu)O_2$, (MOX);
ii) thorium-based mixed oxides $(Th,Pu)O_2$ and
iii) inert matrix fuels (IMF).

In these types of fuels, PuO_2 is dispersed in a neutron transparent ceramic carrier [3] (such as yttria, YzO_2, zirconia, ZrO_2 etc.). Plutonium oxide is diluted in inert ceramic oxides and/or thoria. The $(Th,Pu)O_2$ fuel contributes to increase the Pu consumption with respect to $(U,Pu)O_2$ fuel in terms of lack of uranium. Furthermore, the $(Th,Pu)O_2$ fuels exhibit good capability under irradiation conditions and they have high stability which enable us to produce a ^{233}U having high gamma activity in burnt fuel.

2. THORIA FUEL STUDIES

Recovery of thorium from the ore and production of thoria fuel powder were carried out in the past, including sol-gel with oxalate precipitation [4] and calcination of thorium oxalate to obtain thoria fuel. Mixed thoria-urania fuel pellets are prepared by mechanical blending in various proportions. In the above study [4], more than 90% theoretical pellet density is obtained. Sintering kinetics of prepared pellets is investigated at a high sintering temperature.

The survey on the thorium and rare earth element deposits and tenor determining studies is expected to be speeded up when sufficient financial support is provided. Therefore the exact amount of thorium and rare earth elements are not yet to be determined. But recent determination of thorium oxide reserves is about 380,000 tonnes in Eskisehir estate. Thorium and uranium reserve distribution in Turkey is mapped in Figure 1.

Reserve determination survey exhibited a various thorium and rare earth element contents according to the different deposit regions. While some ore deposits seem to be rantable to operate thorium ores, in contrast, some deposits do not seem to be rantable to operate ores. However, it seems to be rantable in case of operation in both thorium and rare earth elements ore together.

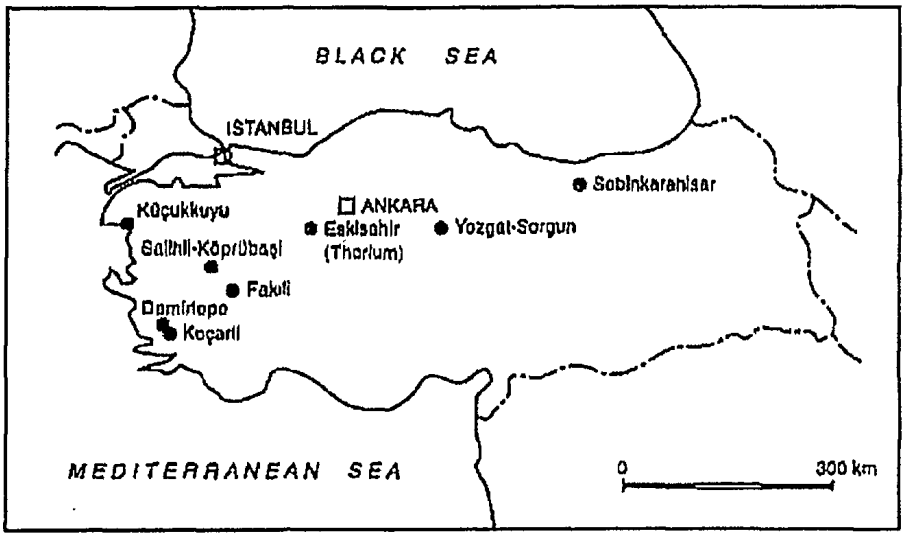

Figure 1. Thorium and uranium deposits and occurrences in Turkey.

Rare earth contents in thorium deposits are about 3% in a cumulative average. Yttria content in determined in rare earth deposits is considerably a high amount. Some deposits in Eskisehir have reached to 8-9% rare earth element tenor and 0.12-0.02% thoria content. In China, rare earth element and thoria content have reached to 7% and 0.035% respectively. Thorium and rare earth element contents in examined ore samples are tabulated [5,6] in Tables I and II.

Table I. Elemental Concentrations of Ore Samples.

Element	KTS-1	KTS-2	KTS-3	KTS-4
Na(g/kg)	-	7.2	7.8	8.3
Fe(g/kg)	26.7	96.5	26.0	18.8
Sr(g/kg)	-	2.50	1.3	18.8
Y(g/kg)	-	-	-	42.3
Ba(g/kg)	111	132	36	145
La(g/kg)	3.84	9.42	4.35	12.5
Ce(g/kg)	15.3	13.6	4.8	12.0
Nd(g/kg)	14.8	3.4	1.3	2.5
Sm(g/kg)	3.00	0.63	0.15	0.35
Th(g/kg)	5.3	2.9	2.9	1.5
Sc(mg/kg)	18.0	14.6	10.6	8.4
Cr(mg/kg)	500	39.1	30.1	168
Co(mg/kg)	25.8	1.8	1.4	1.0
Zn(mg/kg)	-	-	360	210
As(mg/kg)	200	-	160	-
Zr(mg/kg)	143	87	94	104
Nb(mg/kg)	3.0	1.2	1.6	7.0
Sb(mg/kg)	5.6	2.9	3.8	10.7
Cs(mg/kg)	10.3	1.0	1.8	5.0
Eu(mg/kg)	370	71.1	27.4	41.3
Gd(mg/kg)	540	420	520	150
Tb(mg/kg)	54	12.5	8.2	11.3
Yb(mg/kg)	62.6	28.4	21.9	37.4
Lu(mg/kg)	-	-	36.2	-
Hf(mg/kg)	4.2	10.6	30.1	4.4
U (mg/kg)	9.4	181	8.0	8.61

As seen from the Tables I and II, thorium content does not seem to be rantable by itself except rare earth elements. Thoria deposit ores almost include all the following rare earth elements in various contents. Since the importance and usage of rare earth elements are continuously increasing, more research should be carried out for both rare earth elements and thorium estimation in Turkey.

Reserve determination survey is not over, it is expected that the amount of thoria and rare earth elements would increase more as survey studies sustain at these regions.

Table II. Elemental Concentrations of Ore Samples.

Element D(	Dolamitte-2	Dolamitte-4	K. Hoyuklu	K. Devebagirtan
Na(g/kg)	-	16.0	-	-
Fe(g/kg)	4.30	5.18	2.69	3.19
Sr(g/kg)	-	1.55	2.34	-
Ba(g/kg)	0.44	0.45	109	72.6
La(g/kg)	5.0	-	10.3	12.4
Ce(g/kg)	6.2	5.0	21.3	22.0
Nd(g/kg)	-	-	3.9	-
Gd(g/kg)	1.13	0.41	0.19	0.41
Th(g/kg)	11.5	4.0	2.4	3.0
Sc(mg/kg)	35.2	32.2	14.0	42.5
Cr(mg/kg)	27.7	82.8	74.4	66.6
Co(mg/kg)	27.2	22.2	5.3	4.6
Zn(mg/kg)	210	-	200	320
As(mg/kg)	-	-	-	6.66
Y (mg/kg)	-	-	840	-
Zr(mg/kg)	144	165	97	95
Nb(mg/kg)	6.08	4.20	1.05	1.27
Mo(mg/kg)	-	-	46.7	-
Sb(mg/kg)	20.5	23.8	11.4	4.0
Cs(mg/kg)	11.5	4.8	9.2	8.4
Eu(mg/kg)	20.6	11.8	46.0	67.8
Tb(mg/kg)	17.4	6.6	8.0	18.5
Yb(mg/kg)	69.0	50.6	19.2	28.7
Lu(mg/kg)	-	-	2.15	-
Hf(mg/kg)	4.22	3.37	1.38	9.71
Ta(mg/kg)	1.0	0.95	-	1.46
U (mg/kg)	-	996	24	45

3. BORON CARBIDE COATING OF URANIA-GADOLINIA FUEL

Controlling a nuclear reactor with strong neutron absorbers causes flux depressions at certain locations, therefore fluctuation of power production, and inefficient burning of fuel is inevitable. However, the new generation control rods are made of weak neutron absorbers. In this case, the excessive reactivity is compensated with the amount of burnable absorbers used in fuels. Thus, burnable absorber consumption and build up of fission products minimise the need for the control rods.

Recently longer operating fuel cycles and more efficient fuel management have been proposed to utilize some form of burnable absorbers for power distribution and reactivity control. For this aim, suitable burnable absorbers can be Gd, Er, Ho or B. Gadolinium is being used as a burnable absorber in the commercial reactors. In recent studies, erbium, Er, seems to be used as the best compromise, in spite of a fairly large residual absorption among the effective burnable absorbers [7].

Burnable absorbers in nuclear fuel are commonly used in current nuclear reactor design. Since they are a part of the fuel rod, separate handling of the absorbers used in fuels can be classified in two categories : burnable absorber mixed with fuel (gadolinium oxide mixed

with fuel), and burnable absorber coated with fuel (zirconium diboride, ZrB_2, and boron nitride, BN). ZrB_2 coated gadolinium oxide fuels are commercially available.

There are many advantageous of gadolinia on soluble absorbers: first, Gd has higher thermal cross section than boron ; this contributes to the significant control of the reactivity at the very beginning of the cycle when fuel is very fresh. In addition, it does not cause residual reactivity at the EOC. Secondly, boric acid changes the properties of the moderator (water) such as thermal conductivity, pH, corrosivity, density. Thirdly, the cooling system creates additional waste, ion exchange resin and necessitates additional storage and disposal. Lithium formed due to the reaction of $^{10}B(n,^{7}L)He$, must be removed in the ion exchanger.

However, the use of excessive amount of gadolinia creates some problems. The thermal conductivity of the fuel decreases as gadolinium content increases. Gadolinia retards sintering and so results in higher porosity. In addition to the above, it causes solid solution formation in the fuel.

Another method of using burnable absorber is to coat the fuel pellets with a thin layer of burnable absorbers. In this method the fuel is coated with ZrB_2 and boron nitride [8]. Boron has a low neutron absorption cross section compared to the gadolinium. Therefore boron does not totally burn out when it is mixed with the fuel and introduces residual negative reactivity at EOC.

However, as a burnable absorber exists on the surface of the fuel, it interacts with the thermalized neutrons on their return from the moderator before the fuel does. As the thermal neutrons are partially absorbed by burnabl absorber on the fuel surface the spectrum becomes hardened. So burning of boron is achieved at high rate while sufficiently hardened neutrons go inside the fuel. This naturally increases the conversion efficiency of ^{238}U into ^{239}Pu.

The fabrication technology of ZrB_2. is quite complicated and the dissolution of unburned ZrB_2 creates solubility problems. On the other hand, boron nitride coating of the fuel is relatively a new technique and more advantageous than ZrB_2 coating. However, the BN coating has a disadvantageous since reaction with $^{14}N(n,p)$ results in undesired radioactive ^{15}N on the surface of the fuel.

In this study, a new approach is applied during the coating of the fuel pellets with burnable absorbers. This is boron carbide coating of the fuel by chemical vapor deposition technique. Some difficulties and solubility problems in ZrB_2 coating technology and also radioactive neutron reaction with nitrogen in BN coating have however eliminated by coating fuel pellets through B_4C.

3.1. Experiment

In this experiment, UO_2 and UO_2-^{203}Gd nuclear fuels are obtained by sol-gel technique, and then these fuels are pelletised and sintered. After that pellets are coated with boron carbide in a thin layer (about 5 μm thickness) through chemical vapor deposition [9].

The B_4C films prepared in this experiments are formed by reacting BCl_3 and CCl_4 under excess H_2, at temperatures of 1000°C, 1100°C and 1175°C at 1 atm in a CVD tube furnace. BCl_3 and CCL_4 are carried by argon and hydrogen respectively into reaction medium. CVD

tube furnace used in this experiment is shown in Figure 2. The general overall reaction to form B_4C is the following:

$$mCCl_4 + nBCl_3 + H_2 \rightarrow B_nC_m\ (+HCl)$$

This reaction gives homogeneous boron-carbon compounds with rhombohedral structure and a carbon ratio of 9-20% ($B_{10}C$-B_4C). The electrical and thermal conductivities were found to change with the carbon content.

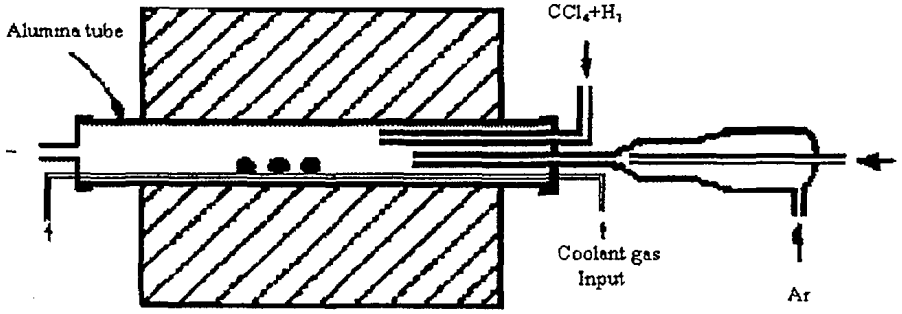

Figure 2. Thermal CVD tube furnace.

As substrate, silica plates were also used besides three nuclear fuels. CCl_4 was kept warm at 76 °C and its vapor was carried to the reaction medium by H_2 gas. BCl_3 kept in a steel tube was fed to the reactor with Ar gas through a nozzle. The ratio of BCl_3/CCl_4 was kept between 4-9 throughout the experiment.

4. PRELIMINARY NEUTRONIC CALCULATIONS OF THE COATED FUEL

The preliminary neutronic calculations performed by WIMS-D/4 have been mainly based on k_{eff} and burnup. Fresh fuels are replaced near the center of reactor vessel to decrease neutron exposure to the vessel and thus to increase the reactor vessel life in in-core fuel management. In this case, fresh fuels result in power peaks in the reactor core. Burnable absorbers, like gadolinium oxide, have been added natural or slightly enriched fresh fuels to remove these power peaks.

In this study, standard and slightly enriched standard fuels are assumed to be coated with boron carbide and used in PWR. Boron carbide coated fuel is examined in comparison with boron nitride coated fuels due to its superiorities [9].

A standard PWR has been assumed with 17X17 fuel bundles. There is a neutron detector or neutron source tube together with 24 blank tubes for control rods in the center of the bundle. The remaining 240 fuel rods consist of 4% enriched UO_2, and 24 fuel rods consist of 4% enriched UO_2 or B_4C coated 4% enriched UO_2. These are symmetrically situated in the fuel bundle.

Three different unit fuel cells are used in this computation: a standard fuel with no burnable absorber, fuel containing burnable absorber gadolinia and also boron carbide coated fuel.

Burnup versus k_∞ data for boron carbide and boron nitride coated fuel is shown in Figure 3. B_4C coating is more effective in terms of k_∞ increase in comparison to BN coating owing to the higher boron content in B_4C compound.

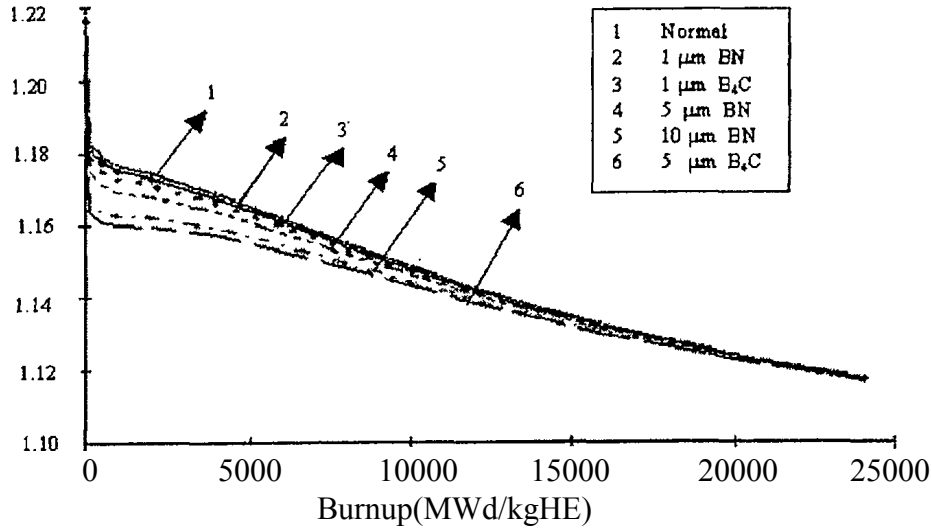

Figure 3. Comparing k-infinity.

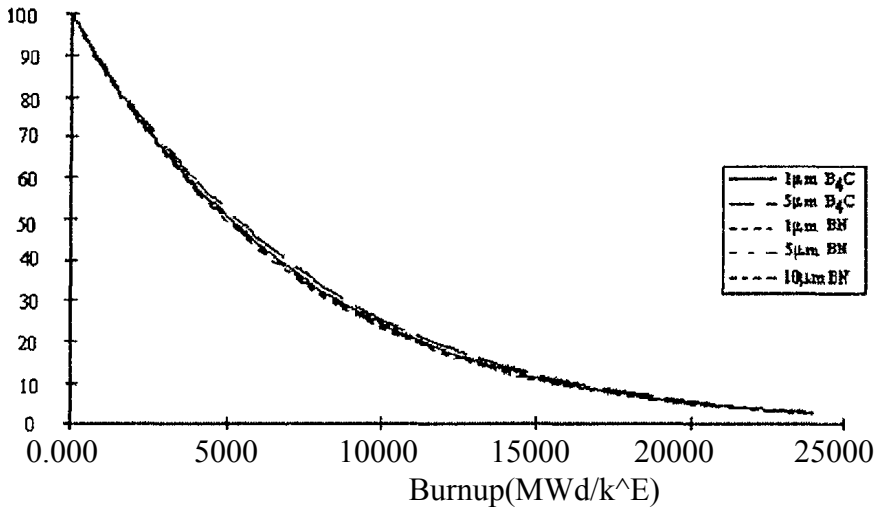

Figure 4. Comparison of burnable poison.

Burnable poison and burnup curves are almost same and overlapped in both B_4C and BN cases given in Figure 4.

Burnup and amounts of fissile ^{239}Pu and ^{241}Pu produced during burning are given in Figure 5. As seen from the Figure 5, the curves are overlapped in both boron carbide and boron nitride coated cases. Thus B_4C coating and BN coating are comparable due to fissile Pu-production.

.B_4C and BN coated fuel bundles are examined in terms of power distributions and compared in 5 μm coating thickness. Power peak values for B_4C and BN coating is almost constant at the order of 1.05 value.

B_4C coating yields the same k_∞ effect as BN coating even with smaller thicknesses due to the high amount of boron content in B_4C. Therefore B_4C coating compared to BN coating is more effective and superior in terms of neutronic perspective especially in k_{eff} case.

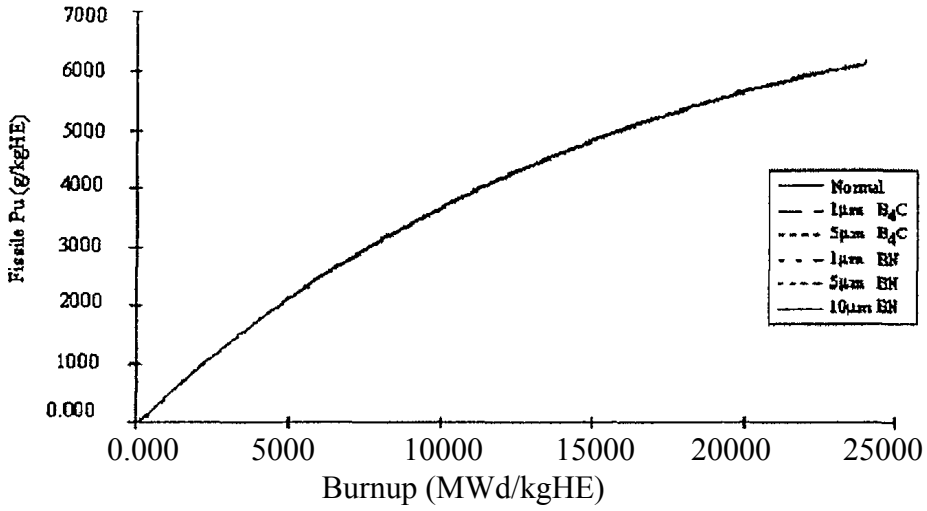

Figure 5. Comparison of fissile plutonium.

5. CONCLUSIONS

Thorium deposits

1. Thorium survey studies together with rare earth elements have not been finished until today. This survey should be finished as soon as possible, as Turkey has considerable amounts of thorium deposits.
2. At the present time thorium and rare earth deposits seem to be rantable due to co- operation of them. Especially yttria (Y_2O_3) between rare earth deposits is to be considered.

B_4C coating

1. Homogeneous and thermodynamically stable B_4C compound was successfully coated on pure urania and urania-gadolinia fuel.
2. Boron carbide coats were formed in layered, particle and rod shaped on the fuel experiments.
3. Coated B_4C compound did not penetrate and interact with the fuel but it only well-adhered on the fuel surfaces.
4. B_4C coating thicknesses were mostly 5 μm however 12 μm thicknesses were rarely observed.
5. B_4C concentration was decreased almost in a negligible amount in any step after burnup at 24000 MW·d/kgHE (MW·d/kg Heavy Element).
6. B_4C coating compared to the BN coating is more effective and superior at the same coating thickness in terms of neutronic perspective. Power peak values both B_4C and BN coating cases are almost constant at the magnitude of 1.05 value.
7. ZrB_2 coating is quite complicated and also there are some solubility problems in the fuel processing management. On the other hand, the BN coating has disadvantage since reaction with $^{14}N(n,p)$ results in an undesired radioactive ^{15}N reaction on the fuel surfaces. However, B_4C coating of the fuel has mostly eliminated such the above problems due to the coating of the pellets with ZrB_2 and BN.

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V. PROBLEMS TO RESOLVE: SAFETY, NON-PROLIFERATION, ENVIRONMENT PROTECTION

POTENTIAL ADVANTAGES AND DRAWBACKS OF THE THORIUM FUEL CYCLE IN RELATION TO CURRENT PRACTICE: A BNFL VIEW*

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Abstract. Thorium could extend the availability of nuclear fuel beyond the necessarily finite reserves of uranium ore, particularly if used in a thermal breeder system with the uranium-233 formed by transmutation serving as fissile content. The cycle produces virtually no plutonium, nor the other transuranic elements that contribute substantially to anxeties about the disposal of nuclear waste. Thorium-based fuels have therefore been proposed as a substitute for uranium, both in existing power reactors and in advanced systems such as the 'energy amplifiet,' with a sub--critical assembly of fissile and fertile material driven by an independent neutron source. The benefits and drawbacks of thoriurn need careful evaluation. A self-sustaining, breeding cycle should be possible with good neutron economy, but whether existing modern reactor types meet that condition is questionable, particularly at high fuel ratings where parasitic absorption by ^{233}Pa tends to pre-empt decay to ^{233}U. Radiation from thallium-208, formed in the decay of by-product ^{232}U and ^{228}Th, complicates storage and refabrication. Public perception would favour the cycles producing no transuranic elements and its particular capacity for consuming those already stocked; however, although they contribute largely to the long-lived content of nuclear waste, fission products also do likewise, and since the amounts of these are not greatly changed, any resulting improvement to long-term safety would by no means be decisive. BNFL has recently assessed the outstanding development requirements of the Thorex process. Commercial realisation would require a huge investment with no certainty of success. So far, the potential advantages do not seem likely to justify the risk, but the position is being kept under review in case the balance should be seen to shift.

1. INTRODUCTION

Thorium has long been recognised as a possible nuclear fuel, since although the natural element consists almost entirely of the non-fissile ^{232}Th, it is fertile in being capable of transmutation by a neutron flux into the fissile uranium-233. In favourable circumstances, given a start with some other neutron source such as ^{235}U, plutonium or an accelerator-driven generator, it can sustain a thermal breeding cycle in which as much fissile material is generated as consumed. This cycle has been adequately demonstrated in principle.

Thorium, particularly with breeding, could therefore add to the available nuclear fuel currently dependent on the established uranium cycle. For neutronic reasons, [1] thorium is also somewhat more suited than uranium to high-temperature reactors (HTRs) which are capable of better thermodynamic efficiencies than the currently dominant water-cooled types, or of serving as sources of process heat. However, uranium is still plentiful, HTRs have never been commercially deployed, and the thorium cycle has not been widely adopted. The chief reasons for a revival of interest after a period of neglect are that:

- in the absence of ^{238}U, the cycle produces virtually no plutonium or other transuranic elements;

* 1997 meeting.

- the waste products are therefore free from the long-lived alpha-emitters that arouse particular anxiety about the ultimate safety of permanent disposal;
- in various schemes intended to consume plutonium (where seen as a liability rather than an asset), stocks could be depleted more rapidly than if consumption were offset by fresh generation from uranium as principal fuel component; and
- rightly or wrongly, ^{233}U may be thought to present lower proliferation risks than plutonium.

Thorium has therefore been suggested as a substitute for uranium-based fuels both in existing reactors and in advanced systems such as the proposed "energy amplifier," [2] where a sub-critical fissile-fertile assembly is driven by an accelerated proton beam impinging on a heavy-metal target as an independent neutron source.

The true thorium cycle requires breeding and recycling ^{233}U, i.e. reprocessing irradiated fuel to recover at least the fissile content. The necessary reprocessing technology is unproven on a commercial scale. Advanced dry processes are especially so in this context, and for the foreseeable future, reprocessing means a wet-chemical operation in which the fuel substance is dissolved in nitric acid and separated by solvent extraction. Thorium is considerably less amenable than uranium to such processing.

The weakest link in the thorium cycle has always been, and remains, reprocessing to recover fissile and fertile materials from irradiated fuel, a necessary step if the breeding potential of the system is to be realised. It has been done only under experimental, makeshift or small-scale arrangements far from commercial practice. Any attempt to graduate from such trials to the rigours of full-scale industrial application would require much time-consuming and expensive development work, with prospects of technical success uncertain and of commercial viability even more so. Before a programme of this magnitude is undertaken, the likelihood that the benefits would justify the costs needs to be carefully assessed. This paper represents the view, based on the literature, of BNFL as an organisation with considerable interests in the development of fuel cycle technology.

2. THE THORIUM-URANIUM CYCLE

Transmutation of ^{232}Th into ^{233}U is closely analogous to that of ^{238}U into plutonium.

$$^{238}U \xrightarrow{n} {}^{239}U \xrightarrow[23.5\ min]{\beta} {}^{239}Np \xrightarrow[2.355\ days]{\beta} {}^{239}Pu$$

$$^{232}Th \xrightarrow{n} {}^{233}Th \xrightarrow[22.3\ min]{\beta} {}^{233}Pa \xrightarrow[27.0\ days]{\beta} {}^{233}U$$

The obvious difference between the half-lives of intermediate ^{239}Np and ^{233}Pa is unimportant once fuel is discharged from the reactor, since a lapse of several years is customary to allow partial decay of fission products before reprocessing, and during that time, conversion from either intermediate would be essentially complete. Within the reactor, however, the relatively long half-life of ^{233}Pa allows its absorption of neutrons to compete significantly with decay [3]. The ^{234}U daughter of the resulting ^{234}Pa is non-fissile, and although it could be

transformed by further absorption into ^{235}U, the overall three-neutron shift from the initial ^{232}Th means that the breeding ratio is necessarily poor and the absorption by ^{233}Pa essentially parasitic. Even if reducing production of ^{233}U by only a few percent, this could tip the balance to a net loss of fissile material. Efficient breeding is therefore incompatible with high neutron flux, and so with high fuel ratings.

If the cycle is to be self-sustaining in fissile material, fuel must be constructed and irradiated in such a way that the formation of fresh ^{233}U at least keeps pace with fission and incidental losses, either homogeneously or in a fertile blanket around the fissioning elements. In the former case, the fuel should remain in the reactor until as much as possible of the fertile component has been transmuted and consumed; the HTR type, comprising impermeably-coated ceramic particles dispersed in a moderator matrix, is said to be capable of achieving about 50% burn-up. However, in view of the neutron-absorbing fission products which would then accumulate extensively and compete with breeding, such irradiations might need:

- an unusually high initial fissile content with a consequent requirement to control excess reactivity by means such as burnable poisons or enhanced control-rod worth;
- much reshuffling of fuel components to maintain throughout the core a balance of fresh and irradiated fuel, such as may be feasible with pebble-bed or CANDU reactors but not compatible with economic light-water reactor (LWR) regimes; or
- a breeding efficiency higher than is likely to be attained in current LWRs, where significant parasitic absorption of neutrons occurs in both the coolant and (with relatively high ratings) the ^{233}Pa intermediate; other types using moderators such as graphite or heavy water would probably be required, with low fuel ratings implying large cores and high capital cost per unit power.

The alternative breeding regime, not relying on high burn-up in a single pass, is to process fuel periodically in order to separate fissile, fertile, and parasitically neutron-absorbing constituents, and to reconstitute the fissile-fertile mixture with whatever replenishment may be needed from other sources. Such processing is a current industrial practice in the uranium-plutonium cycle, but much more difficult and less firmly established with thorium [4]. This is a particularly important issue to be resolved; other comparisons with the uranium-plutonium cycle are mainly related to the overall value of the system.

Reprocessing difficulties are especially severe with HTR-type fuels of whatever composition, since the particle coatings and graphite matrix are chemically resistant and troublesome to break down mechanically. The cost of reprocessing would therefore be higher than for LWR types, while an extreme burn-up would diminish the value of recoverable material, at the same time reducing the volumes of discharged fuel for a given electrical output. The balance of advantage between reprocessing and direct disposal would then be much more favourable to the latter than with LWR or similar fuels irradiated for instance to 30 - 60 GW·d/t.

Accordingly, reprocessing is most likely to be needed only for fuel of much lower irradiations. The necessary techniques of breaking down fuel ready for presentation to a chemical separation process, and notably the problems peculiar to HTR fuels, are common to both uranium- and thorium-based cycles. Accordingly they are not directly relevant to the comparison, which can thus start at the first step towards separation.

3. REPROCESSING

The choice of methods is limited. Dry processes have been much investigated as a way of avoiding the aqueous wastes associated with the solvent extraction currently universal in commercial practice; halide volatility and electrochemical or pyrochemical separations in molten salts are the chief contenders.

Fluoride volatility, while inapplicable to thorium itself, might at first sight appear attractive to separate the fissile uranium content, but would require a supplementary means of penetrating the thorium matrix to allow attack on the dispersed uranium. High-temperature chlorination has been tested on a pilot scale with some success, [5] although whether it could be applied industrially without insuperable problems of safety and corrosion remains doubtful. Despite early interest in the application of molten-salt methods to thorium, [6] they are so far experimental, and of late considered chiefly in relation to advanced metallic uranium-plutonium fuels; extra steps would be needed for oxide. For the foreseeable future, therefore, the only likely route is by solvent extraction, which in practice means by some form of the Thorex process where thorium and uranium are extracted by tri-*n*-butyl phosphate (TBP) in a hydrocarbon diluent. [4] To this end, the fuel substance must be dissolved in nitric acid.

4. DISSOLUTION

For the purpose of comparison, fuel is assumed to be oxide. Herein lies one particular difficulty: thorium dioxide, like plutonium dioxide, is scarcely attacked by plain nitric acid unless homogeneously dispersed in a less resistant matrix. The difference is that plutonium is very much a minor component in thermal uranium fuels, well below the limit of about 30-40% at which a true solid solution of the two oxides is still soluble without the need for complex-forming additives; in the thorium cycle, uranium is the minor component and the fuel practically inert to nitric acid alone. For this reason, the Thorex dissolver reagent contains hydrofluoric acid which does attack thorium dioxide, albeit slowly. Unfortunately it also attacks most likely materials of construction, so the reagent includes aluminium nitrate to buffer the concentration of free fluoride ion at a level tolerably effective for its purpose but not unduly corrosive. Of course its palliative effect does not extend to the vapour space, where the worst problems of corrosion are liable to be met. The problem may be alleviated by advances in fluorine technology, but to avoid unexpectedly severe corrosion in other process areas, it demands extreme caution and can hardly fail to increase costs significantly.

The difficulty might in principle be avoided by using a different thorium compound less inert than the oxide. The carbide, for instance, is readily hydrolysed [7], and has other advantages such as higher thermal conductivity and heavy-metal concentration that would be beneficial in the reactor, but some of the organic compounds formed on dissolution have surfactant properties expected to interfere seriously with the operation of solvent-extraction equipment.

A preliminary combustion to oxide is said to be the best way of avoiding this particular problem but in terms of dissolution restores the original difficulty, while an acid-free hydrolysis that could produce a more tractable hydrated form of oxide would still presumably require the organic materials to be separated or destroyed. Moreover, carbon in the fuel would be a
source of ^{14}C which could have a significant radiological impact, an objection that also applies to nitride. The problem of dissolving oxide appears to be the least troublesome of the alternatives.

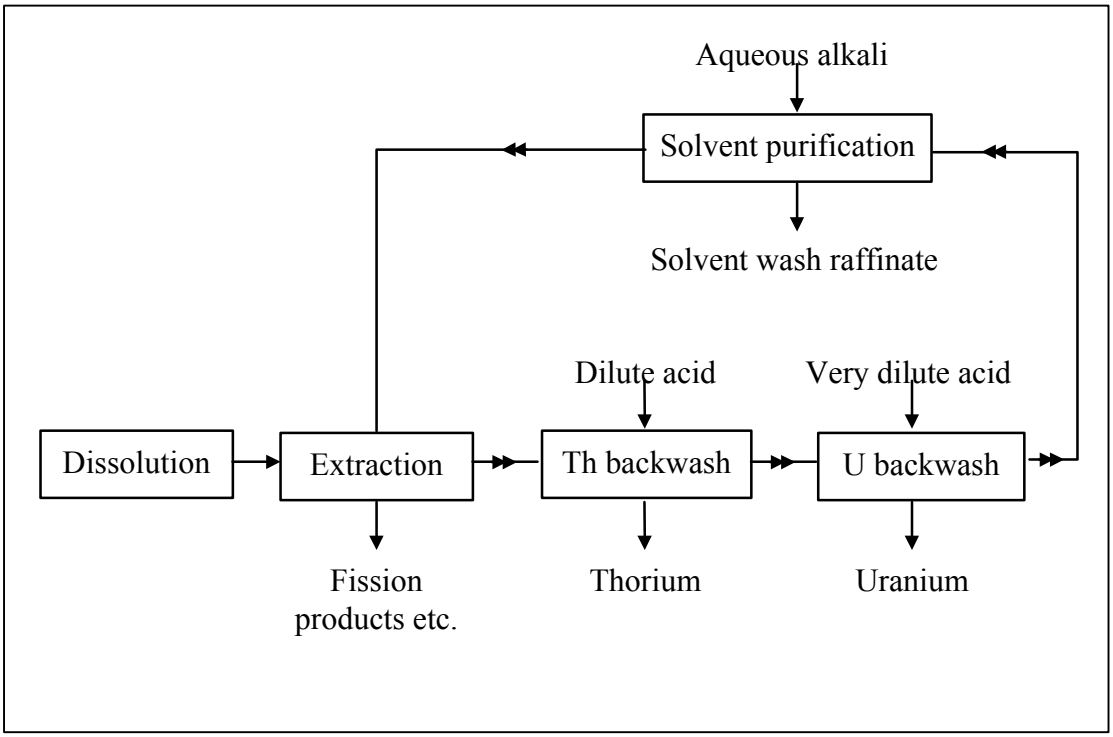

Figure 1. Outline of Thorex process.

The classical Thorex process, whether feeding acid-deficient fuel solution as in the original version or the later acidic alternative, may be illustrated by the outline flowsheet of Figure 1. Essentially, thorium and uranium are extracted away from the bulk of the fission products by the TBP-based solvent, from which first the thorium is backwashed with dilute acid, and then the uranium with acid still more dilute; the solvent is washed with alkali to remove degradation products such as dibutyl phosphoric acid and remaining traces of extracted metals before recycling to the extraction stage. Parallels with the Purex process used to process uranium-plutonium fuels are obvious, but there are significant differences:

- There is no equivalent to the valency changes used to enhance the separation of plutonium from uranium, so that in Thorex the separation depends solely on the difference in extractability.

- This difference is substantial; thorium, here the major component, is an order of magnitude less extractable than uranium, [8] so that the concentration of free extractant in the solvent must be kept higher than in Purex by a considerably lower loading.

- The extracted thorium complex is less soluble in the hydrocarbon diluent than is the equivalent uranium complex, [9] so a further limitation is to avoid forming two distinct organic phases that would prevent proper functioning of the solvent-extraction equipment.

- The low solvent loading prevents utilising the “squeeze” effect to restrict the co-extraction of certain fission products. However, the resulting relatively poor decontamination may be tolerable, since the products are likely to need remote handling for other reasons described below.

- More seriously, the dimensions needed to accommodate the greater solvent flow could require more elaborate measures to ensure nuclear safety than in an equivalent Purex plant.

- Furthermore, the fissile element is the more rather than the less extractable component, so that the safety case cannot be a straightforward extrapolation from that for Purex. In particular, there is a greater risk that fissile material might accumulate in the alkaline solvent wash system, which because of the flows required must be relatively large.

Difficulties connected with the poor extractability of thorium could of course be avoided by extracting only the uranium, although besides abandoning a rather valuable material, this would perhaps more seriously mean increasing the volume of waste. Not only the recycling process itself needs to be considered; the loss of material would have to be made good by additional mining, which in producing particularly large volumes of waste, incurs the greatest environmental penalty of the whole cycle - a serious consideration with uranium, [10] for thorium comparable in terms of radiation dose to individuals, [11] and chemically probably more troublesome, given the harsh treatment needed to break down the ore. This consideration should not be overlooked simply because the operation tends to be geographically remote from the rest of the cycle, particularly when the wastes are allegedly more radiotoxic than those from uranium production.

The thorium cycle is especially likely to be considered in relation to a programme of plutonium incineration, where the presence of a third component with intermediate extractability would greatly complicate the separation process unless a mixed product were acceptable. Even if it were, conditions suitable for backwashing uranium from the solvent without precipitating plutonium could be hard to find and still harder to ensure to the satisfaction of regulatory bodies.

A three-stage backwash is probably the simpler solution. Considerable cross-contamination of products could be acceptable, although it would reinforce the argument for applying to the major (thorium) stream precautions against criticality otherwise inherently necessary only for flows an order of magnitude lower.

Criticality problems could be eased by using intensified contactors such as centrifugal mixer-settlers with their short residence times, provided that the risk of blocking orifices could be effectively eliminated despite the tendency of thorium to form precipitates with solvent degradation products, [12] said to be greater than with uranium. Solvent degradation should of course be reduced in proportion to the residence time.

Other differences between the cycles, less directly related to the process, are that:

- Although the fission-product spectrum of ^{233}U has a similar general form to that of ^{235}U or ^{239}Pu, there is a slight shift so that on the lower slope, the yield of the mass-85 chain is roughly two to four times greater. [13] In this chain krypton appears first as the short-lived Kr-85m, most of which decays directly to stable rubidium-85, but some undergoes internal transition to the 10.7-year ^{85}Kr. In a borderline case, the increase in this radiologically significant isotope might tip the balance towards having to remove it from off-gases before discharge, and so add appreciably to costs. The consideration is unlikely

to be crucial, particularly if krypton removal becomes an unconditional requirement, but it could be a relevant factor.

- An important complication to the thorium cycle is that a small but significant proportion of the ^{233}U undergoes an (n,2n) reaction to form ^{232}U, also formed by the sequence.

  This nuclide decays with a 70-year half-life to ^{228}Th (half-life 1.9 years), then by way of

$$\text{Th-232} \xrightarrow{n,2n} \text{Th-231} \xrightarrow[25.5\ \text{hr}]{\beta} \text{Pa-231} \xrightarrow{n} \text{Pa-232} \xrightarrow[13\ \text{days}]{\beta} \text{U-232}$$

  radium-224 and other short-lived intermediates (Figure 2) with a minor branch to thallium-208 which has an unusually penetrating beta-gamma emission. Thus both thorium and uranium products would require remote handling within a few days after separation; hence the lack of concern for thorough decontamination from fission products.

- This fact implies a need for heavier shielding than for uranium-plutonium fuel during refabrication into new elements, transport, and storage at the reactor site. Refabricating facilities would have to be dedicated for the additional reason that the risk of cross-contamination would rule out sharing with uranium-plutonium fuel. Otherwise these operations would be much the same in both cycles.

5. EXAMINATION OF CLAIMED ADVANTAGES

In view of the problems to be expected in the practical implementation of a thorium cycle, the extent and reality of advantages claimed for it need to be evaluated very carefully. Taking in the order of those mentioned in the introduction:

(a) Producing no plutonium

This is true of the pure thorium cycle such as might eventually be established simply to make up for a failure of uranium supplies. However, that is not the reason most strongly advocated for favouring thorium at present, and a hybrid cycle with uranium or plutonium as driver is more likely at least for the foreseeable future, indeed virtually essential to start the cycle. Plutonium production would certainly be reduced considerably and might on balance become negative, but whether and how far that would be an advantage depends very much on a subjective rather than a technical view of the element (c and d below).

(b) Avoiding long-lived alpha-emitters in waste

Again this is true only of the pure thorium cycle. With plutonium present, transplutonium actinides would inevitably be formed. In any case, transuranic elements are by no means the only long-lived component of nuclear waste. Eliminating alpha-emitters would not be the decisive improvement to long-term safety that it might appear if fission products were neglected.

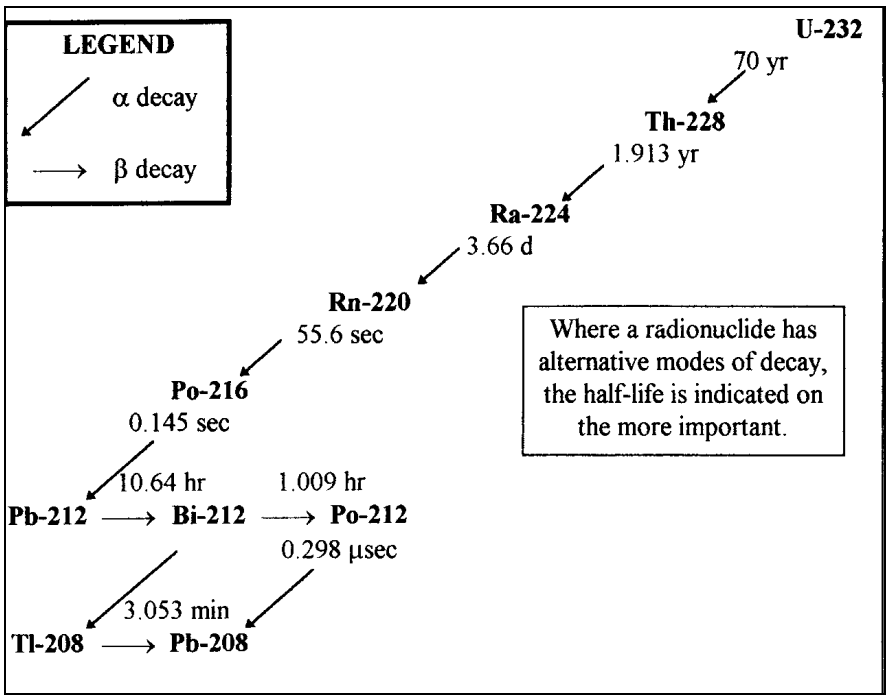

Figure 2. Part of the "4n" decay series of heavy nuclides.

(c) Rate of consuming plutonium

Plutonium could evidently be consumed considerably faster if driving a thorium breeder than with further generation from ^{238}U. How far that may be an advantage, when plutonium could meanwhile become a prized commodity to fuel fast reactors, depends on the view taken of the future.

A distinction sometimes overlooked is that between stockpiled and utilised plutonium. Thorium fuel with plutonium as initial driver would utilise a certain amount, providing an effective safeguard against misuse until the remainder were reprocessed or discarded. Conventionally fuelled fast reactors generating the same power would at first quarantine several times that amount from the stockpile. The longer-term pattern would depend on whether such reactors were operated in breeding or burning mode and so on the currently perceived balance of advantage, to which the fast-reactor option may thus be more readily responsive.

(d) Proliferation resistance

Although pure ^{233}U appears to be usable for nuclear weapons, the isotope produced by a thorium breeder is sometimes claimed to be proliferation resistant because the decay products of the concomitant ^{232}U would create a radiation hazard sufficient to require remote handling within a short time after chemical separation. We imagine that this is almost certainly enough to rule out its military use by the Nuclear Weapons States, and they already appear to have enough of the more conventional weapons materials (highly-enriched uranium and weapons-grade plutonium) for their present or probable future purposes; indeed, a more immediate preoccupation in Russia and the USA is how to manage the surplus stocks which come from dismantling weapons.

On the other hand, terrorist groups or rogue states, who not conspicuously concerned for their own or anyone else's personal safety, would be likely to want fissile material for immediate use rather than for stockpiling. In such circumstances, ^{233}U could be decontaminated from decay products for a few days' relative freedom from troublesome radiation. Whether it would be much less attractive to them than plutonium is thus at least open to question, supporting the international view that ^{233}U requires the same level of safeguards oversight and physical protection as does plutonium.

6. DISCUSSION

The various problems in applying the thorium cycle on a commercial scale may be amenable to technical solutions, given sufficient effort and determination. Whether the costs of that effort or of subsequent operation would be commercially acceptable is another question altogether and could not be answered with confidence until the development was largely complete, i.e. until a very considerable cost had already been incurred. A provisional judgement is therefore necessary on the information available before commitment to any substantial tranche of that development.

Much work has already been done on the necessary reactor physics, while fuel of conventional form can apparently be manufactured according to established principles and methods (subject to the proviso already noted on handling recycled material) with relatively slight adaptations. Reprocessing is the weakest area, with the following topics needing special attention:

- Dissolution; conditions necessary and rates realistically achievable. Fluoride is assumed to be the reference catalyst, but a less troublesome alternative might be sought.

- Corrosion, not only in the dissolver but elsewhere in the process equipment where fluoride might cause damage. Since hydrogen fluoride is volatile, while the customary aluminium palliative is not, this could well mean throughout the plant. Vapour spaces and off-gas lines in waste-treatment facilities may be particularly vulnerable. The requirements of development are (a) tolerably inexpensive materials and methods of construction capable of resisting the various liquors and vapours under working conditions, besides of course being sufficiently robust and stable to radiation, and (b) accelerated but otherwise realistic corrosion trials to demonstrate the required durability under such conditions.

- The safety case, in particular an assurance against criticality. As indicated earlier, this must be more than a mere extension of that for the uranium-plutonium cycle and is liable to be more difficult. In order to avoid the need for extremely expensive and time-consuming physical trials of every credible variation or maloperation in the process, the most limiting must be identified for detailed study. In current practice, this is done largely by means of computer simulations based on a mathematical model. To this end, the minimum requirement is a set of correlated distribution measurements covering the whole range of compositions that could be met, on a mesh fine enough for confident interpolation. It must locate the boundary surface of third-phase formation where the loaded solvent splits into solute-rich and diluent-rich fractions. If the process is sensitive to kinetics as well as to equilibrium conditions, the hydraulic and mass-transfer properties of the contactor must also be represented, greatly increasing the necessary amount of data,

the effort in obtaining it, the complexity of experimental equipment and the difficulty of modelling.

- The effect of fluoride on waste treatment and the resistance of ultimate forms to environmental leaching will have to be established satisfactorily.

The technical problems are real and tangible. The advantages are largely subjective, in so far as they depend on a commonly adverse perception of current nuclear practice. That is not to say that they are unimportant; a sufficiently aroused public opinion might close down the entire industry within its area of influence, as is the declared aim of some pressure groups. On the other hand, such groups tend to be motivated by a fundamental hatred of nuclear energy in principle rather than in its particular manifestations, and not always intellectually consistent or scrupulous. Although much of their current propaganda is created by exaggerating the dangers of plutonium, and they may tactically favour thorium so long as its use remains hypothetical, they are unlikely to be appeased by its adoption. Indeed, wherever a new installation or system of whatever kind is proposed, they are sure to pick upon any real or apparent weakness in the industry's case that suits their immediate purpose. It is therefore imperative that the technical and logical basis of the case should be as robust as practicable.

That applies whether the preference is for action or inaction. Accordingly, even if the merits of using thorium were believed on the present state of knowledge to be more apparent than real, some further study of the problems, possible solutions, likely costs and benefits over the whole cycle could be expedient, at least as a defensive measure. The work should then be restricted in the first instance to the crucial issues, with a view to assessing whether a full-scale development programme followed by industrial implementation could be expected to recoup its costs on an acceptable time-scale, or yield benefits of equivalent value in other terms.

To serve its defensive function, the process of assessment would have to be made known publicly, with some inevitable risk of giving an over-optimistic impression of its prospects that could add to the difficulties affecting current or intended operations of the industry. Care would be needed to minimise this risk by judicious presentation.

As an eminently pre-commercial project, the assessment would be a fitting topic for international co-operation. With its recent experience in developing and building the thermal oxide reprocessing plant and the MOX fuel fabrication plant (SMP) at Sellafield, and its interest in more radical alternatives for the future, BNFL is well placed and could be willing to participate in such a study.

7. CONCLUSION

Thorium as a nuclear fuel is technically well established, yet it has never been widely adopted, presumably because it is judged to be commercially uncompetitive with uranium. One reason is that its potential as a thermal breeder cannot be realised without reprocessing, which is subject to severe outstanding difficulties. A re-examination of the thorium cycle is required so that the advantages, problems and possible solutions could be realistically assessed under current conditions. In view of the commercial risks attached to a full-scale development programme, prospects of success should be assessed clear-sightedly before any major commitment. This assessment, with a limited supporting programme of experiment, could appropriately be an international project.

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ON SOME ISSUES OF THORIUM FUEL CYCLE STUDY*

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Abstract. In conceptual aspect, various reactor systems have been under study. They include PWRs fueld with Pu-Th and ^{233}U-Th, molten salt reactors, fast reactors with mixed Pu-^{233}U -Th ^{238}U fuel cycle. Burning of plutonium in thorium matrix makes it possible to convert plutonium into ^{233}U. In the long term, ecological hazard of ^{233}U thorium fuel cycle may appear to be lower, compared to uranium-plutonium one. ^{233}U plus ^{238}U blend may be good fuel for thermal reactors. Considerable efforts have been concentrated on studies of initial steps for mastering thorium cycle. In this connection, investigated were issues such as ^{233}U accumulation in thorium in various neutron spectra, methods for obtaining ^{233}U with ^{232}U levels acceptable for its simpler handling. A series of such experiments was carried out with different reactors. These experiments have confirmed the probability o initial accumulation of ^{233}U in fast reactors in the form allowing a simpler subsequent handling. This is important for experimental works on mastering the technologies of ^{233}Th fuel cycle. The program includes also the development and tests of technologies for radiochemical separation of ^{233}U from thorium, technological studies with samples of thorium and^{233}U. Experiments with critical assemblies and nuclear constant measurements are also a part of the programme for thorium and ^{233}U studies.

1. INCENTIVES FOR THE APPLICATION OF THORIUM FUEL CYCLE

The accumulation of new facts and information on the development of thorium-based fuel option is not very fast as the attitude towards this problem in many countries is not of top priority for financial support.

The material provided in [1] and [2] reflects the state-of-the art in thorium fuel cycle in Russia. Some additional considerations and results are presented in this paper.

Speaking about incentives for an application of thorium cycles, it would be appropriate to point out that thorium insertion into nuclear power plants enables the LWRs life to be prolonged; and experience accumulated, and industrial technology mastered, to be used more effectively.

It accounts for the fact that the use of thorium cycle in its different options results in favorable changes in passive safety. The application of new fuel compositions reducing the fuel temperature will also contribute to this.

An understanding of these prospects in Russia resulted in the Arsamas-16 Research Weapon Centre being set to solving the thorium problem [10].

In discussing the incentives for the application of thorium cycle, the following consideration could be pointed out:

In an advanced nuclear-power fuel complex with different reactor types, facilities on the production and reprocessing of spent fuel mixed and combined cycles are possible. For example ^{233}U build-up may be ensured in fast reactors; while for thermal ones plutonium and

* 1997 meeting.

HEU may be converted into ^{233}U. Combined use of ^{233}U and plutonium in one reactor may be considered beneficial.

At the same time, the high radioactivity of ^{232}U may be an obstacle for unauthorized use of nuclear material.

Many mixed fuel cycles are yet to be studied in details.

2. STUDY OF CONCEPTS

Some new results from different concept studies have recently been added to those given in "Working Materials" [1].

Thorium fuel cycle with the composition of metallic thorium and highly enriched weapon-grade uranium oxide is initiated for utilization in light water reactors [3]. Highly enriched uranium need not be diluted to low concentrations in this case. The WWER-1000 has been taken as an example to evaluate this idea.

Owing to the physics properties of accumulated ^{233}U, and to the low temperature of the fuel composition; characteristics of this reactor on fuel use, reactivity effects, and safety change to the better. A lot of details of the analysis are given in the reference.

Joint Russian-American proposal on a WWERT reactor of "seed and blanket" type with thorium blanket in each fuel assembly [4] was reported at the 8-th ICENES Conference.

The thorium part is to be used as a dioxide and not subject to reprocessing after unloading from a reactor. Central parts of each fuel assembly are proposed to be made replaceable irrespective of thorium. Uranium-zirconium metallic alloy is used there. This alloy has been studied extensively and applied in reactors.

The idea is to create a reactor without plutonium accumulation and, with better resistance against the risk of nuclear material proliferation. Reactor technologies assimilated are stored at the same time.

The Kurchatov Institute, the Institute of Inorganic Materials, the plant in the town of Electrostal (Electrosteel) from the Russian side; and the Radkowsky Thorium Power Corporation, and BNL from the USA side, are taking part in the work.

Conceptual developments on thorium fuel cycle in other reactor types (MSR, HTR) are being continued.

3. INTEGRAL PHYSICAL EXPERIMENTS

Estimated nuclear data files for the thorium cycle are not as validated as for the uranium cycle. To test and correct them one must carry out integral experiments on critical assemblies and reactors.

IPPE is gradually realizing such a programme. Experiments on several critical assemblies with various neutron spectra have been carried out recently. The main trend is to test the neutron cross-section capture with thorium as well as obtain data on other reactions.

Experiments with insertion of various material sets were carried out at the KOBR assembly. The first experiment was undertaken for obtaining the critical ^{235}U and thorium ratios in an infinite medium [5].

Later, media with various spectra, from thermal to tight water lattice spectra; were studied in this same assembly.

A set of preliminary experimental data on these assemblies is available, but their evaluation has not been completed due to difficulties with processing. Cooperation with specialists from other countries could largely contribute to obtaining final results.

Prolonged irradiation of thorium samples with the following ^{232}U content measurements in ^{233}U were carried out at BN-350 reactor [5]. Irradiated samples analysis confirmed the ^{232}U content in the outer radial blanket of 2 to 11 ppm, with uranium content in thorium being ~1,3 g/kg.

Average cross-sections of several isotopes essential for thorium cycle were measured in BN-350 reactor core. ^{231}Pa cross-section capture was measured as well. These measurements showed the current isotope cross-sections need are to be made more precise [6].

The validity of nuclear data for thorium cycle may be expected to improve appreciably after completing the processing and analysis of available integral experiments.

4. TECHNOLOGICAL DEVELOPMENTS

Technological research and developments on fuel fabrication for thorium cycle showed the Russian specialists the need to make new long-term decisions.

Technology for fabrication of mixed uranium-thorium-oxide pellets was tested by different groups of specialists as the first step. Experimental fabrication of compact mixed uranium and thorium oxide pellets was carried out at IPPE. The experiment on technology of fabrication and properties of pellets was a success.

Kurchatov Institute together with the plant and Institute of Inorganic Materials conducted more thorough work. To fabricate uranium-thorium mixed oxide pellets, the whole technological cycle and tooling were developed. An experimental set of pellets was fabricated, quality investigations were carried out. The result was successful. This work was done for the blanket parts of WWERT fuel assembly [4].

IPPE technologists propose various dispersional fuel compositions for thorium WWER of traditional constructions, but with a new level of safety. This fuel with a matrix high heat conductivity has a low temperature with a small level of accumulated heat. As a result reactors with this fuel will achieve a higher level of safety [7].

The development of dispersional uranium composition with a metallic matrix for WWER reactor types is being carried out in Russian institutes. Large work on substantiation of UO_2 (60% vol.) - Zr (40% vol.) alloy and UO_2 - Al(silumin) alloy [8] pellet compositions has been done for some years in IPPE. The authors of these developments think UO_2 could without essential difficulties be replaced for ThO_2 - UO_2 mixture. Cermet compositions were tested in

a set of experiments as well as in pile tests of experimental pins with man-made failed claddings.

Another kind of dispersional fuel with an extended pyrographite matrix [9] has been proposed as well.

A composition with 50% vol. oxide particles (thorium, uranium, plutonium) and 50% pyrographite is proposed. A graphite matrix high heat conductivity will ensure a low fuel temperature and affect safety performance favourably. Unfortunately, complex tests of this concept have not been carried out so far.

Any thorium oxide compositions will pose some problems in reprocessing spent fuel with water extraction techniques. Dry electrochemical reprocessing may seem to be in prospect. Some experimental tests with thorium oxide compositions seem to be quite promising.

However, in some concepts of thorium insertion into power reactors, reference [4], thorium oxides have not been offered for reprocessing - they must be disposed of.

Russian technologists have not considered metallic thorium compositions so far. At the same time a cermet based on thorium metallic matrix for thorium cycle in any reactors could be a good decision, in fuel reprocessing, in particular. So far in Russia physicists, not technologists, are dealing with this composition.

5. SOME GENERAL REMARKS.

Taking into consideration that accumulation and preservation of new information takes a lot of time it would be useful to extend and specify of what has already been done and given in published documents.

It might be expedient to hold international meetings of specialists from various countries on some issues of thorium cycle, for example, nuclear data and their verification, technology of fuel production based on thorium without ^{233}U and with ^{233}U, possible technologies of fuel reprocessing and so on.

Based on these extended reviews on particular questions it might be necessary to organize a more extensive international discussion.

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THE NON-PROLIFERATIVE COMMERCIAL RADKOWSKY THORIUM FUEL CONCEPT*

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Abstract. Thorium is found in large quantities and is plentiful available throughout the world. Today almost all existing pressurised water power reactors as well as boiling water power reactors can be retro-fitted with the novel seed-blanket core called the "Radkowsky Thorium Fuel Core". Because the new thorium-based cores fit into the same space as the current uranium cores, no or only minor changes to existing operating nuclear plants are necessary. "Radkowsky Nonproliferative Light Water Thorium Nuclear Reactor Concept" was developed and is under test in the USA and in Russia. Advantages of the RTF nuclear reactors may lead to a change of the present public opinion against nuclear power bringing about a better understanding of the benefits of nuclear fission for energy production.

1. INTRODUCTION

The Non-Proliferation Nuclear Arms Treaty (NPT) originally entered into force in 1970. Meanwhile 178 countries of the United Nations have extended the Treaty indefinitely. The Strategic Arms Reduction Talks (START) by the Super-Powers came to a conclusion in the middle of 1991. As a consequence Russia is dismantling approx. 30 000 and the USA about 15 000 nuclear warheads. Therefore vast amounts of weapon material in form of plutonium (260 tons) and highly enriched uranium (1 000 tons) will become available for possible use in the peaceful civil fuel cycle. In addition, by the year 2 000 reactorgrade plutonium of about 1 000 tons will have piled up. To make economic use of the weapon materials the so-called „Radkowsky Nonproliferative Light Water Thorium Nuclear Reactor Concept“ was developed and is under test in the USA and in Russia. The nonproliferative reactor is of a seed-blanket design. Initially the seed used nonproliferative enriched uranium and the blanket, which acts as a converter, is fed by ThO_2. In an alternative plutonium-burner version the seed will be plutonium in form of plutonium-zirconium alloy. The need of cutting the vicious circle of producing more nuclear weapon materials in existing conventional uranium power reactors stands vis-à-vis the consequences of a global nuclear exchange.

A number of seed-blanket units make-up the new thorium power reactor core. The heterogeneous seed-blanket unit forms two separate regions. The outer blanket mainly consists of thorium, while the inner region is made-up of nonproliferative less than 20 % enriched ^{235}U. When the seed material undergoes fission neutrons are produced and after being slowed down will create in the blanket ^{233}U which is also fissionable and therefore can take over the necessary neutron supply and reactivity in due time. The actual blanket consists of rods of thorium oxide which in stoichiometric form can withstand very high burn-ups and thus allow the blanket to remain in the reactor for nearly 10 years. The seed material will use metallic fuel in the form of U/Zr and/or Pu/Zr-alloy designed to allow an efficient thermalising effect of the neutron spectrum. The seed material will have to be replaced more often and needs reshuffling similar to conventionally fuelled uranium reactors. If plutonium is used instead of enriched uranium as seed material, the system will act as a plutonium burner. The Radkowsky thorium core design utilises established and proven light water nuclear reactor technology.

* 1999 meeting.

One of the advantages of the new fuel concept is a nearly full-proof nonproliferative reactor core. Conventional light water reactors produce about 50 times as much plutonium compared with the Radkowsky thorium fuel. Since the costs of uranium are appreciably higher than the costs of thorium, the economy of existing reactors will be improved. The amount of radioactive waste is drastically reduced both in quantity and toxicity as well as in radioactivity and heat emission. By implementing the thorium ^{233}U conversion route the production of high level actinides will be avoided. The nonproliferative system is especially suitable for a once-through fuel cycle with no reprocessing requirements. The long residence time of the blanket permits burn-ups of the order of 100 GWd/t. Means can easily be provided for instance by admixing to the blanket denatured uranium to hinder attempts to extract and use ^{233}U for non-peaceful purposes.

Of particular interest for implementation are approximately 210 PWRs with a net electrical power of over 200 000 MW(e). Thus a majority of the world's nuclear power reactors could benefit from the thorium concept including some 55 Russian-type WWER-reactors operating and under construction in a number of countries.

All the advantages above may lead to a change of the present public opinion against nuclear power bringing about a better understanding of the benefits of nuclear fission for energy production and putting in proper perspective questions of economy, safety, waste management as well as reduction and elimination of nuclear weapon materials.

2. THE PAST AND THE FUTURE OF THE NON-PROLIFERATION TREATY (NPT)

Since 5 March 1970 a Non-Proliferation Treaty (NPT) is in force. At that time the three depositories Soviet Union, United Kingdom and the United States of America together with forty non-nuclear weapon countries signed the agreement in London.

The NPT-concept was to curb the spread of nuclear warheads to Non-Nuclear-Weapon States and to reduce the number of nuclear weapons of the superpowers and Nuclear-Weapon States. The Treaty was in conformity with a number of previous resolutions of the United Nations General Assembly dating back as far as 1946 and warning of the devastation all mankind would experience by a nuclear conflict. It was also believed that the proliferation of nuclear weapons would seriously enhance the dangerous possibilities of starting-off a nuclear exchange warfare.

To all signatories it was evident that the NPT had political, economic and technical aspects. Therefore in the preamble the principle was affirmed that the benefits of peaceful applications of nuclear technology derived by the Nuclear-Weapon States, when developing nuclear explosive devices, should be made available for peaceful purposes to all parties of the Treaty.

Eleven articles define the Non-Proliferation Agreement. Article 1 spells out that all Nuclear-Weapon States do not transfer to any recipient nuclear weapons or explosive devices directly or indirectly and do not in any way assist, encourage or induce any Non-Nuclear-Weapon State to manufacture or acquire nuclear weapons. Article 2 makes it clear that Non-Nuclear-Weapon States should not undertake to receive and/or control nuclear weapons directly or indirectly. Article 3 defines a safeguards system, which each Non-Nuclear-Weapon State should negotiate and conclude with the International Atomic Energy Agency (IAEA). The IAEA safeguards apply to virtually all nuclear materials in facilities outside the five declared Nuclear-Weapon States China, France, Russia, UK, USA. Also a number of peaceful nuclear installations in the five Nuclear-Weapon States are covered in form of voluntary agreements.

All declared Nuclear-Weapon States are now party to the NPT, while only three started out twenty-five years ago. Articles 4 and 5 refer to non-discrimination of production and use of nuclear energy for peaceful purposes, particularly in the Non-Nuclear-Weapon States. Article 6 says that all parties of the Treaty including the Nuclear-Weapon States should pursue negotiations relating to the cessation of the nuclear arms race and undertake nuclear disarmament efforts under effective international control. The last four paragraphs define possible amendments, the mode of accession for countries at a later stage and in Article 10(2) it is explicitly stipulated that twenty-five years after the entry into force of the Treaty a conference shall be convened to decide whether the Treaty shall continue in force indefinitely, or shall be extended for an additional fixed period or periods. The decision shall be taken by a majority of parties to the Treaty. This Review Extension Conference was held in New York from 17 April to 12 May 1995. In the course of the negotiations it became evident that a solid majority of the 178 participating countries was in favour of an indefinite extension. On the other hand 15 countries under the leadership of Indonesia, Egypt, Nigeria, Malaysia, Syria, Libya, Iran and North-Korea were driving for a time-limited extension, while a number of other non-weapon countries tried to connect their votes for indefinite extension with an agreement for further disarmament measures and security guarantees to be given by the 5 nuclear weapon countries.

A continuing global nuclear arms control now and a nuclear weapon-free world in the future seem irrevocably necessary for the furtherance of the harmony of world cultures. Nevertheless a number of countries are not happy with the NPT as it stands, as it prohibits the possession by a great majority of states of one of the most destructive weapons yet invented and on the other hand tolerating the retention of the same weapons by a handful of nations. The inequality of treaty rights and obligations of the „haves“ and the „have nots“ has nevertheless brought a record number of adherence for an arms control agreement of about 169 nations.

The NPT regime requires nuclear trade restrictions. In particular developing countries are constantly complaining about export restrictions of nuclear materials and know-how. These obstacles can only be overcome, when exports are covered by clear and comprehensible rules, which both exporters and importers abide. Experience showed that trade possesses a non-vanishing risk. The present discussions concentrate on greater international harmonisation of nuclear export rules and regulations, but also measures are discussed for rules of punishment, when countries try to cheat. The IAEA will need more authority to report immediately any violation to the Security Council of the United Nations. IAEA safeguards are applied under the terms of agreements concluded between the International Agency and their member states.

These agreements are generally concluded in connection with bilateral nuclear cooperation and supply agreements. At the end of 1991 180 safeguard agreements with 105 states were in force. During 1991 IAEA carried out 2145 inspections at 475 nuclear installations in 56 countries. More than 360 photographic and video-surveillance systems were in operation and approximately 1400 seals previously applied to verified amounts of nuclear material were detached and subsequently verified. About 1100 plutonium and uranium samples were analysed at the Agency’s Seibersdorf laboratory. At the end of 1991 as a result of severe budgetary constraints the Agency experienced difficulties in maintaining a safeguards program at an acceptable level of effectiveness.

The Nuclear-Weapon States agreed thirty years ago to pursue comprehensive disarmament negotiations. Many countries feel that this promise has not been fulfilled. Unfortunately a number of so-called „threshold states“ are still outside of the NPT. Their arguments are of political nature mainly and they condemn the discriminatory character of the Treaty. In reality

there are always at least two countries who mistrust each other and fear that their counterpart might acquire nuclear weapons and therefore arguing they need also nuclear explosives for counterbalancing.

Leaders for a nuclear-free world will be definitely the USA and Russia, the Western industrial world and the neutral and non-aligned countries taking advantage of the current peace situation to reinforce the already existing constraints. Over the last twenty years intensive negotiations took place between the Nuclear-Weapon States to arrive at a comprehensive test ban (CTB) agreement. Unfortunately a number of states deeply regret that the comprehensive multilateral nuclear test ban treaty banning all nuclear tests by all states in all environments for all time has not been concluded so far and this fact was brought forward at the New York extension negotiations of the NPT.

3. STOP AND RUN-DOWN OF THE NUCLEAR ARMS RACE

Strategic Arms Reduction Talks (START) between the USA and USSR began in 1982. Negotiations were aimed for to reduce the strategic nuclear forces on both sides. The talks were suspended in 1983 and opened again in Geneva in 1985. The START-Treaty was finally signed in Moscow on 31 July 1991 at a summit meeting between President Bush and President Gorbachev. The Treaty between the United States of America and the Union of Soviet Socialist Republics on the Reduction and Limitation of Strategic Offensive Arms is formulated in nineteen articles and a number of statements and exchanges of letters annexed. At the end of January 1992 the United States and then Russia already announced additional unilateral nuclear arms control measures.

In the preamble of the START-document the two parties stated their consciousness that nuclear war would have devastating consequences for all humanity and that it cannot be won and must therefore never be fought. They were also convinced that the measures for the reduction and limitation of strategic offensive arms will help to reduce the risk of outbreak of a nuclear war and strengthen international peace and security. Article 2 specifies the general reductions and limits.

Neither side may exceed a limit of 1600 Strategic Nuclear Delivery Vehicles (SNDV), which include Submarine-Launched Ballistic Missiles (SLBM), InterContinental Ballistic Missiles (ICBM) and heavy bombers. These delivery vehicles may carry no more than 6000 accountable warheads. A maximum number of 4900 warheads may by carried by ballistic missiles and no more than 1100 warheads by intercontinental ballistic missiles on mobile launchers. Article 3 defines the counting rules. Article 4 relates to the non-deployed missiles and non-deployed mobile launchers. Each side is permitted to have only 250 non-deployed Inter-Continental Ballistic Missiles. Articles 5 and 6 deal with basic prohibitions, particularly on the movement of deployed mobile systems. In Articles 7 to 15 the verification principle and the verification regime are defined. In Article 16 the Treaty prohibits either side to assume international obligations that would conflict with treaty provisions. Finally Articles 17 to 19 are concerned with the conditions for entering into force and future discussions of possible amendments. The Treaty will remain in force for a period of fifteen years and can be extended successively for five-year periods. Each party has the right to withdraw from the Treaty, if it decides that continued adherence to the Treaty would jeopardise its supreme interests. Withdrawal from the Treaty requires a six-month notice and a declaration for the reasons to withdraw.

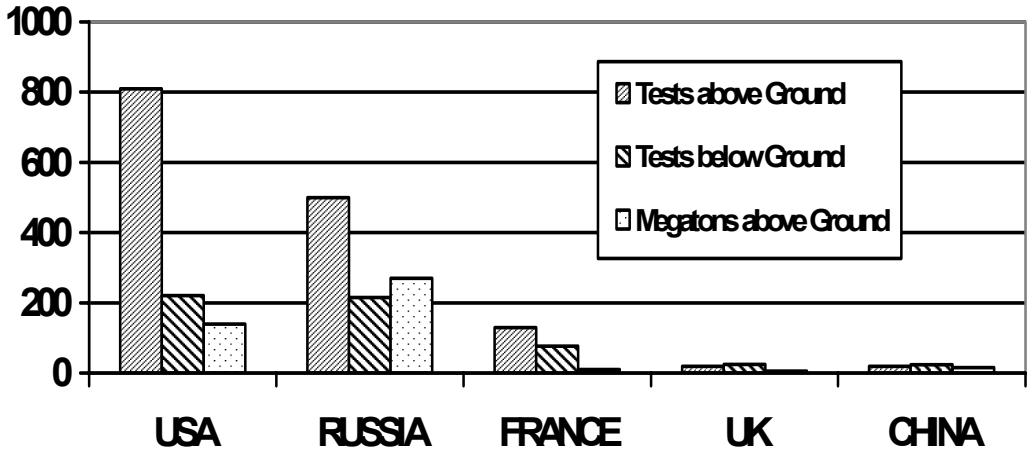

Figure 1 Number of nuclear tests 1945 - October 1999

In the unilateral statements reference was made to ban nuclear tests. Consequently the number of nuclear test explosions was drastically reduced, an overall agreement on a complete nuclear test ban does not exist yet. About 2040 nuclear test explosions in the atmosphere and underground were carried out since mid-July 1945. USA ranks first with 1030 explosions followed by the former USSR and its successor Russia with 715 detonations. Third is France with 207 test explosions followed by UK with 45 and China with 43 explosions. India has detonated one nuclear device described for peaceful ground excavation purposes. The total explosion power of all tests above ground amount to over 430 megatons of TNT equivalent (*Fig. 1)*.

Nuclear weapons world-wide totalling to about 60 000 warheads consist of fission weapons with ^{235}U and/or plutonium, of boosted fission warheads and of thermonuclear fusion bombs. SALT lays down a dismantling of approximately 30.000 Russian and about 15.000 US warheads within a period of seven years after entering into force of the Treaty. To meet with this time scale US is dismantling 2000 warheads a year, while Russia should dismantle approximately 4000 warheads per year. A number of treaties like NPT, Antiballistic Missile Treaty, and special United Nation's Sessions on disarmament are counterforces against nuclear wars, but nuclear weapon technology and production of fissionable isotopes could be acquired by some thirty countries in a relative short period of time.

4. WEAPONGRADE PLUTONIUM AND HIGHLY ENRICHED URANIUM (HEU) BOMB MATERIALS FOR THE CIVIL FUEL CYCLE

Originally weapongrade plutonium was produced in special military nuclear reactors. Highly enriched uranium for the first bomb was produced by electromagnetic isotope separators called CALUTRONS. Later on diffusion was the key method for enriching ^{235}U. Nowadays ultracentrifuges are in use as well as laser-induced separation methods. When nuclear power became an economic alternative possibility for electricity production, large quantities of reactorgrade plutonium were piling-up.

From energy and electricity data of the International Atomic Energy Agency in Vienna the nuclear power electricity production at the end of 1997 was about 16 % of the world's total electrical energy consumption (Fig. 2).

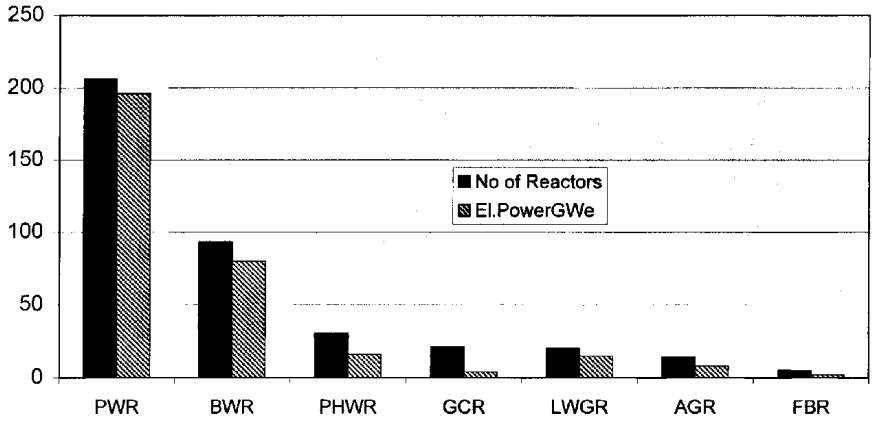

Figure 2. Reactors in operation and net electrical power in 1998.

This energy was supplied by 437 power reactors connected to the grid in 31 different countries including Taiwan; their total power amounted to 352 000 MWe. The nuclear share of electricity generation varies from Lithuania with 82 %, France with 78 %, Belgium with 60 % to former Eastern countries like Hungary and Bulgaria with roughly 40 % and 45 %, Germany with 32 %, Japan with 35 %, the United Kingdom with 28 %, USA with 20 % and Russia with 14 % (*Fig. 3*).

Country	**Nuclear Share (%)**		**Country**	**Nuclear Share (%)**
Lithuania	81,5 %		Armenia	25,7 %
France	78,2 %		USA	20,1 %
Belgium	60,1 %		Czech Republic	19,3 %
Ukraine	46,8 %		Canada	14,2 %
Sweden	46,2 %		Russia	13,6 %
Bulgaria	45,4 %		Argentina	11,4 %
Slovakia	44,0 %		Romania	9,7 %
Switzerland	40,6 %		South Africa	6,5 %
Slovenia	39,9 %		Mexico	6,5 %
Hungary	39,9 %		Netherlands	2,8 %
Japan	35,2 %		India	2,3 %
Korea Republic	34,1 %		Brazil	1,1 %
Germany	31,8 %		China	0,8 %
Finland	30,4 %		Pakistan	0,6 %
Spain	29,3 %		Kazakhstan	0,6 %
United Kingdom	27,5 %			

Figure 3. Nuclear share of total electricity generation during 1997.

437 reactors of nine different reactor types are in service, but only two contribute to electrical power generation considerably. These are the Pressurised Light Water Reactors (PWRs) and the Boiling Light Water Reactors (BWRs). 206 PWRs supply presently 195 990 MWe, while 93 BWRs contribute 79 800 MWe. Since the first demonstration of nuclear power began with operating the reactor in Obninsk near Moscow in 1956, the total reactor years experience until 31 December 1998 has reached over nine thousand reactor years. Altogether 80 reactors were shut-down and taken out of service in the past 30 years.

4.1. World plutonium stocks and surpluses

Plutonium is derived from two major sources:

(a) It comes from dismantled warheads as a consequence of the two disarmament agreements in force between USA and Russia.

(b) It is the result of commercial separation by large scale chemical reprocessing of spent uranium fuel coming from civilian nuclear power reactors.

There are some 60 000 tactical and strategic nuclear warheads stored in the world's nuclear arsenals. The agreed overall cuts between the two largest nuclear weapon countries to approximately six-thousand warheads each will result near to fifteen thousand warheads to be retired by the USA and roughly thirty-thousand warheads by Russia (Fig. 4).

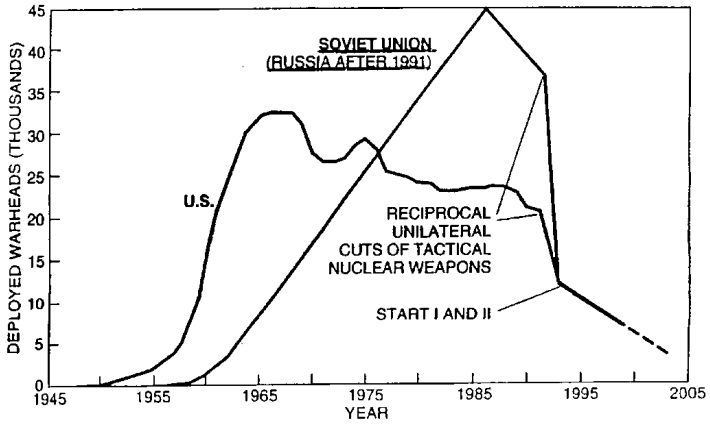

Figure 4. Number of warheads USA - Russia.

Advanced thermonuclear warheads contain about 15 kg fission explosives (highly enriched uranium HEU and/or plutonium), both in the primary and in the secondary compartment, the latter together with fusion fuel. It can safely be assumed that some 256 000 kilograms weapon grade Pu will be available and more than four times as much ^{235}U, when the warheads are dismantled. While ^{235}U can easily be used commercially by mixing it with natural U and thus blending it down, Pu can only be removed by nuclear reactions either by irradiation in nuclear power reactors or by explosion in warheads. As a somewhat unrealistic alternative, mixing plutonium with high-level radioactive waste has been proposed, for instance by vitrification and disposing it in proper geological structures. If, however, significant quantities of plutonium remain in whatever form stored on our planet, the risk of weapons proliferation will greatly increase and the diversion of only tens of kilograms of plutonium for criminal or subversive acts could cause crises world-wide. Reactorgrade plutonium can also be used for weapons purposes, even if it is not as effective as weapongrade material. With 7.5 kg of reactorgrade plutonium a bomb with an output in the kilotons TNT-range can be built.

At the end of 1990 the world plutonium stocks and surpluses had accumulated to about 900 tons, the major portion in the nuclear weapons countries, but roughly 175 tons in non-nuclear weapons countries. Some 20 tons of reactorgrade plutonium were accounted to countries, which had not signed the Non-Proliferation Treaty. Over 500 tons were at that time contained in irradiated fuel. The plutonium-stocks inventory increases year by year (Fig. 5) as shown in the cumulative amount of fissile plutonium in spent fuel alone.

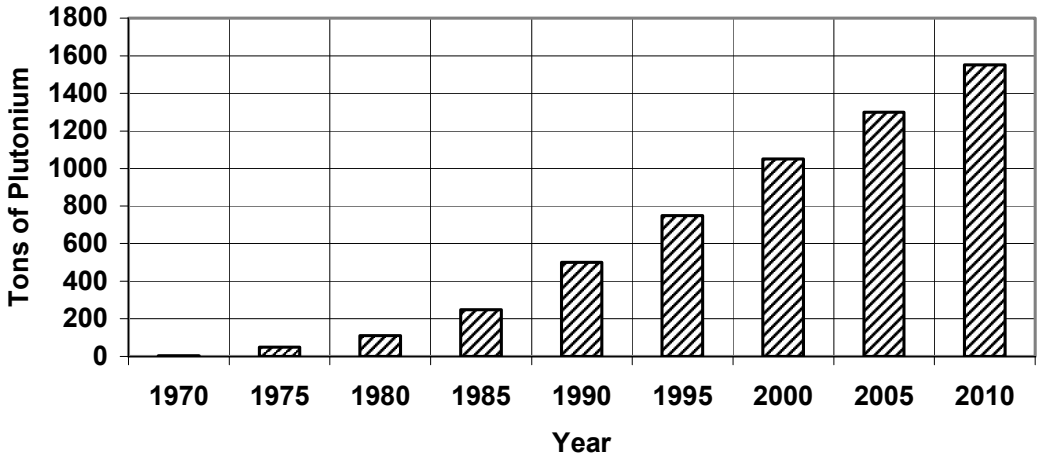

Figure 5. Cumulative amount of fissile plutonium in spent fuel.

The table shows for 1990 an amount of 531 tons in irradiated fuel. In 1995 about 750 tons will be in irradiated fuel and in the year 2000 one thousand tons of reactorgrade plutonium will have accumulated, if no Pu-burners are available. The civilian reactorgrade plutonium contains ^{240}Pu between 25 % and 33 % depending on the uranium fuel type. Weapon-grade plutonium has less than 6 % ^{240}Pu and some 94 % ^{239}Pu (Fig. 6).

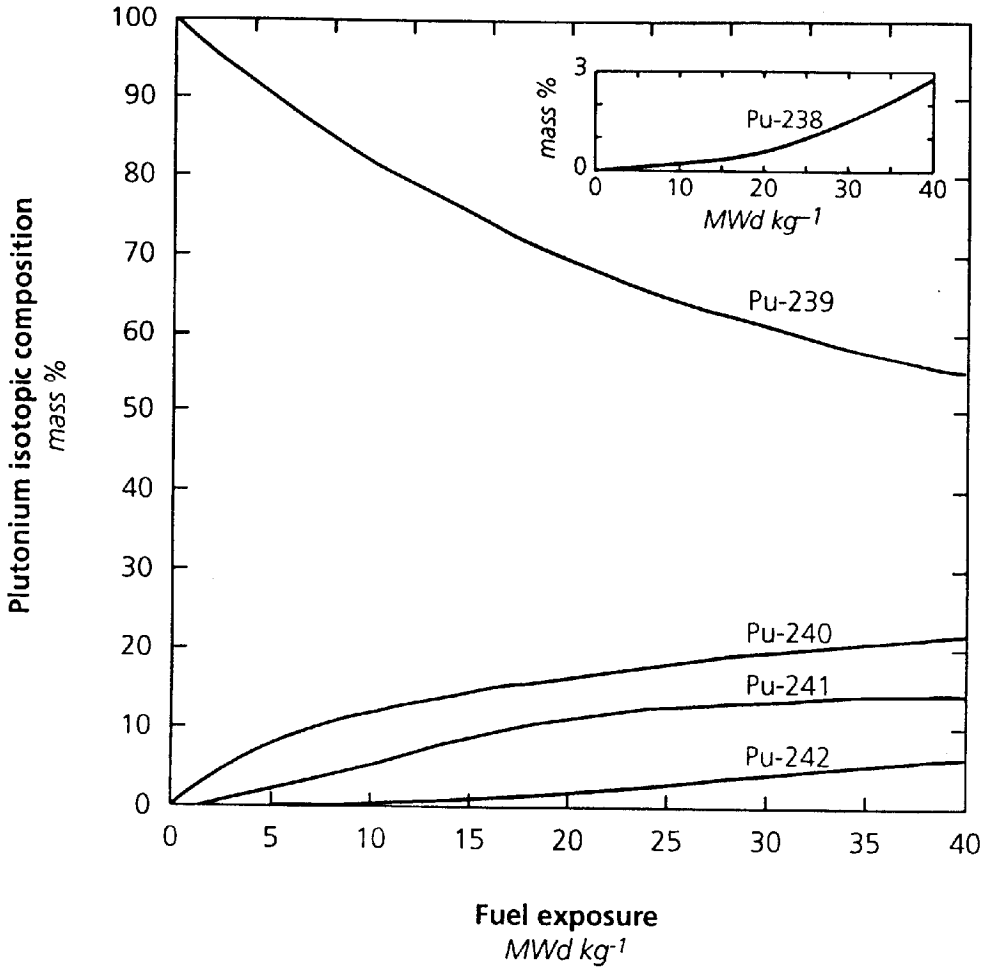

Figure 6. PWR plutonium isotope production as function of fuel exposure.

In the seventies the extraction of civilian plutonium by special separation plants was justified for the use in fast neutron plutonium breeder reactors. Unfortunately the experimental fast reactors encountered a number of serious problems and could in general not be operated safely. Therefore, with very few exceptions, the fast breeder reactor programs were terminated in several countries.

In order to make use of the existing plutonium stocks the use of plutonium as mixed oxide fuel (MOX) in light water reactors was suggested. Several important countries are engaged in Light Water Mixed Oxide Fuel Reactors on the basis PuO_2-UO_2. The method requires reprocessing and produces new plutonium and therefore cannot claim to be nonproliferative. Nevertheless this is an option the nuclear industry has chosen so far for the separated civilian plutonium, even if modifications in existing light water reactors are required.

4.2. The Radkowsky Nonproliferative Light Water Thorium Nuclear Reactor

There is a general consensus that proliferation of nuclear weapon materials should not be tolerated. Legally, however, the non-NPT-countries are free to obey the NPT-rules or not. On the other hand the UN Security Council stated at several occasions that proliferation in all of its aspects is bad and should not be allowed. The concern about a massive increase of nuclear weapon materials arises from the fact that an 1000 MW(e) light water nuclear power reactor produces approx. 200 to 300 kg plutonium in its core annually.

Therefore the present 299 light water reactors with some 276 000 MW(e) power have produced 56 000 kg plutonium in the year of 1998. This amount would be enough to feed additionally some four to five-thousand nuclear weapons in the 100 kilotons range.

In order to overcome these problems A. Radkowsky proposed a new approach to reactor core design under the boundary condition of utilising proven light water technology and thorium.

The core should be suitable to replace present cores in light water reactor systems with none or minor alterations only. The design became known as „Radkowsky Thorium Power Reactor“ (Fig. 7).

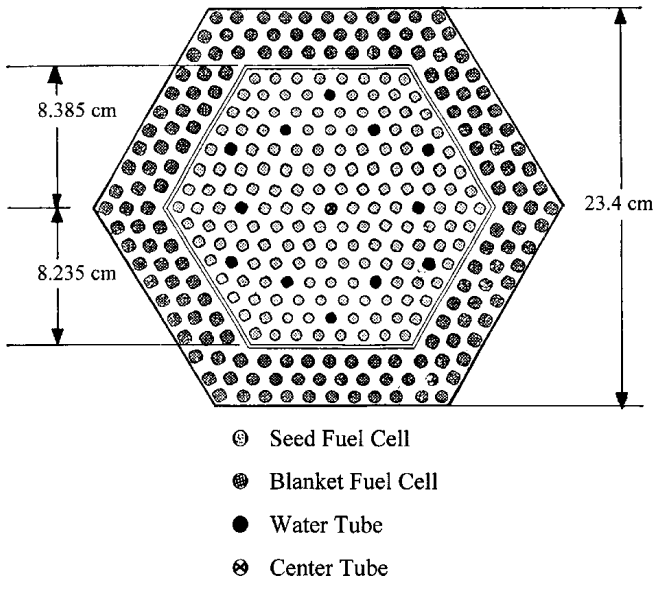

Figure 7. Radkowsky thorium reactor SBU fuel assembly geometry

Already in 1980 Alvin Radkowsky et al. published an article entitled „The Optimisation of Once-Through Uranium Cycle for Pressurised Light Water Reactors“. In this paper the authors point out that the optimum enrichment for nonproliferative ^{235}U is 20 %. A uranium core design under this condition leads to a reduction in core volume by about a factor two, while at the same time the safety will be enhanced as a result of utilisation of metallic fuel elements. The plutonium discharge would be reduced at least by a factor seven.

There is no need to employ soluble neutron absorbers for control purposes. Using the experience gained by the former studies the concept of the nonproliferative light water thorium reactor evolved.

The concept provides an economic approach to the utilisation of the nuclear potential of thorium in an „Once-Through Put Away Cycle“. Thorium is at least as plentiful as uranium despite of the fact that there have not been any exploration efforts so far. Thorium offers no attraction from either nonproliferative or economic standpoints, if it is used uniformly mixed with uranium of relative low enrichment. Studies revealed that the plutonium burn-up was less than in standard light water reactors, but a relative large amount of ^{233}U was created, which has weapon potential.

A. Radkowsky embarked on a completely different core layout utilising a special multiple-seed blanket arrangement. The seed regions are fuelled with nonproliferative enriched uranium in zirconium alloy. The blanket fuel elements are supposed to be thorium oxide spiked with a few percents of nonproliferative uranium oxide (<20 % enriched to ^{235}U). The seed regions have a very high water to fuel volume ratio. This leads to a good thermal spectrum and minimises the capture in ^{238}U with the result of a high value of the seed multiplication constant, which in turn maximises the fraction of core power obtained from the blanket. Another advantage is the minimisation of plutonium production.

Natural thorium contains no fissionable isotopes, but thorium is by neutron capture converted to the fissionable isotope ^{233}U. The transformation of thorium goes via ^{233}Pa. For a given neutron input the energy obtained from thorium is less than from uranium at short irradiation, but if thorium remains for a longer time in the core, this disadvantage is offset. In order to get an optimum power output from thorium, the geometry of the core arrangement is essential (seed-blanket system).

It is now planned to refuel the seeds at three-year intervals and the blankets at nine-year intervals. By use of successive seeds the blanket can be irradiated to the full metallurgical lifetime of about 100 000 MWd/T. This fact is supported by earlier Oak Ridge experiments.

The importance of the concept is that the energy from thorium is obtained by burning in place the ^{233}U as it is formed. It is not necessary to extract the ^{233}U and fabricate it into fuel elements. Thus thorium can be utilised for production of nuclear energy without the need for a new fuel cycle.

The inclusion of small amounts of uranium in the thorium oxide blanket rods leads to an economic gain and eliminates the need for soluble boron control during operation. Nevertheless soluble boron can be foreseen for emergency shut-down purposes.

The seed blanket core arrangement has a strong negative moderator coefficient, which will simplify adjustment of load variations. The residual ^{233}U could conceivably be utilised for weapons. However, as stated above, the blanket fuel elements also will contain some

nonproliferative uranium oxide (enriched up to 20 % ^{235}U). As a result the ^{233}U will be denatured by being uniformly mixed with non-fissionable ^{236}U and ^{238}U. An important difficulty in utilising ^{233}U is the very high Gamma-radioactivity accompanying it. This Gamma-activity arises in a complex chain reaction from the isotope ^{232}U. Over the ^{232}Th (n,2n)-reaction and a Beta-decay one arrives at ^{231}Pa. By neutron capture of ^{231}Pa ^{232}Pa is formed and this by Beta-decay goes to ^{232}U.

Because of the very high Gamma activity of ^{232}U it would be very difficult to separate out ^{233}U making the whole system even more nonproliferative. To make proper use of plutonium in connection with thorium in analogy to the 20 % U/Th-cycle an „Once-Through Put Away Cycle“ with Pu/Th was studied (Fig. 8).

A further important feature of the Pu/Th and the enriched U/Th-cycles is the virtual absence of transuranium elements, which are very difficult to be handled in a final storage of normal uranium spent fuel. The actinides have a high biological risk and hundred thousands years of half-life as seen in Fig. 9.

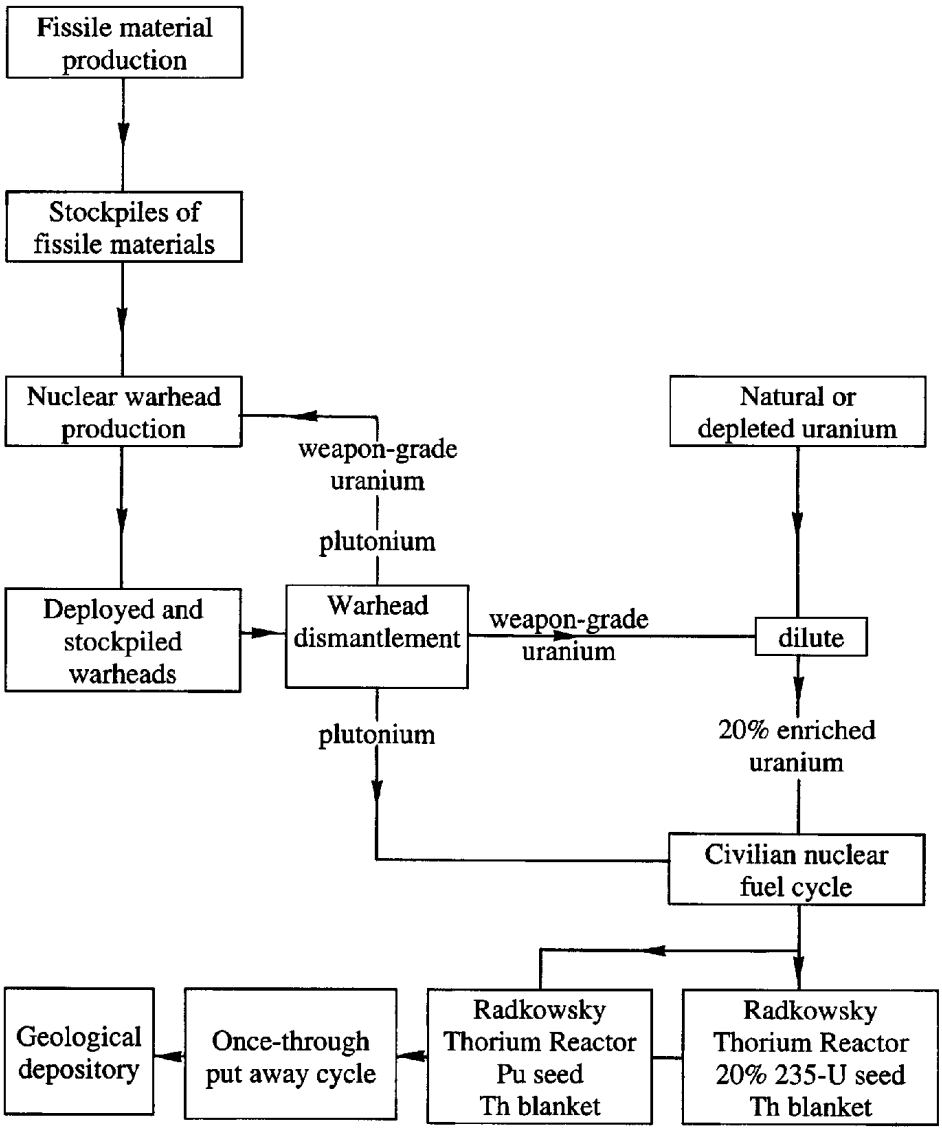

Figure 8. Warhead production and elimination activities.

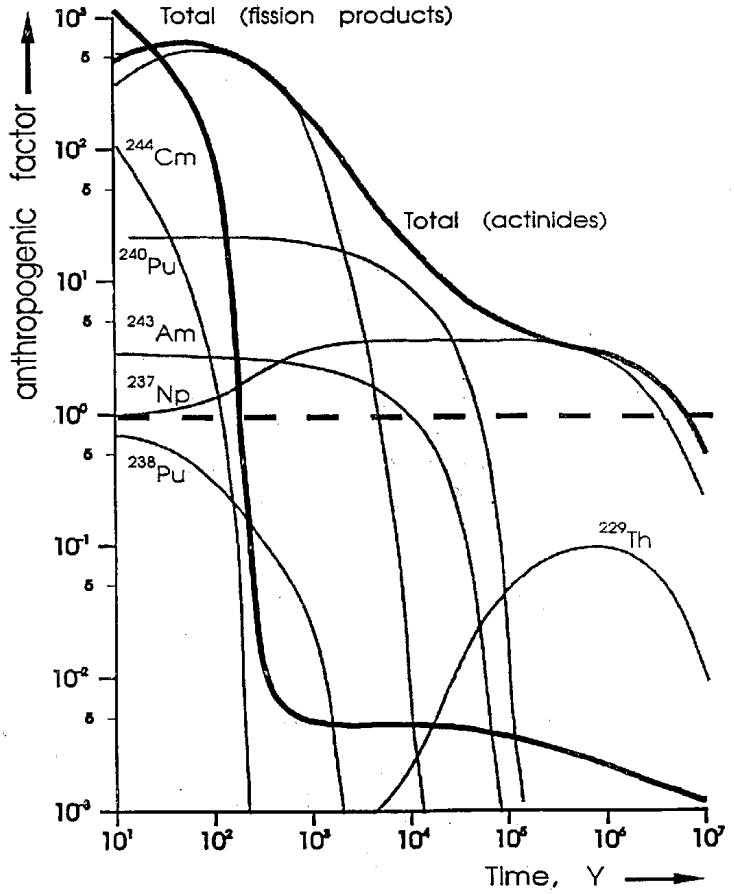

Figure 9. Time-dependence of radiotoxicity in a spent LWR fuel of 33 GW·d/t normalized to the radiotoxicity of the uranium ore (dashed line) mined to produce the fuel

In the absence of the actinides the final underground depository comes to the biological risk level of uranium ores contained in the earth crust after about 300 years. With actinides the risk level would be several orders of magnitude higher.

In the radioactive waste management issue thorium/^{233}U has a considerable advantage against uranium-plutonium systems resulting in much lower quantities of by-product actinides. In the production of ^{237}Np (neptunium) a factor of 100 less is achieved in a ^{233}U/thorium core compared with the usual ^{235}U/^{238}U core, if the calculations are carried out for a PWR with a burn-up of 30 000 MWd/t. For the isotopes of americium ^{241,242,243}Am about six orders of magnitude difference exist and for curium ^{243}Cm to ^{246}Cm roughly seven orders of magnitude or ten million less is the result. It must be kept in mind that the above calculations are performed for the thorium blanket region, while in the seed region the actinides production depends on the uranium enrichment. For 20 % ^{235}U enrichment compared with natural or low enriched uranium a factor of at least 5-10 reduction in actinides can be achieved. For safety reasons the reactivity control should be separately done for the blanket and the seed.

In recent publications costs for underground storage of spent fuel elements with actinides were quoted to be about ten thousand millions of US dollars. Adopting the Radkowsky Thorium Power Reactor concept would reduce these quoted expenditures considerably.

The main advantage of the thorium-based fuel cycles in thermal reactors is that is has a higher neutron yield of ^{233}U in comparison with the neutron yield of ^{239}Pu in the U/Pu-cycle. One possible approach to adjust the absorption rate in the fertile species is to alter the neutron spectrum by changing the degree of moderation. This can be achieved by adjusting the water volume to fuel ratio. The above idea is also incorporated in the RTPR concept.

4.3. Other work on thorium utilisation in PWRs

Proper utilisation of thorium was part of several national and international programs already in the past. Unfortunately none of the previous investigations, however, were of nonproliferative nature. In 1969 a government agreement of cooperation in the field of science and technology between Germany and Brazil was signed and a program between KFA Jülich and NUCLEBRAS on the thorium utilisation of PWRs was started in 1979. The program was planned to run in three phases with phase 1 from 1979 through 1983. In this period the technological basis for further work on $(Th,U)O_2$ fuel for PWRs was established and the feasibility of the chosen fuel cycle was proven in principle. In phase 2 nuclear core design and initiation of development of $(Th,Pu)O_2$ fuel as well as spent fuel treatment were the main themes. The program was terminated in 1988 after the advantageous features of a once through Pu/Th fuel cycle with high burn-up were confirmed.

Major activities in the area of thorium based nuclear fuels have been reported besides Brazil and Germany from Argentina, Australia, Canada, China, France, India, Italy, Japan, Pakistan, Romania, USA, USSR and by the International Atomic Energy Agency (IAEA), Vienna, Austria. A number of countries terminated their efforts in the eighties, but others have still programs running directed towards high temperature reactors, heavy water reactors, light water reactors and fast breeders. The theoretical and experimental studies comprise of nuclear core design and strategy calculations, thermal and mechanical fuel rod evaluations, technological development for $(Th,U)O_2$-PWR fuel such as palletising ex-gel technology, transfer of $(Th,U)O_2$-fuel technology to $(Th,Pu)O_2$-technology, irradiation testing and post-irradiation examination as well as fuel storage assessment and reprocessing studies such as the THOREX process.

Between 2-4 December 1985 the IAEA convened a Technical Committee Meeting to assess „Thorium-Based Nuclear Fuel: Current Status and Perspectives“. At this meeting main emphasis was given to the utilisation of thorium fuels in once-through nuclear fuel cycles. In an overview of world thorium resources it was stated that **r**easonable **a**ssured **r**esources (RAR) of thorium are estimated at about 1.16 million tons. About one third of this amount is available in the beach and in inland placers of India. Other countries which have sizeable reserves are Brazil, Canada, China, Norway, the former USSR, USA, Burma, Indonesia, Malaysia, Thailand, Turkey, and Sri Lanka. Thorium appears mainly in association of uranium and **r**are **e**arth **e**lements (REE). The present knowledge of the real thorium resources is poor, because there is practically no exploration effort due to insignificant demand.

As an outflow of the German-Brazilian cooperation a nuclear core design for the KWU-standard 1300 MW(e) PWR was performed. It was found that the KWU-type reactor can be operated without changes and restrictions in open and closed fuel cycle modes with all types of fissile material investigated. In the Th/Pu cycles without recycling great savings can be realised, when using Th/Pu instead of uranium fuel in existing reactors. In order to avoid the need of early reprocessing and to strive for reasonable savings, the „Once-Through Put Away Cycle“ with extended burn-ups was recommended. This means for the Radkowsky Thorium

Core after remaining for about 9 years in the reactor being put away to final storage without reprocessing.

For more than ten years the Commissariat á l'Energie Atomique and the Electricité de France have jointly carried out experimental design studies for the thorium cycle in unmodified PWRs. The studies first concerned the use of plutonium with thorium to start the cycle. The French investigators came also to the conclusion that burnable poisons are no longer necessary, when the assemblies are loaded in rings. To start the Th-^{233}U cycle two possibilities were considered:

(a) The high enriched Uranium/Thorium/^{233}U cycle and
(b) The Plutonium/Thorium/^{233}U cycle.

The French group proposed the solution to start the Th/^{233}U-cycle in unmodified PWRs in loading the whole reactor with Th/Pu-assemblies from the first core with three different plutonium assembly concentrations in ring form. The use of thorium as a fertile material is also especially suited for heavy water moderated reactors. High conversion ratios are reached and even breeding might be expected. It is, however, a fact that light water reactors, not the heavy water reactors, have been commercially established in the last two to three decades.

Nine papers were presented at the before mentioned IAEA 1985 Technical Meeting exploring the following subjects in some depth:

- evaluation of world thorium resources and incentives for further exploration;
- basic research results of physical, chemical and nuclear properties of thorium;
- reactor core and blanket concepts regarding utilisation of thorium-based fuel;
- advanced thorium fuel fabrication technology and reprocessing of thorium-based fuel.

The final panel discussion concluded: for a long-term fuel supply thorium could be recovered at costs less than US $ 80 per kg in the amount of about 2.4 million tons. For a long time it was believed to be impractical to breed with light water reactors; however, since the value of η for ^{233}U is only slightly lower in the epithermal region, while that of ^{235}U and ^{239}Pu is greatly reduced, the thorium cycle appears to be most attractive for thermal conversion.

The Radkowsky Thorium Power Corporation (RTCP), owner of the property rights of the Nonproliferative Radkowsky Light Water Thorium Reactor, joined forces with UE&C Nuclear, Inc. (Raytheon) to fully develop and build a Thorium Power Reactor as described before. Raytheon in particular is providing engineering, construction and support services based on their experience with a great number of nuclear plants operating in the United States.

Both companies are in contact with the Russian Research Center „Kurchatov Institute“ in Moscow. This Institute has a vast know-how in the nuclear arms development, dismantling of nuclear arms and the utilisation of thorium for power reactors. The Institute was prepared to verify the nuclear data base and reactor codes for the thorium power reactor concept in using benchmark models and experience of thorium irradiation. A further objective is a confirmation of the reactivity effects and control mechanism and validation of neutron-physical and thermohydraulic characteristics of the core design. Finally a 1000 MW(e) WWER-reactor will be made available to accept a full Radkowsky type thorium core. A feasibility study was submitted in Dec. 1994 by the Russian Research Center „Kurchatov Institute“ confirming the basic Radkowsky concept. The above relationship is also supported by efforts of US national

laboratories such as the Brookhaven National Laboratory. Since Sept. 1998 MIT's Nuclear Energy Department has joint forces of the Radkowsky Thorium Power Reactor concept.

Test fuel was produced in April 1999 at MSZ-Electrostal in Russia and thermal hydraulic testing started in May 1999 at the Russian Research Centre "Kurchatov Institute". In Russia and the former Eastern Bloc countries as well as in Finland 45 light water pressurised power reactors are in operation; 14 are under construction. The total power output of these reactors will be more than 42 000 MW(e). Seven WWER-1000 units are operative in Russia, ten in the Ukraine and two in Bulgaria (Fig. 10).

	Operational	Under construction	Electric output in MW(e)
WWER 230			
Bulgaria	4	-	1.760
Slovakia	2	-	880
Russia	4	-	1.760
WWER 213			
Slovakia	2	4	2.640
Czech Republic	4	-	1.760
Finland	2	-	880
Hungary	4	-	1.760
Russia	2	-	880
Ukraine	2	-	880
WWER 1000			
Bulgaria	2	-	2.000
Czech Republic	-	2	2.000
Russia	7	2	9.000
Ukraine	10	6	16.000
Sum	**45**	**14**	**42.200**

Figure 10. WWER reactors operational and under construction.

As some of the WWER reactor types do not meet Western safety standards, a number of multimillion dollar upgrading programs are under way. A full Pu/Th or 20 % ^{235}U/Th core of the Radkowsky design could contribute to safety improvements of Russian built Pressurised Water Power Reactors (PWPRs), but in the Western countries there are about 160 PWPRs with sufficient licensable life to justify retrofit deployment of the Radkowsky thorium reactor core concept.

5. CONSEQUENCES OF A GLOBAL NUCLEAR EXCHANGE

The two atomic weapons used in warfare were dropped on Hiroshima and Nagasaki, Japan, in August 1945. The yield of the Hiroshima bomb was about 15 000 tons TNT equivalent, while the Nagasaki bomb had a yield of slightly over 20 000 tons of TNT. It is reported that the casualties were 120 000 people immediately and some 250 000 fatalities up to 1990. In Nagasaki approximately seven square kilometres were destroyed, while in Hiroshima about thirteen square kilometres of urbanised area were devastated. The two bombs were exploded in the air some 500 meters above ground. The blast damage and the damage by heat irradiation were for these heights at a maximum, but the radioactive fallout was at a minimum. The heat intensity had the most direct consequences so that people being away several kilometres from the hypocenter suffered serious skin burns. The enormous pressure

waves damaged buildings and other construction works three to four kilometres away. The relative low yield nuclear explosions showed that the destructive power of nuclear weapons is immense. Nowadays weapon yields can reach million tons of TNT and thus can destroy also large cities within a few seconds. Typical strategic warheads are in the range of about 200 000 tons of TNT. The atmosphere is effected by the explosion of nuclear devices over large areas and radioactive fission products and neutron-induced radioactivity contaminate the environment and can extinct any life being plants, animals or human beings. When a nuclear device is detonated, the fissionable materials uranium and/or plutonium become volatile within 10^{-6} seconds. The effective radiation temperature reaches about 40 million Kelvin. A blast of X-rays is the result of the intense power. Fires initiated by the nuclear explosion are mostly of secondary nature, because the blastwave extinguishes fire ignited by the primary heat sphere. Scenarios for a global nuclear war have been discussed in a number of publications such as the US National Academy of Sciences, the Office of Technology Assessment and the National Research Council. The National Research Council published its nuclear war scenario in 1985 assuming that 6 500 megatons of TNT equivalent were detonated by 25 000 devices between 50 kilotons and 1.5 megatons plus tactical 1 500 megatons in surface bursts and the same amount in urban zones and 500 megatons of smaller tactical devices. The term „overkilling capacity" evolved from these scenarios and to the layman it is frustrating to hear that the overkilling capacity is reaching one hundred, which means that all life on planet earth could be extinguished one-hundred-fold, or putting it in other terms, if only one percent of the nuclear weapons available reach their targets, life would virtually be coming to an end.

It seems needless to discuss the consequences of a global nuclear warfare, but also a local nuclear exchange would have pronounced ecological and climatic effects, in particular on agricultural productivity and the availability of food after a local nuclear exchange. To arrive at precise estimates of a limited nuclear war on humans and the duration with severe effects for humans, is difficult or even impossible. But from the energy involved it is no over-estimation to say that billions of human beings may die immediately or within a short period of time. It is known that current strategic deterrence policies imply that in an escalating nuclear conflict the majority of warheads may be targeted directly against urban and industrial centres.

In summary the effects of a nuclear war can be characterised:

1. Direct consequences:
a. Devastation by shock-wave; 500 km^2/MT (Megaton)
b. Mushroom fireball has a vertical velocity of 100 m/s; explosions over 100 000 tons reach up to the stratosphere and fire damages extend to 250 km^2/MT (Megaton)
c. Radioactive fallout with lethal dosis value of 4.5 Sv:
   in the first 48 hours 1000 km^2/MT (Megaton)
   within 50 years 2000 km^2/MT (Megaton)

2. Indirect consequences:
a. Meteorological and climatic effects
b. Ecological and biological effects
c. Adverse effects on food production

Because more than fifty years have passed since the first and only use of nuclear weapons for warfare a great number of political leaders claim that the nuclear balance between the two super-powers made this possible. The large number of nuclear warheads in stock on both sides convinced their leaderships that nuclear wars cannot be won by neither party.

Independent of the encouraging NPT decision to keep indefinitely in force the validity of the Treaty, it was more or less a general consensus that the present nuclear weapons states are allowed to maintain a minimum deterrence force. It must be remembered that at the time, when the NPT was signed in 1968 less than ten-thousand warheads existed. At its maximum a few years ago the number of warheads was approximately 60 000. The NPT review conference was not an end for itself. It is playing a key-role to lead to a cessation of the nuclear arms race. It could well lead to a complete disarmament under effective international control. Since 1950 for the elimination of nuclear weapons rhetorical lip services were paid by practical all delegates from all countries all over the world at different occasions. It would be a good time now to start this process as the Cold War ended. What is happening to nuclear weapons could be extended to other mass destruction means including chemical and biological warfare. One can only hope that a large majority of countries identifies itself and supports such ideas.

6. NUCLEAR WEAPONS VERIFYING AND SAFEGUARDS SYSTEMS

The accumulated amount of weapongrade and reactorgrade plutonium was discussed before. By now the total plutonium inventory amounts to about 1100 tons. Highly enriched ^{235}U in nuclear warheads both in the United States and Russia exceed 1000 tons and in thermonuclear devices about 200 kg tritium is available. Not only the number of nuclear warheads but also the nuclear materials in the warheads must be safeguarded and verified from time to time. Sophisticated technical regimes have been developed for control and verification. Particular attention was given to submarine-launched cruise missiles (SLCMs), submarine-launched ballistic missiles (SLBMs), intercontinental ballistic missiles (ICBMs) and aircraft bombs and missiles.

Already in 1962 the Government of the United States of America, in order to lend its support to building-up a safeguards system by the International Atomic Energy Agency, signed an Agreement that the Agency's safeguards could be applied for test purposes to four US reactor facilities. As a conclusion the inspectors recommended that qualified auditors, statisticians and highly qualified technical personnel could assure themselves with reasonable accuracy on the situation of a given facility. It was, however, strongly pointed out that the most convincing and accurate information on a possible non-peaceful diversion of nuclear materials could only be obtained by destructive or non-destructive analysis of the fuel in question. Gamma-spectroscopy of the fission products was pointed out as one possibility, since it is possible to select an appropriate number of characteristic fission product Gamma-lines covering different half-lives. This way information on fissionable material burned could be obtained and also information of the history and the total neutron flux seen.

Modern technology has made the cruise missiles a strategic nuclear weapon able to target very precisely any location in the world over long distances. Also nuclear detector equipment was improved dramatically. A short summary for the verification of nuclear arms is given in Fig. 11.

Portable monitors for detecting fissile materials and chemical explosives are available as well as very improved Gamma-ray and neutron detectors, which can also be operated from helicopters. Satellite observation and guiding techniques with resolutions in the cm-range exist besides video-surveillance and monitoring electronic seals. Further control possibilities are environmental sampling to detect releases of radionuclides and detection of so-called „signatures“ typical for a particular nuclear fuel cycle. Institutional control mechanisms are on hand, which involve various political, economic and diplomatic strategies to control sensitive materials and facilities up to a complete technology.

1. Warheads: All 55.000 warheads contain U-235 and/or Pu-239
either as fission bomb or trigger for hydrogen bomb

HEU (Highly Enriched Uranium) 93.5 % U-235
(U-234 1 % and U-238 5.5 %)

WGPu (WeaponGrade Plutonium) 93 % Pu-239
(Pu-240 6 % and others 1 %)

Fission bomb consists of inner sphere of HEU and/or WGPu
followed by 2 cm Be reflector, 3 cm temper (tungsten or uranium),
10 cm high explosive and 1 cm of Al-case).

2. Detection possibilities:

Neutrons: HEU 1.6 N/s/kg
WGPu 56 000 N/s/kg
Neutron production mainly by spontaneous fission occurring in isotopes
with even numbers (Pu-238, Pu-240, ...)
HEU weapon with tungsten 60 N/s
HEU weapon with depleted U 2500 N/s
WGPu weapon with tungsten 1.500.000 N/s
WGPu weapon with depleted U 1.500.000 N/s

Figure 11. Verification of nuclear arms.

7. OUTLOOK

If 1100 tons of plutonium must be stored, 275 000 shipping containers are required (a maximum of 4 kg plutonium in each container). Cost requirements for storing 1 gram of plutonium per year have a band-width of US $ 1 to 2. Therefore between 1.1 and 2.2 billion US $ must be provided per year. When using MOX-fuel in light water reactors twelve reactor units of 1000 MW(e) each would be able to burn in a ten-year period 100 tons of weapongrade or reactorgrade plutonium, but MOX-fuel will produce for each unit plutonium burned two-third units of new reactorgrade plutonium. The nonproliferative Radkowsky thorium reactor system will be able to burn about 850 kg plutonium in a 1000 MW(e) PWR per year. As a plutonium burning system there would be virtually no plutonium produced.

In the last decade opinion polls in favour and against disarmament inspections were taken in the United States, UK, France, India, Germany and Japan. A majority of the population - between 70 and 92 % - supported disarmament inspections. Being asked, if mankind should drive for a nuclear weapons-free world, a huge majority find the idea desirable but believe that it is not yet feasible. An almost unanimous support was given to the proposal to undertake every effort for reaching finally a nuclear weapon-free world with the argument that otherwise self-extinction may be the consequence for mankind.

Since the Cold-War ended the risk of nuclear wars between the super-powers has become remote and this is one of the reasons, why stocks of nuclear weapons both in the United States and Russia are being drastically reduced by some 90 %. The Strategic Arms Limitation Treaty

(SALT) initiated this huge weapon reduction, but in each of the two countries there are still about 6 000 warheads available as deterrence force. It can be argued that a survivable force of about 2 000 warheads would be more than enough to be considered a „finite deterrence force". In reality several hundreds of warheads would be enough. Under these circumstances pressures have been exerted onto the United States and Russia at the nonproliferation extension discussions to cut their military budgets and make the free resources available to deal with urgent national problems. In the United States alone it is estimated that over a ten-year period as much as 150 to 200 billion dollars could be shifted from military requirements to civilian purposes.

The weapon dismantling process would not be worth the effort, if it were not connected with a policy, what to do with the fissile bomb materials. The Radkowsky Thorium Reactor gives one possible answer. It gives also a positive answer to the continued need of the nuclear energy sources, because, in order to get rid of the plutonium and the highly enriched ^{235}U bomb materials, more than 100 existing pressurised water reactors in the world will be required for the next decades to burn the fissionable materials and eliminate these products for ever. Radkowsky Thorium Reactor Cores would not produce new plutonium and latest studies indicate that truly nonproliferative nuclear power reactors could evoke broad public acceptance of such nuclear power systems. Only one alternative way exists theoretically, namely, to explode the bombs with all the consequences elaborated earlier.

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DEVELOPMENT REQUIREMENTS IN REPROCESSING THORIUM FUELS*

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Abstract. Thorium fuels have been much discussed over the past few years as a replacement for uranium in main-line power reactors, or in special systems to consume minor actinides or unwanted stocks of plutonium. Less attention has been paid to the reprocessing. Existing knowledge is a useful basis for a tentative scheme to reprocess thorium fuels, but the reasons for favouring thorium, the types of fuel in which it is most likely to be used, and external circumstances with their consequent requirements have all changed since the early work was performed. The amount still to be done should not be under-estimated.

INTRODUCTION

Much of the discussion on thorium fuels has centred on the form and composition of such fuels, their behaviour in various neutron spectra and the burn-up or degree of consumption attainable. Less attention has been paid to the reprocessing that would be needed if a single pass failed to achieve the desired ends, and there is evidently a widespread assumption that at least with respect to solvent extraction processes, all problems have already been solved.

Reprocessing trials so far reported have covered mixtures of thorium with a little uranium, such as might be bred from it, although even for this limited purpose they leave much further development to be done before industrial implementation. Plutonium-enriched fuels now need particular attention. The residue of plutonium remaining from the original fissile component, or the smaller but still significant amount arising by transmutation of uranium-238 if initially added to denature the ^{233}U product for anti-proliferative reasons, would add considerably to the difficulties and require a modification of the basic scheme.

Eventually the required products would have to be defined with thorium, uranium and plutonium either separate or in some combination. The present paper considers both possibilities, concentrating mainly on the more demanding, i.e. to provide for three distinct products, albeit with perhaps lower mutual decontamination than is specified for current reprocessing. In addition, for the sake of logical consistency with the expected purposes of using thorium, the highly-active waste stream would need partition although with one exception – the presence of protoactinium-231 – this is unlikely to raise significant new issues.

The three-way product separation might in theory be achieved simply by backwashing a common extract with progressively more dilute acid. This is the principle adopted in the two-way separation of the classic Thorex process (Figure 1a), where the order-of-magnitude difference between the extractabilities of uranium and thorium would probably be adequate (Figure 2, taken from [1]). However, the intermediate extractability of plutonium is unlikely to allow sufficiently reliable separation from either of the other main components, and a more elaborate scheme must therefore be considered.

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* 1999 meeting.

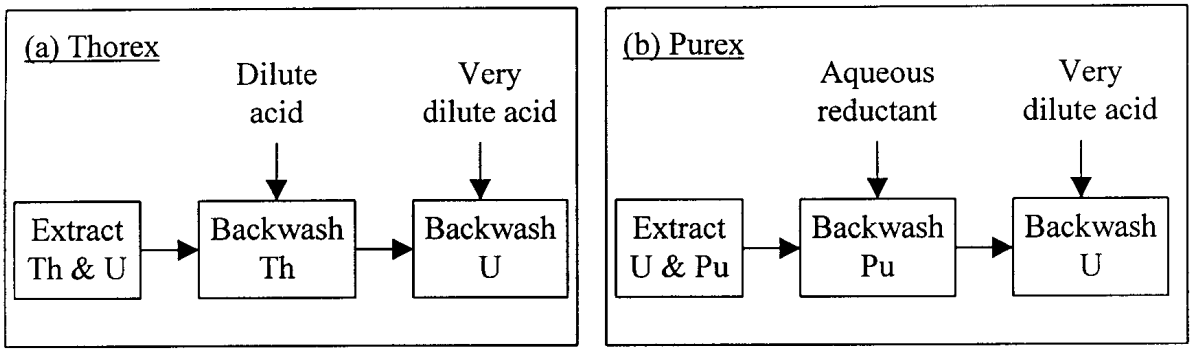

Figure 1. Essentials of Thorex and Purex separation schemes.

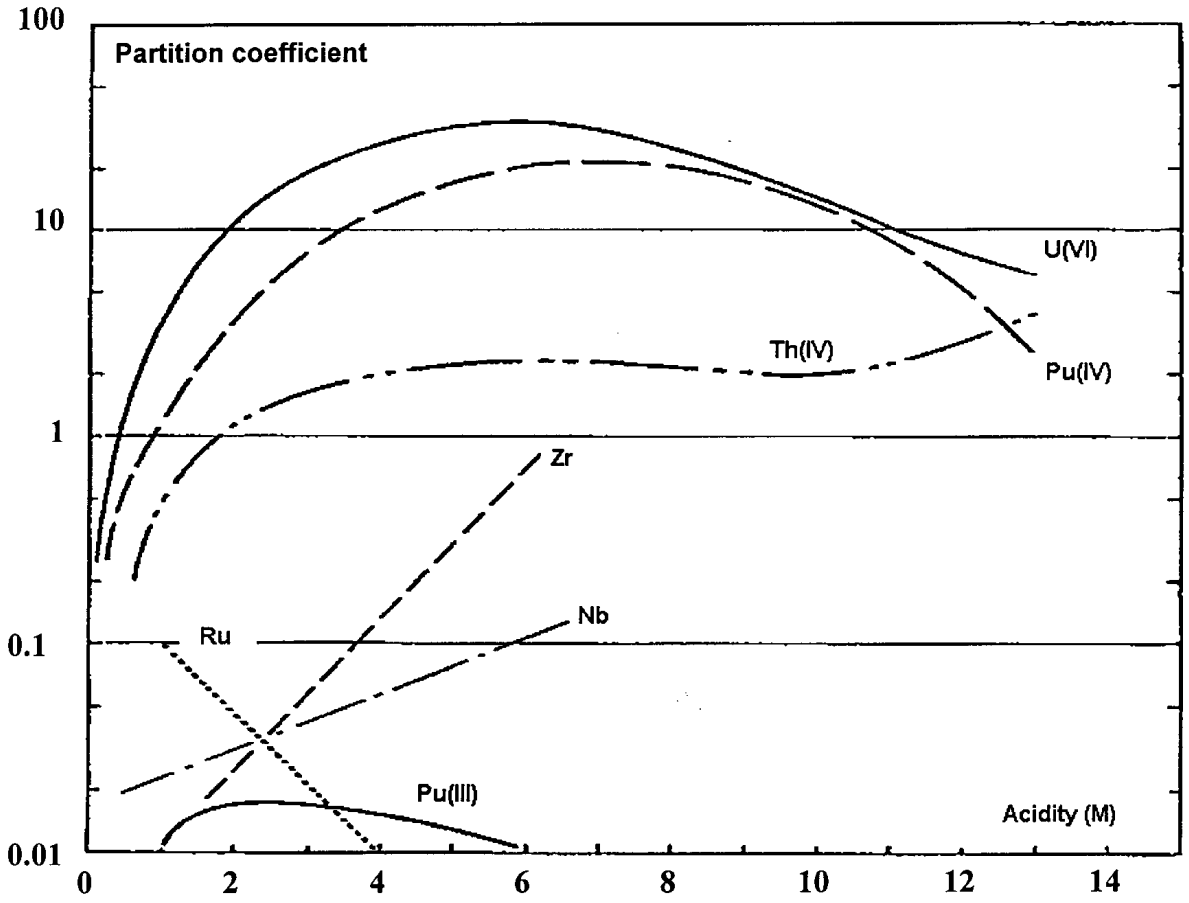

Figure 2. Extraction by 20% TBP/Kerosene from nitric acid [1]

The suggestion that a simple combination of the Thorex and well-established Purex processes would meet the need appears unduly optimistic, as discussed below, although the principles

behind the processes may still be applied. Schemes more likely to prove satisfactory are therefore suggested and the lines of necessary development work indicated.

Any process chosen in the first instance is likely to be as close as possible to familiar practice. Whatever form the rest of the scheme might take, the first step would be dissolution in nitric

acid with a fluoride catalyst and with aluminium nitrate to restrict corrosion and prevent precipitation of thorium tetrafluoride. [2] Thereafter, extraction with tributyl phosphate (TBP) is generally assumed with some sequence of differential backwashing or re-extraction to separate the common extract into thorium, uranium and plutonium fractions. Alternative extractants are possible, with similar considerations applying. Any subsequent purification of the products is a refinement that for the time being may be disregarded.

As solvent, TBP would be diluted probably to 30% with a hydrocarbon such as dodecane or kerosene. Solvent loading would have to be rather low to ensure effective extraction of thorium and prevent third-phase formation, and a significant amount of zirconium would also be extracted [3]. Most of the technetium would almost certainly follow it, with serious potential implications for a later stage [4].

THOREX/PUREX COMBINATION

The idea of simply combining Thorex to separate uranium from thorium with Purex to separate plutonium from uranium apparently assumes that

(a) plutonium would remain associated with uranium in Thorex or thorium with uranium in Purex, according to the order in which the processes were taken; and
(b) the change in the proportion of uranium from predominance to being one of two minor components would not adversely affect Purex.

Neither assumption is necessarily true, and although a cursory examination of the solvent-extraction relationships between the elements might suggest the first to be credible, more detailed consideration indicates difficulties.

If the principles of Thorex (Figure 1a) were to be applied first, then the separation would aim initially to backwash thorium while leaving plutonium in the solvent with the uranium. This might appear to offer the great advantage of segregating the main bulk from the fissile stream, which if itself then backwashed into a small volume could be further processed in equipment of dimensions less favourable to criticality. However, the high extractability of uranium coupled with the large volume of solvent needed to extract thorium raises some doubts about this prospect.

In the first backwash, plutonium would be intermediate in extractability between thorium and uranium with comparable differences from both, and so would tend to distribute itself between the two streams according to the precise conditions. Provided that it mostly followed the uranium this might be acceptable, particularly since the expected relatively poor decontamination from fission products would require stricter containment of thorium during refabrication than its own radiotoxicity suggests. Considerations of nuclear safety might however require the main thorium stream to be kept essentially free from plutonium, so conditions should preferably be arranged to retain all the uranium and plutonium in the solvent, together with whatever proportion of the thorium this might imply. This is in fact the basis of a reduced scheme considered later.

A possible schematic flowsheet for the full separation is shown as Figure 3. It assumes that sufficient thorium would be separated in the first backwash for the rest to remain with the uranium at the following stage; if however too much were backwashed with the plutonium to be tolerable in the product, a plutonium purification cycle as in the alternative scheme of Figure 4 would be needed.

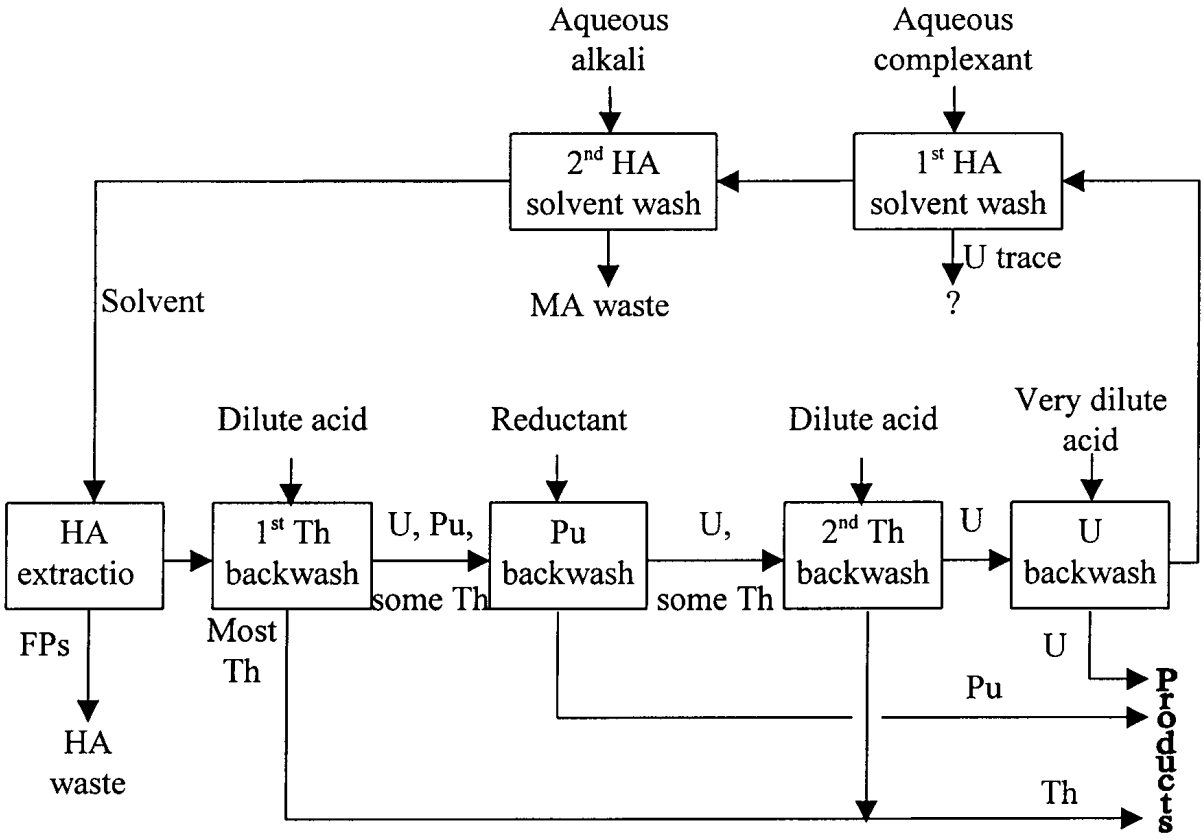

Figure 3. Suggested scheme for reprocessing thorium-plutonium fuels.

A point to notice is the first solvent wash. Because of its high affinity for TBP, some uranium is likely to remain in the solvent after the final product backwashing. Given the solvent flow required to accommodate thorium at a relatively low loading, the amount could be significant even at a low concentration. Since its fissile content could be almost 100%, and would certainly be much higher than the 1% or thereabouts in conventional Purex, routing such uranium to the general medium-active waste stream would be undesirable, perhaps to the extent of preventing acceptance by regulatory authorities whether on grounds of safety or of proliferation resistance. Uranium is therefore backwashed with an aqueous complexing agent, for instance sodium carbonate. This is normal practice to prevent precipitation by caustic alkali in a later washing step; the difference here is that the complexing wash raffinate would be segregated. The amount of uranium might be small enough to be routed to high-level waste, or otherwise it would be combined with the major uranium stream. In either case elimination of salt content and acidic phosphate esters would be desirable if not mandatory.

Since the greatest difference in extractability is between Pu(III) and the rest, a reductive step might instead be the best start as shown in figure 4. This again would raise the question of whether the plutonium as first backwashed would be sufficiently free from thorium to be acceptable as a product without a purification cycle. Such a requirement, or lack of it, could be a major consideration in deciding the choice between the two schemes, but would become apparent only after both had been tested.

Separation in Purex (Figure 1b) depends essentially on the reduction of Pu(IV) to the almost inextractable Pu(III), since the difference in extractability between U(VI) and Pu(IV) is insufficient. The tendency for plutonium to be re-oxidised in the solvent phase, particularly

under the autocatalytic influence of nitrous acid carried forward from the highly-active extraction, leads to extensive cycling between solvent and aqueous phases and the need for six or more times the theoretical quantity of reductant, besides a lower solvent/aqueous ratio than might otherwise be necessary to ensure complete backwashing. To maintain favourable kinetics and minimise cycling, the acidity at the reduction stage has to be kept as low as practicable. If thorium were to replace uranium as major component, its lower extractability by an order of magnitude would almost certainly cause some considerable proportion to accompany plutonium rather than uranium.

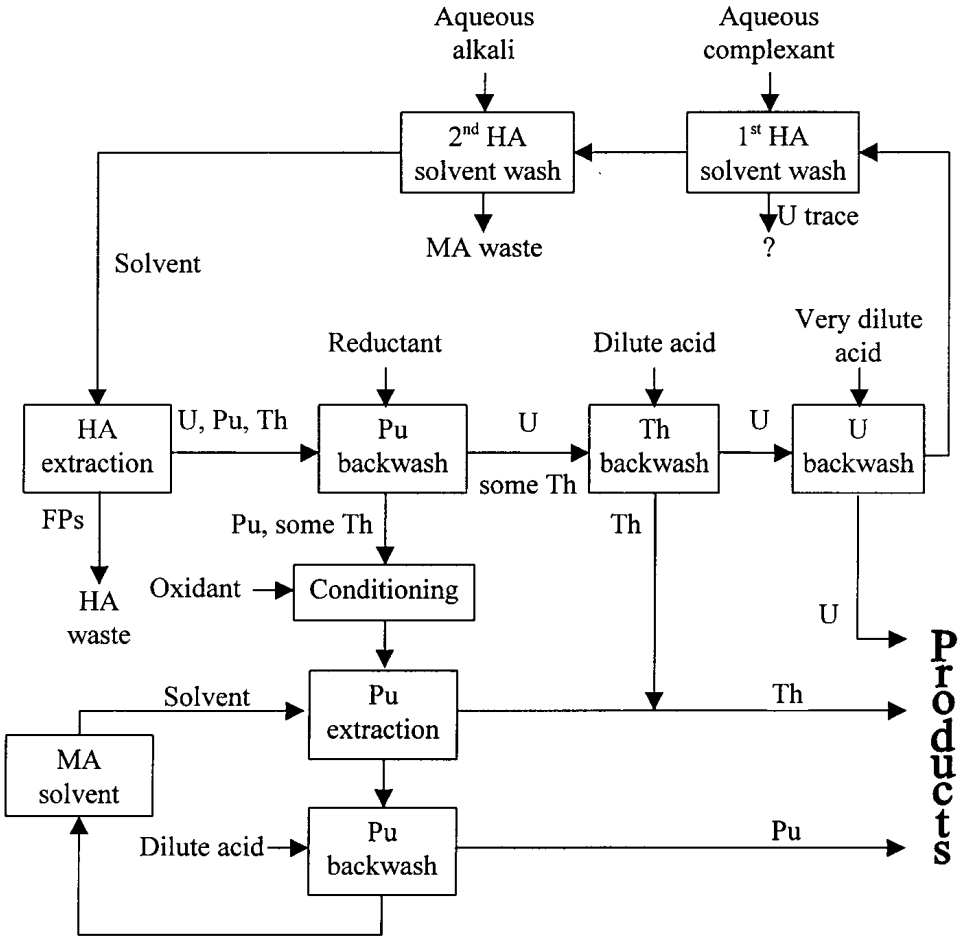

Figure 4. Alternative scheme for reprocessing thorium-plutonium fuels.

The reductant currently favoured is U(IV) nitrate, protected by hydrazine against oxidation by nitrous acid. Indications are that in thorium fuel initially enriched with civil plutonium, the amount of uranium at discharge would be similar to that of residual plutonium or rather less. Although with military rather than civil plutonium the residue would be about half as great and the U/Pu ratio correspondingly higher, [5] in neither case would the amount of uranium be enough to serve as reductant unless repeatedly recycled, which would add to problems of criticality control or at least complicate them. A supplement from natural or depleted sources might be acceptable, and ^{238}U deliberately added to denature the ^{238}U product would presumably meet the need, although in the next cycle of irradiation it would conflict to some extent with the expected purpose of using thorium. An alternative reductant may be desirable in any case for the sake of directing neptunium as Np(V) to the plutonium stream, and avoiding difficulties in controlling the technetium-catalysed destruction of the protective hydrazine [4].

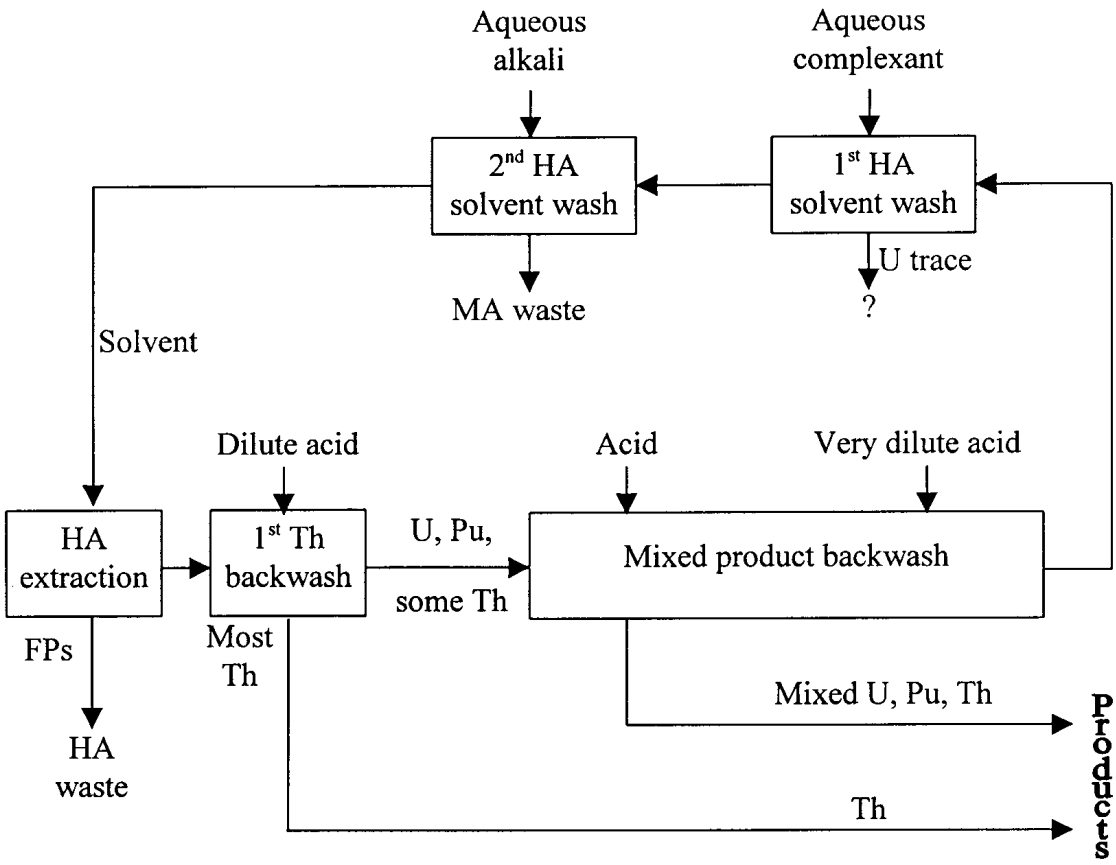

Figure 5. Simplified scheme for reprocessing thorium-plutonium fuels.

Hydroxylamine might be used to control neptunium valency, [6] but kinetically is still more sensitive to acid than U(IV). In the presence of technetium, it promotes a rapid oxidation of U(IV) by nitric and nitrous acids [7]. Although a similar mutually destructive reaction with the more weakly reducing Pu(III) is less probable, the mechanisms of catalysis by technetium are complex and by no means fully understood, so the possibility should not be dismissed without examination. Various organic compounds such as aldehydes and oximes have been considered as alternative reductants and might be preferable, particularly if effective in both solvent and aqueous phases. The options and related development requirements warrant careful attention.

A considerable simplification would be possible if the mutual separation of uranium and plutonium were deemed unnecessary, especially if a moderate admixture of thorium with these elements were acceptable. The flowsheet outline might then be as in Figure 5, with the requirement for reducing plutonium avoided altogether. The mixed-product backwash is in fact very similar to that in the first (co-decontamination) cycle of the Sellafield Magnox reprocessing plant, with the important difference that while there the fissile content is a fraction of 1%; here it would be much higher and require precautions against criticality, for instance to ensure that the plant could not operate without the supplementary acid feed to prevent precipitation of plutonium. With no obvious reason to separate the two fissile elements from each other when both are presumably to be incorporated into new fuel, this reduced scheme is probably the most sensible to adopt in the first instance.

In principle a still greater simplification might be achieved by dispensing altogether with separation of thorium from uranium and plutonium. At the backwash stage, however, the high level of thorium tetranitrate would tend to salt out the uranium, increasing a degree of cycling within the contactor bank that is already liable to be substantial, with consequently greater losses to the solvent wash.

A further short-term possibility derives from the currently low value placed on thorium itself, maybe less than the cost of recovery. It might for the most part be left with the fission products after a partial extraction, eliminating the need for the first backwash of Figure 5. Although wasteful and in the long run unsustainable, this might be temporarily acceptable to permit a first demonstration of other stages in the system. Since it would raise no additional development issues apart from the apparently undemanding conditioning of thorium into a waste form, it is not considered further. However, a variant with thorium subsequently extracted by a second, larger batch of solvent [2] might be worth consideration if there were a special need to confine uranium and plutonium to a small solvent flow. Exactly how much thorium would then accompany them would depend on the particular conditions.

As mentioned previously, if the purpose of introducing the thorium cycle is to help dispose of long-lived radionuclides, then protoactinium-231 should be taken into account. Being poorly extracted by TBP from nitric acid, [2] it will accompany the bulk of the fission products to highly-active waste. How it might be separated is uncertain; past developments have been said to be adequate for construction of a plant on demand, [1] although statements of this kind tend to gloss over a great many practical issues that would still need attention, particularly after a lapse of decades during which standards have changed substantially. Moreover, methods in the literature do not appear very attractive industrially, and there would be a case for leaving the nuclide in the waste with various others that could not profitably be incinerated. An important part of that case might be the consideration that a facility to separate ^{231}Pa could also be used in principle to separate ^{233}Pa from short-cooled fuel (as indeed was the original purpose) and so constitute a proliferation-prone route to isotopically pure ^{233}U.

NON-AQUEOUS PROCESSES

Dry processes have been suggested as a way of avoiding the difficulties inherent in the aqueous routes, with several considered or already applied to uranium fuels albeit generally on a small scale. Of these, fluoride volatility can be virtually ruled out since thorium fluoride is involatile. It might conceivably be used to separate uranium and less easily plutonium from the general mass, but would first require the thorium matrix to be dispersed in some way, for instance by dissolution in a molten salt mixture. Once this had been done, there would be an obvious incentive to continue by a route such as electrolysis specifically developed for molten salts. So far these studies have concentrated on chloride media, but the possibility of synergy with the development of molten-salt reactors would put a premium on using fluorides instead.

Thorium is said to be thermodynamically suited to processes of this kind, but its behaviour in relation to uranium and plutonium needs to be determined in practice, especially for the purpose of controlling associations and proportions in mixed products if desired or unavoidable. Also needing attention are methods of separating fission products from the salt, or separating most of the salt from the fission products and forming the residue into a suitably stabilised waste for disposal, although this issue is clearly not specific to thorium fuel. At the present stage, more detailed consideration would be premature.

Although thorium fluoride is not volatile, the chloride is, and this was at one time considered a possible basis for reprocessing HTR-type fuels, comprising particles of fuel substance with a hard, impermeable coating compacted with graphite into spheres, annuli or prismatic blocks. While considered unlikely to meet cost and safety considerations for this purpose,[1] it might conceivably be worth re-examining where the impediment of a graphite matrix is absent.

DEVELOPMENT REQUIREMENTS

A first requirement is to gather together all the accessible information on the subject, not only such as has been formally reported but also anecdotal experience that in practical terms may be equally important. A valuable start has been made in the form of a European Commission review of the whole cycle in general [8] and a report on its status as a waste-management option, [9] but for the present purpose more detail is needed. Moreover, where experience is derived from plant operation, consideration must also be given to relevant circumstances, for instance whether they demanded compromises or expedients acceptable at the time (commonly several decades ago) but not under present industrial and regulatory conditions.

The following discussion disregards HTR-type fuels, which appear not at all amenable to processing at acceptable cost [1]. It assumes for the time being minimal extensions beyond current practice so as to require the least possible development work; more radical changes could be studied later if desired.. Consideration is therefore restricted to oxide fuel in metal cladding, processed by an aqueous route. This immediately raises an obstacle common to all the options, in getting the fuel into solution at the start.

Dissolution

Thorium dioxide is essentially inert to nitric acid, and the classic Thorex reagent therefore contains small proportions of hydrofluoric acid to catalyse dissolution and of aluminium nitrate to reduce corrosion of the dissolver vessel itself. Aluminium may adequately protect immersed surfaces, but corrosion can also be significant in the vapour space, where no such protection could be given, and an adequate lifetime for the ventilation system must be assured. The optimum composition of the dissolvent mixture may already have been determined, together with the rate of fuel dissolution under realistic conditions, but tests will be needed to establish (a) corrosion rates on existing or proposed alloys of construction, (b) whether any measures will be needed to remove hydrofluoric acid from the aerial effluent, and (c) the durability of ventilation ducting, filter housings, off-gas equipment and anything else liable to be exposed to the vapour or entrained spray.

Solvent extraction

Solvent extraction is a reversible process and seldom complete in a single contact. Each block in the various schemes illustrated above may represent a series of up to about twenty contact stages with the two phases flowing countercurrently between them. Sometimes the stages are physically distinct and operate as effectively complete equilibrations with intervening separation. More commonly in modern plant, the stages are a theoretical and approximate representation of a system in which solvent and aqueous phases flow past each other more or less continuously, always in contact but nowhere in mutual equilibrium. Future plant may revert to discrete stages but with centrifugal enhancement of settling and possibly too short a contact time for equilibrium to be completely established. Developments in Purex are likely to cover all engineering requirements, with none obviously specific to thorium.

The design and safety analysis of a new plant requires, as for reactors, a reliable prediction of how it would behave under conditions that must not be allowed to arise, in equipment that does not yet exist. It is therefore modelled either physically, which is time-consuming and expensive if not totally impracticable, or more often mathematically, although some form of confirmatory demonstration will always be needed. Where the plant can be treated as comprising discrete stages at equilibrium, dominated by a single extractable species such as uranyl nitrate, the modelling may be graphical. Nowadays however this is inadequate for anything more than a preliminary indication, and where several interacting species are involved the system is simulated by computer. Apart from any chemical reactions that add a further dimension of complexity, an additional extractable species generally affects an existing equilibrium such as

$$UO_2^{++} + 2NO_3^- + 2TBP \leftrightarrow UO_2(NO_3)_2 \cdot (TBP)_2$$

by two opposing mechanisms: adding to the overall nitrate level and so favouring extraction of the TBP-nitrato complex, but competing for free TBP and to that extent diminishing it.

If the concept of equilibrium stages is wholly inappropriate, then kinetic effects have to be taken explicitly into account. They include mass transfer and hydraulic factors such as the degrees of dispersion and agitation, all interacting and together constituting a very complex system, difficult to characterise and liable to need at least a measure of semi-empirical approximation.

In thermodynamically ideal systems the various relationships at equilibrium could be expressed rather simply and deduced from relatively few separate measurements on individual species. The systems are however far from ideal, and measurements are needed over the whole range of credible compositions with a fine enough mesh for reliable interpolation; skimping them, especially at the extremes, is liable to prove a false economy.

Extensive data have long been available on the distribution behaviour of uranium, plutonium and nitric acid, [10] also for thorium itself, [1] and perhaps but by no means certainly on that of uranium and nitric acid in its presence. However, they are probably incomplete, badly scattered (e.g. Figure 6), and will almost certainly need confirmation and extension to cover the presence of plutonium in thorium systems. In existing data there is often some uncertainty about the concentration of free nitric acid, one of the strongest single influences on the distribution of other species, but difficult to measure in aqueous solutions of hydrolysable ions, much more so in the solvent, and in some regions subject to disturbance by reversible and temperature-sensitive reactions such as

$$3Pu^{4+} + 2H_2O \leftrightarrow PuO_2^{2+} + 2Pu^{3+} + 4H^+$$

which liberate or absorb acid. The more fundamental quantity, although seldom if ever measured directly, is the concentration of nitrate ion. Without a reliable figure at least for the aqueous phase, the utmost accuracy in measuring other quantities would be valueless. For this reason if for no other, extreme caution is needed in using data gathered for different purposes where precise determination of acidity may have been considered relatively unimportant.

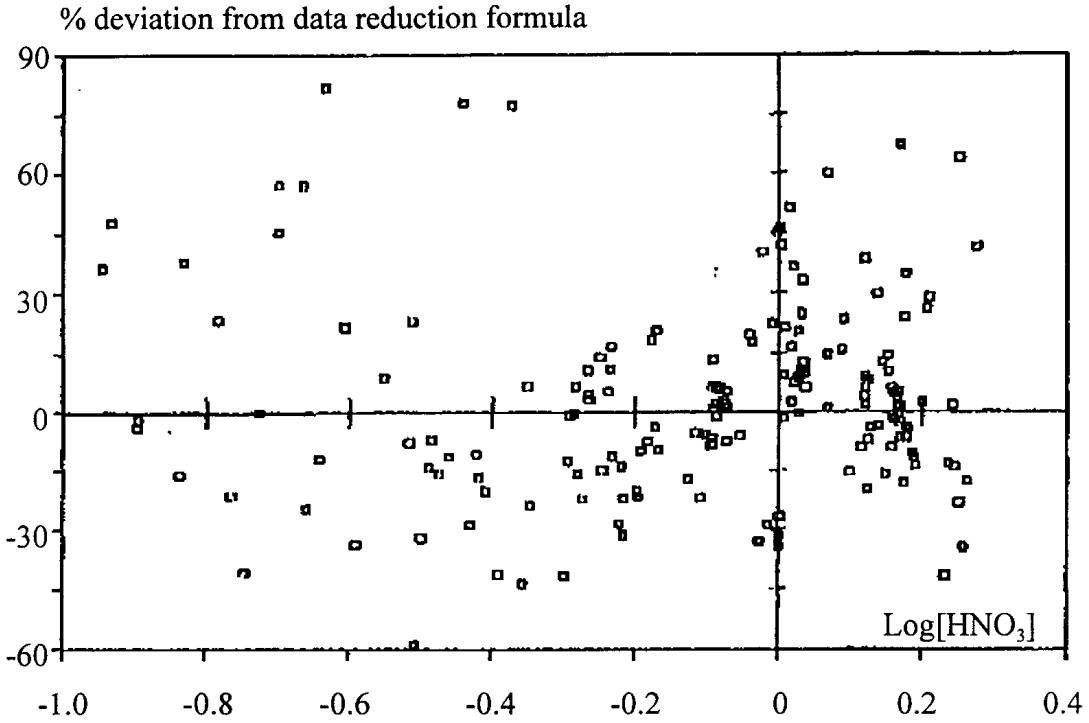

Figure 6. Scatter in plutonium distribution data (N. J. Hill, data from ref. 10)

Once the data are obtained, devising functions to represent them mathematically is itself a major task needing a concentrated specialised effort to meet the needs of subsequent computational procedures.

Close attention must be given to the fundamental issue of chemical analysis. If at all possible it should be conducted within and as part of the development project, not deputed to a separate analytical service, even within the same organisation and particularly in an industrial situation. Even with the best of intentions, such services with their different criteria and priorities may prove unsuitable in practice. Besides often inconvenient reporting times, techniques are not always robust enough to be applied without consideration of the particular circumstances, and those satisfactory for routine analysis may be much less so for the very different needs of development.

The question of reductants for plutonium has already been mentioned. Although common to all aqueous processes involving its separation, it could need special study in the thorium project given the question of whether uranium(IV) would still suffice. If however there were no intention to separate plutonium, the issue would not arise.

The effect of solvent degradation must however be studied whatever the nature of the process. In the past, precipitation of zirconium dibutyl phosphate has been known to cause severe problems, and the analogous thorium compounds have been noted elsewhere.[3] Amide-based alternatives to TBP in Purex, although investigated primarily for other reasons, are said to have less troublesome degradation products, [11] and if a clear choice emerges from the range examined, it should be considered for thorium despite the general preference for familiar systems. Indeed, there could be an advantage in starting with a virtually clean data sheet uncluttered by old values of uncertain reliability. On the other hand, a full experimental programme undertaken before a definite choice of extractant could waste a great deal of effort.

In the pilot-scale studies that would be needed once enough basic data had been gathered to construct a detailed flowsheet, some surprises are still to be expected as progressively more realistic feeds are introduced. The behaviour of some fission products cannot be adequately represented by inactive simulants, and a real fuel solution must be used at least in the concluding tests.

Especially at the early stages, it is a great convenience if the behaviour of the process can be observed directly. The lack of colour in thorium nitrate is a severe impediment, requiring instrumental methods to follow the material's progress, although it avoids masking uranium and plutonium. Instrumentation would in any case be needed in opaque equipment, the more comprehensive the better. It is expensive, but as elsewhere, skimping can lead to greater costs later on.

Demonstrating the intended operation of the flowsheet may be relatively straightforward. Difficulties tend to arise in convincing regulatory bodies that no danger to public or operators can arise however seriously the plant is maloperated. The range of possibilities is too great for practical trials in every hypothetical situation, but if a tolerably accurate computer model is available, simulations may be used to identify a limited number of worst cases that, if sufficiently remote from any dangerous situation, provide assurance that all others will be equally or more so. Confirmatory tests would of course be essential. One of the more interesting problems to be expected, particularly where effects of scale on performance are important, is in devising tests realistic enough to satisfy regulators yet acceptable to the safety management of the experimental facility concerned when safety is the very point at issue.

Waste management

Managing the wastes presents few new issues. The range of fission products in the thorium cycle is much the same as from uranium or plutonium, with some significant differences in proportions especially in the lower peak [12]. The increased yield of krypton-85 may need attention, as would any impact on the durability of waste forms due to the presence of fluoride and aluminium. Such impact is probably slight, since fluorine forms a stable and insoluble calcium salt while aluminosilicates are under consideration as waste forms in their own right, but it must be checked.

If thorium were adopted on the grounds of efficiently incinerating minor actinides, then their residue and any freshly generated from added plutonium would have to be recycled through a repetition of methods already practised. Logic would demand that other long-lived radionuclides including protoactinium-231 should also be separated. Known methods for this element include solvent extraction from hydrochloric acid, which would be unacceptable in industry because of its corrosive properties, or absorption on Vycor glass and subsequent elution. The latter would introduce an unwelcome element of batch processing into an otherwise continuous operation, but might be acceptable. A different method on principles yet unknown cannot be excluded, or (particularly in view of the proliferation risk that separation would introduce) the nuclide might be left in the waste with others such as caesium-135 and tin-126 that cannot realistically be incinerated. The impact of these residual nuclides on the rationale of changing from uranium to thorium would need careful examination.

Intermediate-level wastes would generally be the same as in Purex and need no special investigation. Neither would solvent once degraded beyond recovery, unless it were changed to a new type. Since such a change on an industrial scale appears unlikely for thorium alone, it again raises no special issues.

CONCLUSION

Reprocessing thorium fuels would demand more development than has sometimes been recognised. Even supposing the familiar principles of solvent extraction to be adopted, existing methods could not be applied without modification, especially if essentially complete separation of thorium, uranium and plutonium were required. However, as there is no obvious purpose in separating plutonium from uranium when both are largely fissile, the recommended course in the first instance is to aim for a separated product stream containing most of the thorium, and a smaller combined stream comprising the remaining thorium with all the uranium and plutonium. Compared with other possible schemes, this would require a greatly reduced but still very substantial amount of development work.

The reversal in the solvent-extraction relationships between fissile and fertile elements, compared with that familiar in the Purex process, raises issues of criticality control needing careful evaluation. A computer simulation would be needed both for optimisation of the process eventually adopted and to investigate its likely performance under maloperation. For this purpose, a mathematical model of the solvent extraction processes should be based on data covering the whole range of attainable compositions, whether expected to arise in practice or not.. Pilot-scale studies would be needed both to verify the flowsheet under design conditions, and to show that the worst credible case of each maloperation could not lead to unacceptable risks.

Waste management would for the most part be on familiar lines. Apart from the probably slight effect of fluoride and aluminium on the preparation and properties of a compact, durable high-level waste form, the only novel issue concerns protoactinium-231. Being a long-lived radionuclide, it is as much a candidate for partition as other actinides, but the methods of partition appear unattractive, and in creating a means of separating ^{233}Pa and hence pure ^{233}U from short-cooled fuel, any facility for the purpose could be deemed a potential proliferation hazard. Along with other radionuclides that could not practicably be separated or transmuted, protoactinium might therefore be best left with the general mass of fission products, although this and related difficulties could diminish the attractions of using thorium.

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