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Advanced fuels with reduced actinide generation

Proceedings of a Technical Committee meeting held in Vienna, 21–23 November 1995

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FOREWORD

Nuclear energy can play an important future role in supplying the world population with energy. However, this form of energy will be successful only under certain conditions: it must meet very strict safety requirements, it must be economically competitive, and it must be acceptable to the public. Nuclear power produces radioactive wastes and in several countries the public raises concern about safety. Much development work on advanced nuclear power systems is going on in several countries, with participation of both governmental and private industries to meet these conditions.

Advanced nuclear power systems are currently under development with the potential to make a significant contribution to meeting the energy needs of the world in an environmentally acceptable manner. These systems are being developed to meet the challenges of increasingly demanding safety requirements, economic competitiveness and public acceptance. Technological solutions to radioactive waste management and disposal are under intensive investigation in several countries.

Because of the high cost of development of advanced nuclear power systems, especially the innovative concepts, Member States may find it attractive to co-operate internationally in technology development. The IAEA's programme in nuclear power technology development encourages international co-operation through technical information exchange and co-operative research.

In the framework of this IAEA activity the Technical Committee Meeting on Advanced Fuels with Reduced Actinide Generation was organized. The aim of the meeting was to highlight current research activities and to identify new research areas and fields of possible co-operation. The scope of the meeting included advanced fuels for all types of nuclear reactors: light water reactors, heavy water reactors, high temperature reactors, fast reactors, molten salt reactors and for accelerator driven systems. Other topics covered a wide range of investigations made, or to be made in the Member States.

The IAEA wishes to thank all those who participated in meeting. Special thanks are due to the session chairmen. The officers of the IAEA responsible for the preparation of this publication are V. Arkhipov and A. Grigoriev of the Division of Nuclear Power and the Fuel Cycle.

EDITORIAL NOTE

CONTENTS

SUMMARY

1. INTRODUCTION

The Technical Committee Meeting on Advanced Fuels with Reduced Actinide Generation was held in Vienna, from 21 to 23 November 1995 and was attended by 31 representatives and observers from three international organizations (the European Commission, CERN and the IAEA) and 14 Member States (China, Czech Republic, France, Germany, India, Italy, Japan, Republic of Korea, the Netherlands, Poland, the Russian Federation, Sweden, Ukraine and the United Kingdom). Sixteen papers were presented in seven sessions, followed by a panel discussion.

The objective of the meeting was to exchange information on the feasibility and reliability of advanced fuels and related operation modes, aiming at a significant reduction in the generation of actinides in general and of plutonium, especially.

The world is faced with an increasing population, an increasing demand for power but with diminishing resources of traditional fossil fuels and increasingly stringent controls on release of CO_2 and pollutants into the atmosphere. Against this background, there is a need for the increasing deployment of other energy sources. Renewables will clearly play a part but they are not able to meet the full demand. Considerable technology development and demonstration is required before fusion energy can achieve commercialization and the reducing level of funding by many countries is moving the prospects for its eventual use further into the future. Nuclear power will have to play an important role to meet the demand under environmentally benign conditions.

However, a major concern is the public perception of nuclear safety and in the longterm the storage of nuclear waste. There is a further concern about the increasing production and stockpiling of plutonium which presents an ever present threat of diversion into weapons making. Ways in which both the waste problem and the plutonium buildup problem can be alleviated are clearly of great interest and have been the subject of this meeting. The technologies available for dealing with this problem range from those concerned with reducing the amount of radioactivity which has already been generated to the use of the Th cycle in which the problem of undesirable actinides is largely eliminated.

The actinides include plutonium, americium, curium, neptunium and protactinium. There are currently approximately 126 tonnes of commercially produced plutonium in the world of which only a portion has been separated from irradiated fuel by reprocessing.

In some Member States incineration of actinides is being investigated while in the others the emphasis is on reducing plutonium stocks. This view of the value of plutonium clearly has a strong influence on the approach to actinide reduction.

2. SUMMARY OF PAPERS

In this section a brief resume of the coverage of the papers and significant results is given.

The motivation for reducing actinides may come from one or more of the following reasons:

- reduction of plutonium inventory to reduce storage costs and the risk of proliferation,
- reducing the hazard in the final repository for wastes,
- political reasons, e.g. to increase public acceptance of nuclear power,
- to take advantage of the latent energy resource in plutonium.

Recycling of plutonium was a topic in several papers. It is a well established option for LWRs and work on PHWR was also presented. Traditional MOX Pu recycling is a method of using Pu as a fissile resource without losing the possibility of its use for further conversion of fertile isotopes by excess neutrons.

There is considerable interest in using an inert carrier for the plutonium where the intent is purely to reduce plutonium stocks. Materials such as MgO, ZrC and ZrN have been proposed and tested. This type of fuel can be used in both fast and thermal reactors. With sufficient residence time or multiple recycle most of the Pu isotopes can be eliminated almost entirely except for ^{242}Pu which takes much longer to convert to ^{243}Am. Fast reactors can be designed as non-breeders and, with inert carrier fuel, can be effective Pu burners. This line of development has a relatively high level of funding support. Where plutonium is regarded as a valuable resource, there is still some interest in burning the minor actinides (MA).

Thorium based fuels offer a very low level of actinide production relative to uranium based fuels. Although ^{233}U is a potential direct use material, it can be effectively denatured by addition of ^{238}U and furthermore, the ^{232}U activity which is always associated with it means that ^{233}U fuels have proliferation resistant characteristics. A ^{232}Th / ^{283}U cycle requires other fissile materials to initiate it and the use of Pu for this purpose effectively reduces actinide levels while allowing the start up a reactor system which does not have the same level of actinide generation. One paper described carrying out this process in a pebble bed HTR where the plutonium would be contained in one type of ball and the thorium in another. High burnup of the Pu nevertheless requires extra ^{235}U in the thorium balls. On the other hand, a paper pointed out the reduced radiological impact of thorium mining residues, as compared to those resulting from uranium extraction.

Investigations of actinide burning in reactors requires computer codes to simulate the fuel behavior in the core, especially the buildup and burnup of actinides. One paper presented a comparison of calculations with experimental results for U- and Pu-buildup in the AVR pebble bed HTR feed with LEU-fuel. It was impressively shown, that the computer simulation and the real behavior are fairly in accordance.

There was strong interest in liquid fuel reactors based on thorium/uranium salts using the ^{232}Th/^{233}U cycle. There are proposals to initiate the ^{233}U breeding process by the use of plutonium fuel in such systems. Molten fluoride salt reactors were claimed to have advantages in the possibility of continuous refuelling to avoid high excess reactivity, the possibility of

continuous removal of some fission products, especially gaseous ones, and well established structural and moderating material compatibility. Superheated steam temperatures are possible for electricity generation.

If burning of actinides (and long lived fission products) is to be effective in reducing the overall hazard of the nuclear fuel cycle, the burn rates of the relevant nuclides must be greater than the natural decay rate of those nuclides. This points to a need for high fluxes. Accelerator driven neutron sources can provide these sources especially in conjunction with a subcritical multiplying assembly. One proposal is to use a spallation source to regenerate spent LWR fuel so that it can be re-inserted into a reactor without intermediate reprocessing.

Accelerators are also proposed in conjunction with subcritical molten salt reactors as powerful means for both Pu incineration and ^{233}U breeding. A synergetic linking of such an accelerator assisted breeder with several thermal molten salt reactors was proposed as a route for alleviating many of the current problems of reactor deployment. However, it has to be recognized that such a proposal is many years from implementation and the more urgent task is to use near-term solutions to the problems of today's actinide accumulation.

The final papers were on the question of toxicity of the whole fuel cycle over the period of about 10^6 years up to the time when the hazard is no greater than that of the original ores. The current technologies of plutonium recycle have virtually no impact on this long-term toxicity which is dominated by Pu and Am. To make a real impact, innovative techniques to reduce these species by several orders of magnitude are needed. The importance of reducing the actinides content of the waste streams was emphasized by many speakers. For example, if 1% of Pu is discharged from reprocessing wastes, than trying to burn the remaining part to below this level will not contribute to a significant reduction of the overall toxicity. However, the calculations of this toxicity and the time taken to decay to a background level is dependent on the ALI limits which are subject to change.

3. PANEL DISCUSSION

Y. Zhang (China)

The topic "Advanced Fuels with Reduced Actinide Generation" is important and interesting for China, although China is in the preliminary stage in the development of nuclear energy. At present, following the development of electricity in China the problem of reducing the long term radiological hazard associated with the disposal of high-level radioactive waste is being addressed. In China there is interest in the area of transmutation of minor actinides and long-lived fission products and use of plutonium in fast rectors and thermal reactors in the future. In the Th-U cycle area, China's Tsinghua University has made some progress. The design work of experimental fast reactor CEFR (China Experimental Fast Reactor) is under way. The area of ADTT (Accelerator-Driven Transmutation Technology) will be given more attention in China.

M. Hron (Czech Republic)

There are two principle directions to solve the crucial issues of nuclear power:

1.   Any improvement in the solid nuclear fuel concept, up to now almost exclusively accepted, leading to a reduction of actinide generation
2.   To develop rather a new technology based on the concept of liquid nuclear fuel (e.g. in the form of molten fluoride salt mixture)

The principle difference between these two concepts is as follows:

A.   There will always be handicap with the accumulation of the products of the residence of solid immobile fuel block in a reactor core for a relatively long period of time.
B.   Liquid fuels give a basis for the solution of the issue of actinide and nuclear waste, especially when applied in connection with other important innovations leading to efficient transmutation and even to a nearly wasteless version of nuclear power technology like e.g. subcritical, neutron source controlled reactor systems and continuous separation and removal of undesirable components of the fuel-coolant medium

J.-P. Schapira, IPN-Orsey and C. de Turenne, COGEMA (France)

The French nuclear programme is very important (54 reactors in operation). The activities involved are shared among different organizations:

-    R&D, with CEA (Commissariat à l'Energie Atomique),
-    waste management, with ANDRA (Agence Nationale pour la gestion des Déchets Radioactifs),
-    electricity generation, with EdF (Electricite de France),
-    Nuclear steam supply system and fuel design, with FRAMATOME,
-    nuclear cycle, with COGEMA (Compagnie Genérale pour les Matières nucléaires).

To give an answer to the public concern, an act of Parliament was passed on 30 December 1991, on research for radioactive waste management. A global evaluation report, and a bill, must be presented within 15 years, in order to take a decision in 2006.

Three R&D areas are mentioned in the law:

-    partition and transmutation of long-lived elements,
-    underground laboratories for final waste repository purpose,
-    long duration waste packaging and surface storage.

The present situation for partition and transmutation programmes in France is the following:

R&D field:

CEA has launched the SPIN programme for minor actinides partition and transmutation. In parallel, the CAPRA programme studies the use of fast neutron reactors to

burn more plutonium. PHENIX, SUPERPHENIX and test reactors perform these experimental programmes.

The CNRS (Centre National pour la Recherche Scientifique) and the CEA are studying advanced systems with neutron accelerators.

Industrial field:

-   in 1985, EdF decided to recycle plutonium in PWRs in accordance with the general French policy. MOX fuel is loaded in the 900 MWe units, up to 30%,
-   today, 16 reactors are licensed for MOX fuel, 7 of which are already loaded. At the end of 1995, half a thousand assemblies have been manufactured, saving natural uranium and enrichment work needed for 250 tonnes of fresh UO_2 fuel,
-   tomorrow, 12 more reactors are to be licensed, so that by 2000, 28 reactors will participate to plutonium recycling.

The new French-German reactor EPR (European Pressurised Reactor) is designed to be loaded with 50% MOX fuel assemblies. An extension to 100% is under evaluation.

For plutonium recycling, it is necessary to adjust spent fuel reprocessing with MOX fuel fabrication and reactor capabilities. For the time being, the decision is to recycle in PWRs. Nevertheless, in the future, fast reactors could be used for large quantities.

P.-W. Phlippen (Germany)

1. In Germany, nuclear energy usage is in a difficult situation, so that new items or systems may not be adopted at the moment. This creates difficulties for research activities. Nevertheless research activities in the German research centers are tolerated by their administrations if they are not connected with additional funding by the German Government.

2. From the scientific point of view, based on present experience in the research centers, the topic of P&T is of special interest and research should be carried out to identify for example:

    -   possibilities
    -   draw backs
    -   data needed
    -   possible systems/technologies to achieve goals
    -   goals to be met and identify the significance for the further development of mankind

    Special interest should be given to the choice of the fuel cycle U-based, Th-based or even with inert matrices. For example: burning of Pu together with Th, thus breading U^{233}, would combine energy production with the replacement of Pu by a fuel, that could be more acceptable to the public.

3. Research is going on applying fast reactors for transmutation (e.g. burning Pu) and of an HTR specialized for Pu-burning. On the other hand accelerator driven systems are

being investigated. Partitioning is considered for separation of Np, Am, I and Tc. Methods using experience with HPLC (High Pressure Liquid Cromatographics) are being applied to load Np, Am as well as Pu onto solid columns.

4. In addition to the Th-CRP already running, working tasks, possibly organized as a CRP, addressing the remaining items should be installed and cover for example:

- the fuel cycle starting from mining to final disposal including recycling with its draw backs;
- evaluation of potential toxicity of disposed waste with realistic man doses received by workers while recycling fuel;
- identification and qualification of the transmutation capabilities of different systems applying the same methods of comparison;
- as far as possible at this stage estimate the additional costs related to P&T. A reflection on the cost of energy supply by renewables may be of special interest.

D.S.C. Purushotham (India)

Pu is treated as a valuable resource in India. It is essentially planned to use Pu in fast reactors. However, thermal recycling option is being exercised in the interim due to delay in fast reactor programme. Pu recycle as MOX in PWRs and PHWRs is planned and being implemented progressively. This is expected to reduce Pu inventory. Also in view of the abundant thorium resources we have a strong interest in the Th-U cycle and we expect this cycle to significantly reduce actinides while contributing to the power generation.

Further, as the policy of reprocessing is adopted for spent fuel, the separation of actinides is effected. Transmutation is one of the activities under consideration as an eventual solution. There would be interest in new technology based on molten fluoride salt mixture as liquid fuel due to greater efficiency of the system and its contribution to reduction of actinides.

F Vettraino (Italy)

The outcome of the referendum held in November 1987 was actually interpreted by the majority of the political forces and hence by the government, as a go for a total closure to nuclear energy exploitation in the country. The blockage began in 1988 and was extended even to research reactors (e.g, the ENEA experimental fast breeder reactor PEC under advanced construction stage at about 85% completion). Hard measures on human and financial resources: the ENEA employees on the Nuclear Energy Programme (Fusion excluded), have been drastically reduced, with an extremely limited financial support. Same trend for industry and utilities.

Nevertheless, even by carrying out this dramatic cutting, the Italian government agreed for maintaining a small "garrison" on nuclear fission energy R&D aimed at innovative reactor concepts featured by enhanced inherent passive safety, fuel cycle safety and radwaste disposal.

Accounting to this a space has been allowed in ENEA to the Fuel Cycle Safety with particular attention to Pu issue and long lived actinide reduction together to the waste disposal,

being the feeling these are first priority subjects for paving the way to any possible re-proposal of nuclear fission in Italy.

As to the items which made the object of this TCM, we should say that the work which we are doing at ENEA on advanced inert matrix fuel for Pu and MAs burning in LWRs, is good in line with the urgent necessity of reaching in the new-term a safer fuel cycle. Given the peculiar Italian situation, we outlook that the Pu reduction goal is more realistically achievable through burning it in LWRs via a suitable inert matrix U-free fuel, in a once-through cycle scheme. This innovative fuel concept promises for good Pu burning performances, avoids new Pu generation, does not increase significantly the radiotoxicity with respect to un-reprocessed spent fuel, can act as a carrier for burning MAs also, and from first tests on simulate fuel it shows strong chemical stability either fresh or irradiated fuel (very high resistance to current dissolution techniques).

The latter feature, coupled to the poor quality residual Pu in spent fuel, should result in a high anti-diversion potential and should ensure the stability of fission products and long lived actinides, by blocking them in a sort of rock-like spent fuel ready to be sent to final disposal in adequate deep geological formations, by trying to "copy " at the best what the nature has well done during the last one-million of years (e.g. very old rocks stability and behaviour of radwaste from Oklo natural reactors in Africa).

The suitable candidate inert matrices are alumina, zirconia and spinel, employed either as simple compound or mixed compound. A technological program is under-way at ENEA for developing such inert matrix fuel, going from fabrication and out-of-pile characterization to the irradiation tests expected to take place in Halden HWBR.

Inert matrix fuel, should be considered as one main contributor in the attempt of a useful and safe Pu and MAs reduction, being it more compatible than other solutions with the current world-wide established technology, namely commercial LWRs and fuel fabrication processes. Other more exotic solutions, even if good promising at paper level, appear too far from bringing realistic contributions within the near or mid-future. Promoting collaborations among different countries is highly recommended.

K. Furukawa (Japan)

1. Japan has strong emphasis on a Pu-utilization plan: however, practical deployment of Purex-plant and FBR is delayed resulting in an excess of Pu. Therefore, this problem should be examined more deeply. We already officially reported our assessment to MITI on this March, 1995.
2. Recently, interest in the Th-cycle and, more straightforwardly, on the rational Thorium Molten Salt Reactors (Th-MSRs) approach has been quickly increasing everywhere.

This somehow connected with the following trends:

1. Necessity of safer and economical nuclear power stations
2. Necessity of management of spent fuel by molten salt dry processing
3. Necessity of radiowaste incineration by molten salt application in several facilities

3.   A Specialists' Meeting on Th-MSR should be organized on an international base.
            Generally the following approaches should be separated:

            -    LWR-HWR type approach
            -    HTR type
            -    FBR
            -    A more advanced one such as MSR and accelerator driven systems.

They each have incentives depending on the national necessity. Their comparison/competition is then another problem.

Y. K. Kim (Republic of Korea)

1.   Energy security is the primary concern of the energy policy in Korea. In order to meet the increasing demand for electricity, the Korean Government has been committed to an ambitious nuclear power programme. Around 40% of total electricity is generated from the 10 nuclear power plants, which are currently in operation. It is firmly planned that additionally 13 more nuclear power plants (10 PWRs, mostly 1000 MWe Korea Standard nuclear power plants and 3 PHWRs) will be on-line by the year 2006.

Nowadays, approximately 300 tonnes of spent fuels are produced per year, and therefore it is expected that the cumulative amount of spent fuel will grow very rapidly. This spent fuel is expected to be stored on-site until the policy of the back-end fuel cycle is decided. Therefore the management of spent fuels becomes one of the national issues.

Considering the management of spent fuel, which should contribute to the recycling of resources and the reduction of radioactive waste and the protection of the nature, the long-term economics and technological possibilities should also be investigated.

The recycling strategy for the peaceful use of nuclear energy should be complete with the domestic and international agreements related to non-proliferation. In this regard, the development of advanced fuels with reduced actinide generation through international cooperation has significant importance for Korea.

2.   Currently we are carrying out several R&D activities consistent with the following three objectives:

      -    to decrease the total volume of spent nuclear fuel produced;
      -    to improve uranium utilization, and to secure energy resources;
      -    to reduce by incineration the waste volume and the required management period of long-lived waste.

Specifically, in the short and medium-term, technology development of the direct use of spent PWR fuel in CANDU reactors (DUPIC) is under progress in order to decrease the amount of spent fuel and to improve the economics and safety of the nuclear fuel cycle. For both effective resource utilization and the reduction of spent fuel volume to be stored, the

14

feasibility of MOX recycling in PWRs is under review by means of a feasibility study. Research is also under way to search what transmutation system can be the best for the Korean nuclear programmes, in the long run.

W.M.P. Franken and E.H.P. Cordfunke (Netherlands)

Within the Netherlands a research programme on recycling and transmutation of long-lived nuclides, in particular actinides and fission products, is executed with as general objective to indicate that this option is an acceptable waste management strategy.

Two goals are considered: the reduction of the overall radiotoxicity of nuclear waste over a very long period and the reduction of radiological effects to the population due to leakage of long-lived nuclides.

The programme consists of strategy studies (such as: Pu-burning-capacity and incineration of minor actinides in various reactor types, transmutation of fission products in high-flux devices, perspectives of thorium based fuels, etc.) as well as technological development work, including irradiation tests in the High Flux Reactor HFR, Petten, on fission products (technetium and iodine) and on actinide-fuels (plutonium, americium) based on the Inert matrix concept. All the projects are performed in international, mostly European, collaborations.

At the end of the programmes some questions should be answered:

- Does the evolutionary route (MOX and MINOX fuel in LWR or FBR) lead to an important reduction in long-term radiological hazard?
- What may be the contribution of new type reactor systems to the reduction of the waste problem?
- Can the introduction of other fuel cycles, such as thorium cycle, lead to improved options?
- Which consequences for waste management should be foreseen if the nuclear energy production is terminated?

S. Taczanowski (Poland)

1. According to the national policy with respect to nuclear energy the appropriate strategy of management of nuclear waste is to be proposed before the decision for the introduction of nuclear energy in our country. Therefore, research in the field of the nuclear fuel cycle, the reduction of actinide generation included, should precede the making of the final decision.

2. The research should maintain its diversified multidirectional character. Economical aspects should be considered possibly at the same time. The irretrievable disposal of actinides is not recommended, in contrast to long-lived fission products. A smooth launching of externally driven transmutation technology e.g. in a symbiotic system with LWRs deserves attention.

3. - Meetings on transmutation as a measure for reduction of nuclear waste are desirable
   - A similar CRP seems advantageous too.

L. Zabudko (Russian Federation)

Concerns about radiotoxicity of Pu itself and of its decay products have promoted initiatives for considering several options for Pu and MA disposal.

We have special interest in the fast reactor system. Three fast reactors, BR-10, BOR-60 and BN-600 are operating successfully now. The unique characteristics of fast reactors can be utilized for other future needs. In addition to Pu disposal it has become apparent that long-term disposal of wastes from all types of reactors is also important. It is very interesting to examine the technologies needed to solve these issues using fast reactors. The work is carried out aiming at the use of VVER-type reactors for Pu utilization.

Several topics of this TCM can be the subject of separate discussion:

- study of an optimal system for reduced actinide generation
- re-evaluation of the toxicity of different fuel cycles
- feasibility study of different fuels for reduced actinide generation
  (beginning with fuel fabrication technology up to its reprocessing).

The search for suitable carrier materials for Pu is a very interesting subject. Fuel selection will require several years of work, and the long time required for irradiation and PIE. It appears that an opportunity exist for international collaboration in some aspects of this work.

V. Borisov (Russian Federation)

It would be desirable to consider a topic on Recycle of transplutonics to decrease the quantity of a long-lived alpha-bearing radioactive waste in the future international programme. The research in this area is important, because the storage of such waste in the deep geological formations will be very expensive. If the rare earth fraction be free from americium, curium and other alpha-emitting nuclides, it would permit to put this waste in the near surface disposal in some ten year already. After sufficient curium-244 decay, transplutonics could be burned by recycling in fast reactors.

O. Vertsimaha (Ukraine)

In Ukraine atomic power stations produce about 40% of total electricity, storing considerable amount of spent fuel. There are rich resources of U and Th in the country, so that creation of country's own fuel cycle is expected to be profitable from economic point of view. Therefore the topic Advanced Fuels with Reduced Actinide Generation is interesting and important for both the development of fuel cycle and the problem of spent fuel management.

There are two most important questions for further investigations:

(1)   Thorium-based fuel cycle for the PWR reactor;
(2)   regeneration of spent fuel.

Both questions are studied theoretically in Ukraine. For the experimental research it is planned to create appropriate equipment. The experimental investigations are possible under condition of financial support.

J. Brennan (United Kingdom)

It is necessary to distinguish between the UK position on the need for reduced actinide generation and BNFL's position. There is little UK interest in consuming either plutonium or minor actinides by irradiation merely to reduce the inventories of these materials. However, as an international nuclear fuel services company, BNFL takes account of the fact that the situation may be different in other countries. If any of its customers wish to recycle plutonium or minor actinides as a means of controlling the stocks of these materials, BNFL will be pleased to assist in any way it can.

The UK has accumulated plutonium stocks over the past forty years with a view to future use in a fast reactor programme. Although current thinking is that it will be at least twenty years before the economics of fast reactors favour deployment in the UK, the plutonium stocks are still viewed as a resource to be conserved rather than a liability to be disposed of. Interim use as thermal MOX in PWRs is a possibility although no definite plans exist at the moment.

Waste management is not seen as providing any incentive to remove plutonium or minor actinides from wastes in the UK. Past studies have indicated that these materials can be well contained in a deep geological repository and that, therefore, the actual environmental benefit associated with removing and destroying them will be negligible. In view of this, the cost and real hazards associated with partitioning and transmutation are not justified.

Two remarks on the assestments made in the papers presented in this meeting:

The conclusion which I can draw from the works made in UK and from the paper presented here is that the results of current assessments of the impact of MOX recycle on long-term waste toxicity depend very much on the initial assumptions made. By choosing appropriate assumptions, it is possible to argue a case either for or against MOX recycle. Because of this, I believe it is inappropriate to make generalised statements about the effect of MOX recycle on long-term waste toxicity at this stage.

We need to bear in mind that the potential hazard from waste toxicity may not reflect the magnitude of the actual hazard, due to the mechanisms needed to transport the radionuclides back to man. This is vitally important if we are to avoid giving long-term waste toxicity too much emphasis in environmental impact studies, leading to decisions to suffer real hazards today in exchange for hypothetical future benefits which probably will not exist in actuality.

Y. Kadi (CERN)

Some remarks on the assessment of Pu and MA transmutation in LWRs and FRs was given.

Recycling of Pu as MOX fuel in thermal reactors is a proper way to reduce the mass of the residual Pu in the spent UO_2 fuel elements. However, the residual *potential radiotoxicity of spent MOX is 5 to 7 times higher than that of equivalent LWR-UO_2 fuel*.

Multiple recycling of MOX fuel in LWRs leads to a considerable increase in higher actinides and [238]Pu, which makes this option from the radiotoxicity point of view less attractive than recycling in fast reactors (FRs).

Multiple recycling of MOX spent fuel in specially designed Fast Reactors is capable of reducing the transuranium inventory (since with a harder neutron spectrum the fission to capture ratio increases, resulting in a reduced actinide generation), but the time span to realize that option is very long unless very high burnups are reached (e.g. incineration rate of = 15 at.% at 150 GWd/t). Moreover, the recycling of Pu and Minor Actinides in conventional Fast Reactors (FRs) brings about several adverse effects (i) on cycle operations (heat and dose rate at fabrication, reprocessing, transportation), and (ii) on global core performance and safety (Sodium void, Doppler effect, etc.) which limits their content (3 to 5% if recycled homogeneously) at load.

There is, thus, a net incentive in using a Th-Pu fuel (in conjunction with Accelerator-Driven Systems) not only to improve the incineration yield (and toxicity) but to reduce the recycling time interval as well.

4. CONCLUSIONS AND RECOMMENDATIONS

Many ways to achieve actinide reduction were presented and. Possible reactor/fuel combinations, e.g., thermal reactors, fast reactors and accelerator-driven systems with fuels or targets from the U/Pu and Th/U-families, in some cases with inert filler and in solid as well as in a molten state, were described and discussed. In order for actinide burning to be effective, reprocessing seems to be inevitable. However, most of the data presented were from calculations and it would be desirable to have more experimental results in the future. Another observation is that the problems are far from solved, especially from the hazard point of view. Although many options were presented with regard to fuel cycle, reactor type, etc., there was some debate as to whether a significant reduction in potential hazard could be achieved. Because of the importance of this issue, participants recommend the formation of one or two working groups to validate these initial indications and the reference system used. The working groups should also attempt to estimate the most likely real (as opposed to potential) environmental impacts of fuel cycles with and without actinide burning.

PAPERS PRESENTED

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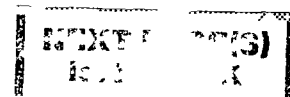
EMERGING APPLICATIONS OF ADVANCED FUELS FOR ENERGY GENERATION AND TRANSMUTATION. OVERVIEW OF IAEA ACTIVITIES

PONG EIL JUHN, V. ARKHIPOV
International Atomic Energy Agency,
Vienna

Abstract

Nuclear power generation is an established part of the world's electricity mix. However, the highly radioactive waste generated during power production is of great concern of public perception of nuclear energy. In order for nuclear power to realize its full potential as a major energy source for the entire world, there must be a safe and effective way to deal with this waste. Therefore, science must come the rescue in the form of new, more effective technology aimed at reducing the amount of long-lived radioactive waste and eliminating nuclear weapons' grade material through transmutation of these isotopes in fission reactors or accelerators. In the framework of IAEA activities on the use of this new technologies the Agency has periodically review and assess the current status of the new fuel cycles, its applications worldwide, its economic benefits, and its perceived advantages vis-a-vis other nuclear fuel cycles.

I   Introduction

Today the world uses a great deal of energy and the usage is increasing dramatically in developing countries. During the 1980s energy use in the developing countries increased 60%.

Nuclear power generation is an established part of the world's electricity mix providing 17% of world electricity (cf. coal 39%, oil 11%, natural gas 14% and hydro 19%). Its use is increasing. Nuclear power is the most environmentally benign way of producing electricity on a large scale. If we replaced nuclear by coal-fired generation, carbon dioxide emissions would increase by about 1.8 billion tonnes per year. Carbon dioxide currently accounts for 50 per cent of the human-contributed portion of the global warming effect of the atmosphere.

The combustion of coal also releases radioactive heavy metals contained in it, while the use of natural gas releases radioactive radon. The amount of radioactivity released is negligible compared with natural background radiation levels, but is often greater than that which we can expect from normal operation of a nuclear power station.

The UN Intergovernment Panel on Climate Change (IPCC) has comprehensively reviewed global warming and has reached a consensus that the phenomenon is real and does pose a significant environmental threat during the next century if fossil fuel use continues even at present global levels. According to the IPCC, to stabilise the carbon dioxide concentration in the atmosphere at current level a 50-80 per cent reduction in all emissions would be required. Nuclear power has a role to play in reducing greenhouse gases.

With the world population growth from current 5.5 billion to 8.5 billion by 2025, energy demand will increase significantly in coupling with increasing standards of living over the next two decades.

Our estimations showed that the share of nuclear power in electricity generation worldwide would be between 12% and 17% at the year 2015 (the low and high cases).

One of the greatest obstacles facing nuclear energy is the highly radioactive waste generated during power production. In order for nuclear power to realize its full potential as a major energy source for the entire world, there must be a safe and effective way to deal with this waste. While mined geological disposal is the method chosen by some countries, it has been consistently stalled by a pervasive public perception that it is

not a safe disposal technology. A primary reason for this is the long life of many radioisotopes generated from the fission, with half-lives on the order of 100,000 to a million years. Another reason is the feeling that weapons' grade material in the repository would be easily obtainable by terrorist groups. Therefore, science must come the rescue in the form of new, more effective technology aimed at reducing the amount of long-lived radioactive waste and eliminating nuclear weapons' grade material through transmutation of these isotopes in fission reactors or accelerators.

In the past years more and more studies were carried out on advanced waste management strategy (i.e., actinide separation and elimination) in various countries and at an international level.

The IAEA provides a mechanism for mutual economic benefit based on international science and technology cooperation.

II   The International Status and Perspectives of Nuclear Power

The world now produces more electricity from nuclear power than it did from all energy sources in 1958. Civil nuclear power had gained more than 7200 reactor years of experience to the end of 1994.

In 1994, 54 countries have civil research reactors operating, and 30 countries have a total of 432 commercial nuclear power stations producing electricity. A further 48 commercial power reactors are under construction.

Although some countries, notably Japan and the Republic of Korea, intend to continue ambitious nuclear power construction programs, the rate of growth of installed nuclear generating capacity over the next ten to fifteen years is expected to be low. One country, Italy, has completely abandoned its nuclear power program and others have canceled or indefinitely suspended partially constructed and planned power reactors and/or imposed a moratorium on any new nuclear construction.

Estimates of nuclear power growth over the near term, are based only on firm orders and known plans for plant expansion, life extension or closure. They do not involve any extrapolation of theoretical models, so are generally conservative and not subject to the large uncertainties that resulted in speculative projections by the nuclear industry in past decades.

After the year 2000 and until orders are placed or construction commenced, forecasts of installed nuclear capacity become much less certain. When the present construction programs are completed, most significant nuclear power growth is expected to continue only in the Asian region. The average annual growth rate of electricity production from nuclear power is estimated to be about 0.6% per year for the period from now to 2015.

Six countries with existing nuclear power programs (the UK, Russia, China, India, Japan and South Korea) and four countries without any nuclear capacity at present (Indonesia, Egypt, Turkey and Iran) have announced firm plans to build new power reactors.

III   A Potential of the Thorium-Based Fuel Cycle

Within the framework of IAEA activities, the Agency has maintained an interest in the thorium fuel cycle and its utilization worldwide. Its periodic reviews have assessed the current status of this fuel cycle, its applications worldwide, its economic benefits, and its perceived advantages vis-a-vis other nuclear fuel cycles.

Prior IAEA reviews have recognized the advantages of the thorium fuel cycle as compared with a uranium-plutonium fuel cycle, based on its nuclear characteristics. These advantages include:

*   a significant reduction in the yield of transuranic actinides, especially plutonium
*   the production of U-233, a superior fissile material for thermal reactors;
*   reliance on an abundant resource, thorium, with superior metallurgical properties in LWRs and D_2O reactors. Thorium is especially important to the nuclear economics of threshold countries such as India.

22

The thorium fuel cycle can address these new concerns:

* A combined thorium-plutonium fuel cycle can destroy plutonium in a safe, and diversion resistant manner, while generating electric power, and generating U-233 as a denatured fissile material.
* Thorium fuel cycles will permit renewed public confidence in nuclear power by avoiding the stigma of plutonium production, minimizing the high level waste disposal issue, and using a valuable resource.
* Thorium fuel cycles can be used in existing reactors with no plant design or operational changes, and the fuel can be manufactured in current facilities. Thus the economic investment of a switch to this cycle is minimal
* Thorium fuel cycles would permit the use of nuclear reactors by nations who are a concern to the world community from the standpoint of illegal development of nuclear weapons. Any change in a fuel cycle would provide an immediate early warning of a weapon problem.

In summary, the thorium fuel cycle offers the potential for a "non-plutonium" nuclear economy utilizing existing facilities and infrastructure. As such, it deserves farther serious consideration and detailed evaluation of these potential advantages.

IV   The Recent IAEA Activities

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 preparation of a Report on Status of **Thorium-based Fuel Cycle** was started to describe the State of the Art of the thorium cycle, to upgrade information presented in the document "A fresh look at the thorium fuel cycle" and to suggest areas that need farther investigations. We intend the document to be the IAEA TECDOC to provide information and to summarize experience and study regarding the subject mentioned above. The report will include the general overview and summary of the Thorium-based Fuel Cycle concepts and contributions from the particular countries/groups presenting in detail their ideas.

The scope and the content of the report were elaborated at the consultancy held from 22-24 November 1994 at the Agency's Headquarters in Vienna. A draft of the Status Report was reviewed and thoroughly discussed by the group of experts from IAEA, France, Germany, India, Japan, the Russian Federation and the USA in June 1995 in Vienna. The necessary corrections and additions were made, and the final version of individual contributions was agreed upon. IAEA will publish this document in 1996.

Coordinated Research Programme (CRP) on the Potential of Th-based Fuel Cycles to Constrain Pu and to Reduce of Long-term Waste Toxicities

This CRP will examine the different fuel cycle options in which Pu can be recycled with Th to get rid of the Pu, or replace the Pu with materials that are less unacceptable to the public. Potential of Th-matrix will be examined through computer calculations. Each participant can choose his own cycle, and the different cycles can be compared through certain predefined parameters (e.g., annual reduction in Pu inventory). As a final recommendation, the CRP could suggest a demonstration of Pu-Th burning in a reactor in one of the members' countries The toxicity accumulation and a transmutation potential of Th-based cycles for current, advanced and innovative nuclear power reactors including hybrid systems will be investigated.

The work will include:

- Analysis of neutronic transmutation potential of the thorium fuel cycle for the long-term nuclear power development, which incudes:
- elaboration of physics criteria (goal) of the transmutation;

23

- a search of the most effective physical means of the toxicities reduction (optimal neutron flux burn-up, recycling schemes, etc.) and
- Potential of the innovative nuclear systems concepts with a thorium fuel cycle (molten salt. lead cooled)
Estimated duration of the programme: Three years.

The scope of a Coordinated Research Programme (CRP) was considered at the Consultancy on Status of Thorium-based Fuel Cycles held from 6 to 9 June 1995 at the Agency's Headquarters in Vienna and attended by representatives from IAEA, France, Germany, India, Japan, the Russian Federation and USA.

Several topics were proposed and the following benchmark was selected and recommended for the first stage of CRP:

"Calculation of the isotopic composition, cross-sections, and fluxes for a typical PWR-cell loaded with $(Pu-Th)O_2$ - fuel, as a function of a fuel burn up."

The PWR-type reactor has been chosen for the first benchmark calculations of Pu-burning because it is the reactor type that gives the largest contribution to the current production of nuclear energy. The IAEA has circulated the detailed description of data for the benchmark to Chief Scientific Investigators. A Research Co-ordination Meeting (RCM) will be held in the middle of 1996 to discuss results of calculations of the first stage of the CRP and to work out a programme for the next stage.

We have also an intention to start a benchmark calculation on Accelerator Driven Systems and Related Fuel Cycles.

Status Report on Accelerator Driven Systems

The IAEA has initiated work on a status report on **Accelerator Driven Systems** (ADS), as recommended by participants of a Special Scientific Programme on "Use of High Energy Accelerators for Transmutation of Actinides and Power Production" held at the Austria Center in Vienna, on 21 September 1994 in conjunction with the 38th IAEA General Conference. The general purpose of the Status Report is to provide, in particular for planners, decision makers, and other parties that are not directly involved in the development of ADS, an overview of ongoing development activities, different concepts being developed and their project status, as well as typical development trends. The document is intended to be the IAEA TECDOC and will be published by IAEA in 1996.

STATUS OF THE NUCLEAR FUEL CYCLE IN
IAEA MEMBER STATES

A. GRIGORIEV
International Atomic Energy Agency,
Vienna

Abstract

At the international level, debate is focusing increasingly on number of industrial operation and processes associated with what is called the nuclear fuel cycle. These activities include uranium mining; enrichment and fuel fabrication; reprocessing, and subsequent recycling, of spent fuel and the management of nuclear waste. The issues related to the nuclear fuel cycle continue to be of high priority in assuring the safe use of nuclear energy. Status of the nuclear fuel cycle in IAEA member states is discussed in this paper.

Since the 1970s, when nuclear power was among the fastest developing sources of energy, the technology has made significant contributions to electricity supplies in many countries. Nuclear power's share of total worldwide electricity generation today is just about 17%, and surpasses 25% in 15 countries. However, its rate of growth has slowed and is projected to remain modest through the next decade.

At the international level, debate is focusing increasingly on number of industrial operation and processes associated with what is called the nuclear fuel cycle. These activities include uranium mining; enrichment and fuel fabrication; reprocessing, and subsequent recycling, of spent fuel and the management of nuclear waste.

The issues related to the nuclear fuel cycle continue to be of high priority in assuring the safe use of nuclear energy. The selection of the appropriate nuclear fuel cycle strategy for a particular country is a complex procedure in which many factors have to be weighed, including political, economical and non-proliferation issues as well as protection of the environment. Consequently, the development of successful spent fuel and waste management technologies will play a significant role in the development of nuclear power in the future.

Two options for back-end of the nuclear fuel cycle exist at present - an open, once-through cycle with direct disposal of the spent fuel and a closed cycle with reprocessing of the spent fuel and recycling of plutonium and uranium. Table 1 shows the back-end approaches selected by different countries.

Current back-end policy in different countries can be divided broadly into the following three groups:

- Countries that have been following the once-through fuel cycle and are focusing their attention on interim storage followed by disposal of the fuel;

- Countries that have selected the reprocessing option and are actively operating or constructing reprocessing plants or have contracts for

reprocessing abroad and/or are returning some or all of their fuel to the country of origin;

- Countries that are still evaluating their nuclear fuel cycle programmes.

In 1994, the nuclear power plant spent fuel arising from all types of reactors amounted about 10 500 t HM, giving an estimated cumulative total of over 155 000 t HM. About 102 000 t HM are presently being stored. The quantity of accumulated spent fuel is over 20 times the present total annual reprocessing capacity. Projections of the annual spent fuel arising will grow gradually from the level of about 10 500 t HM in 1994 to over 11 000 t HM in 2010. The projected cumulative amount of spent fuel generated by the year 2010 reaches 300 000 t HM. Assuming that part of it is reprocessed, the amount to be stored by the year 2010 is projected to be about 200 000 t HM. Since the first large scale final repositories for final disposal of spent fuel are not expected to be in operation before the year 2010, interim storage will be the primary option for the next 20 years.

TABLE I   Spent Fuel Management Approaches Selected in Different Countries

	Deferred decision	Direct disposal	Reprocessing
Argentina	x		x
Belgium	x		x
Brazil			x
Bulgaria	x		x
Canada		x	
China			x
Czech Republic	x	x	
Finland	x	x	x
France			x
Germany		x	x
Hungary	x		x
India	x		x
Italy	x		x
Japan			x
Korea, Rep. of	x		
Lithuania	x		
Mexico	x		
Netherlands			x
Pakistan	x		
Russia		x	x
Slovak Republic		x	x
Slovenia	x		
South Africa	x		
Spain		x	x
Sweden		x	
Switzerland	x		x
UK	x		x
Ukraine	x	x	x
USA		x	

Originally, the objective of the closed fuel cycle concept was to recycle the separated plutonium and uranium in breeder reactors. At present most of existing nuclear reactors all over the world are thermal and have potential capacity for accepting mixed oxide (MOX) plutonium uranium fuel. It offers a large possibility for recycling of uranium and plutonium. Currently thermal recycling of plutonium (as MOX) is being carried out in PWRs and BWRs in Belgium, France, Germany, Japan and Switzerland. Thermal recycling of uranium is being carried out in PWRs in France, in RBMK reactors in the Russian Federation and in AGR reactors in the UK.

A properly closed fuel cycle meets today's requirement for natural resource conservation and reduction of waste toxicity. Natural resource conservation is only achieved if the reusable fissile materials are recycled to generate electricity. Furthermore, doing this efficiently fits in with the aim of reducing the toxicity of the waste. The theoretical gains thus obtained may reach a factor 500 between open and closed fuel cycle. But there is a wide gap between the theoretically achievable gains and the gains realizable with current technology.

In a closed fuel cycle, Pu separated by reprocessing ideally should be balanced with the recycling capabilities. In the case of a closed cycle with the recycling of Pu in the form of MOX fuels in thermal reactors (TR), MOX fuel fabrication plants are a prerequisite and are planned to come on stream to provide the necessary capabilities (see Table II). From an optimistic point of view thermal reactors already licensed for MOX use and other TRs under presently licensing procedures could irradiate the Pu already separated by reprocessing facilities.

Table II   Projected MOX Fuel Fabrication Capacities (t HM/a)

YEAR	1995	2000	2005	2010
BELGIUM (DESSEL) PO	35	35	35	35
BELGIUM (DESSEL EXPANSION) P1			50	50
FRANCE (CFCa)	30	30	30	30
FRANCE (MELOX)	25	160	160	160
GERMANY (HANAU - OLD)				
GERMANY (HANAU - NEW)				
JAPAN (TAKAI) FUGEN	10	10	10	10
JAPAN (TOKAI) DEMO-ATR		30	40	40
JAPAN - LWR MOX			70	100
RUSSIA			60	60
UK (DEMO)	8	8	8	8
UK (SMP)		100	120	120
TOTAL TONNES FABRICATED	1OS	373	583	613

However, it should be mentioned that in the early 1980's, large new uranium resources were discovered. These discoveries, along with a decrease of demand for uranium, caused the price of uranium to fall. Also, the development of fast reactors has been much slower than expected. But the commitment to construct reprocessing facilities had been made and realized. This created a significant imbalance between plutonium separation and plutonium utilization rates. At present the development of MOX fabrication capacity has been slowed down by political difficulties and less attractive economics. According to the IAEA's estimation, the world-wide inventory of separated civil plutonium will be 126 t at the end of 1995. The inventory will hit the maximum (160 t) at around the year 2000 and start to decrease gradually thereafter.

It is expected that by the beginning of the next century, sufficient recycling capabilities could be available and the separated civilian plutonium inventory will start to be gradually reduced. Plutonium will then be treated as an intermediate product to be recycled immediately via plutonium fuel fabrication plants. It should be mentioned that this policy will have the additional benefit of avoiding the in-growth of americium in plutonium and the costs of its removal through the purification process.

Furthermore, with an increased number of existing LWRs being licensed for MOX use in the world, with new LWRs designed to use 100% MOX cores and with new FR specifically designed to consume plutonium, it will be possible in the future to control worldwide Pu production according to the world's needs in terms of energy.

It is also important to note that the thermal reactor closed cycle has some challengers. Radiotoxicity increases with MOX fuel irradiation. High radiotoxic long-lived minor actinides (MA) (their initial radiotoxicity is more than ten-times higher than that of plutonium-239) will pile up. The nuclear power industry accumulates actinide radiotoxicity equivalent to 2.8 tonnes of Pu-239 per GWt/y. Multirecycling of Pu is limited by nuclear physics constraints and MOX spent fuel will again have to be disposed of in geological repositories, which could become potential plutonium mines after thousands years. In other words a closed fuel cycle based only on thermal reactors is in some ways similar to an open fuel cycle, although being more efficient in Pu utilization.

These problems would disappear with a fast reactor fuel cycle where multirecycling could be possible and the inventory of long-lived radionuclides would be reduced. Plutonium also would be used far more efficiently in energy production. In the closed fuel cycle option, the burning of plutonium in the form of MOX in TRs is required as a temporary expedient until fast reactors become commercially available.

Unconventional and effective options for burning plutonium while not increasing radiotoxicity are also being discussed extensively these days. One option is the utilization of thorium in conjunction with burning plutonium. This option could be used in high temperature reactors, molten salt reactors or accelerator driven reactor concepts. The use of plutonium in inert carrier matrices to be irradiated in water reactors is also being discussed.

Some words about Reprocessed Uranium (RepU). The recycling of RepU does not face the same problem as plutonium. The utilities which own RepU can choose when it is desirable to recycle it in their reactors. It has already been demonstrated that in-core behaviour of RepU fuels is very similar to that of natural UO_2 fuel. Dedicated fuel cycle facilities to handle RepU are under review and will be implemented according to the utilities' needs.

I would like to stress again that we are convinced that the current nuclear fuel cycle concept provides an adequate protection to the population and the environment by sufficient confinement of radionuclides. Both the once-through and closed fuel cycles are regarded as safe and maturing technologies. However, there is a strong interest to see whether further potential impact reduction can be achieved and at what cost.

29
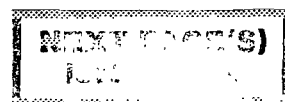
CRITERIA FOR ACHIEVING ACTINIDE REDUCTION GOALS

J.-O. LILJENZIN
Department of Nuclear Chemistry,
Calmers University of Technology,
Göteborg, Sweden

Abstract

In order to discuss various criteria for achieving actinide reduction goals, the goals for actinide reduction must be defined themselves. In this context the term actinides is interpreted to mean plutonium and the so called "minor actinides" neptunium, americium and curium, but also protactinium. Some possible goals and the reasons behind these will be presented.

On the basis of the suggested goals it is possible to analyze various types of devices for production of nuclear energy from uranium or thorium, such as thermal or fast reactors and accelerator driven systems, with their associated fuel cycles with regard to their ability to reach the actinide reduction goals.

The relation between necessary single cycle burn-up values, fuel cycle processing losses and losses to waste will be defined and discussed. Finally, an attempt is made to arrange the possible systems on order of performance with regard to their potential to reduce the actinide inventory and the actinide losses to wastes.

The actinide series of elements begins with actinium (element 89) and ends with lawrencium (element 103). Only the first four of these elements (actinium, thorium, protactinium and uranium) could be found on Earth in significant activities or concentrations before World War II. The development of the nuclear reactor and its widespread use as a large source of electricity have led to large scale synthesis of the other lower members of the actinide series, i.e., neptunium, plutonium, americium and curium. "Actinides" will in the following be used to refer only to the mentioned lower members of the actinide series. Some of these have found use, e.g., plutonium as a complementary nuclear fuel, as an energy source in space research, and as a constituent in nuclear weapons, and americium as radiation source in smoke detectors. However, at present the production is much larger than the consumption leading to increasing stockpiles, increasing amounts in stored spent nuclear fuel and as radioactive waste from reprocessing operations. Improved reprocessing technology might in the future reduce the amounts of actinides in the radioactive waste leading to an increased growth of stockpiles. In this situation means are sought to reduce production rates and to reduce stockpiles of these elements.

In general, there are two ways to reduce the amounts of actinide by: i) minimization of actinide generation in nuclear power reactors, and ii) destruction of actinides generated in nuclear power reactors. The last option is also frequently referred to as transmutation. The available means are:

i) changes in reactor operation modes,
ii) changes in fuel design,
iii) changes in fuel composition,
iv) changes in the nuclear fuel cycle,
v) introduction of improved (or novel) types of power reactors, and
vi) development and use of accelerator-driven subcritical power reactors.

At present, however, generation and utilization of plutonium is usually maximized for economic reasons. E.g., in a typical swedish BWR more than 30% of the energy comes from burning of the plutonium formed by neutron capture in ^{238}U.
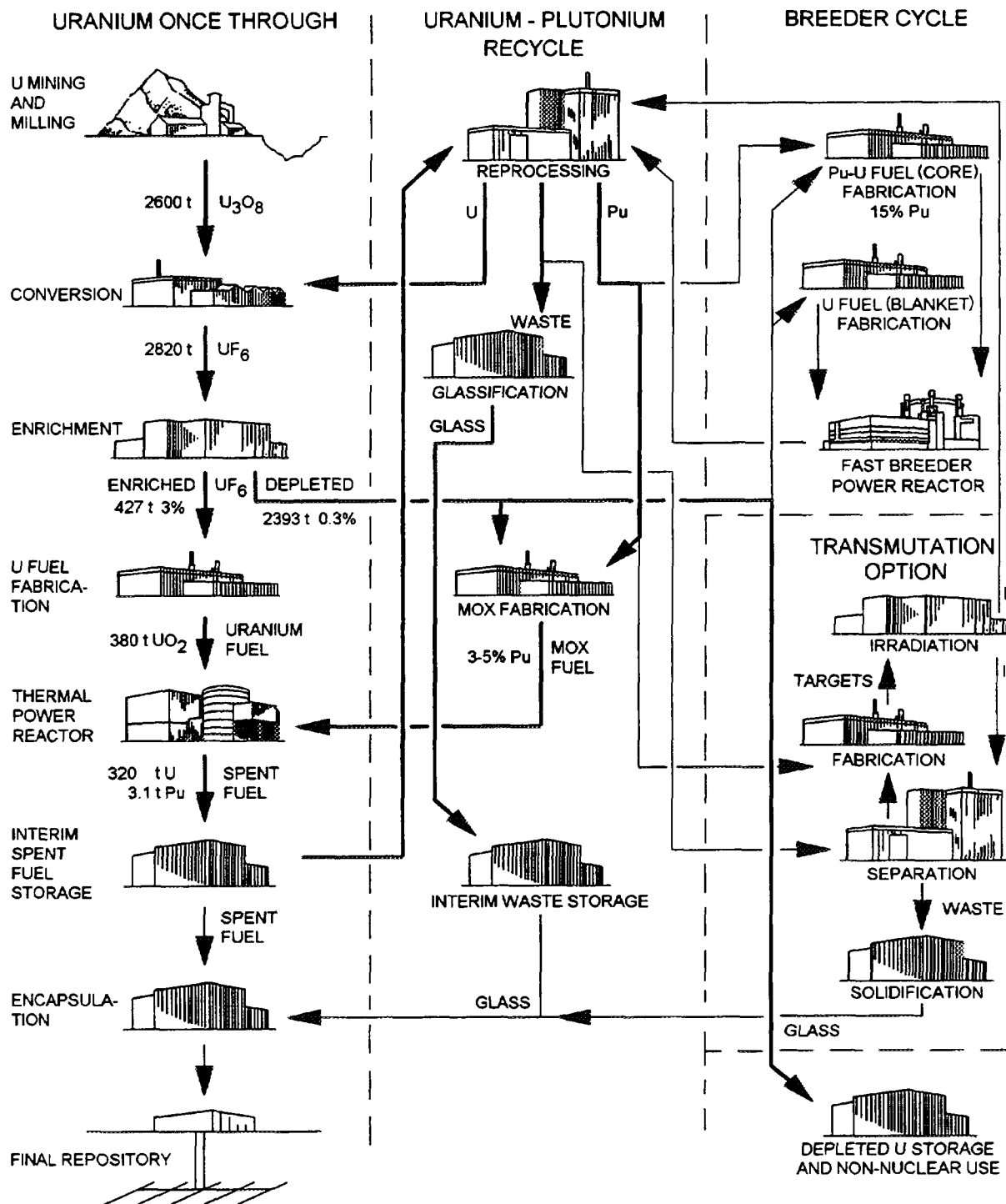
FIG. 1. The nuclear U/Pu fuel cycle with possible variations and extensions.

A trend towards thinner fuel pins in BWR and PWR reactors makes it possible to achieve a higher burn-up by increased conversion of ^{238}U to plutonium, partly compensating the consumption of ^{235}U, and burning of part of the plutonium produced. As plutonium is mainly formed (and burned) in the outer radial zone of a fuel pin, thinner pins yields a relatively higher percentage of plutonium in the spent fuel than thicker pins. However, the accompanying increase in the possible burn-up should in general more than compensate for the higher plutonium concentration in spent fuel. Thus one would expect that thinner fuel pins leads to less spent fuel, but with an increased plutonium content. The combined result could be a decrease in the total amount of plutonium in spent fuel from production of a given amount of electricity.

MOTIVES FOR A REDUCED ACTINIDE GENERATION

Why should actinide generation be reduced? Possible motives could be:

i) A reduced world inventory of plutonium. This could reduce the risk of proliferation. However, with this motive the incentive to reduce inventories of the other actinides should be rather small.

ii) A smaller potential hazard from long-lived nuclides in the final repositories for radioactive waste. In this case the are two different situations; final storage of unreprocessed spent fuel and storage of high level waste from reprocessing. With regard to storage of unreprocessed fuel there is only a very small reduction in actinide content possible. In the case of reprocessing waste, the actinide content depends mainly on the separation process used and its efficiency. However, in order to avoid a large inventory of actinides and corresponding cumulative losses to wastes it is also necessary to achieve a high degree of destruction of actinides during each recycle.

iii) Purely political. These motives could be e.g. to increase public acceptance of nuclear power by removing one of the arguments against its use. In this case we must ask if e.g. subcritical accelerator-driven reactors would be perceived as more acceptable than normal critical reactors, or if the Th/U-based fuel cycle is more acceptable by the general public than a U/Pu-based fuel cycle. An other argument sometimes presented is to reduce the possibility of a future large scale use of nuclear power by forcing a less efficient use of the available uranium resources. However, in this case it must be assured that spent fuel repositories will remain inaccessible for ever.

THE EFFECT OF MINOR CHANGES IN FUEL DESIGN

What can be done with present light water reactors running on the U/Pu cycle? In case of BWRs some difference in actinide generation can be obtained by running the reactors at higher or lower void. It is also possible to vary the fuel pellet diameter in LWRs in order to affect the actinide concentration in spent fuel. The flux depression in the ^{238}U resonance capture region causes most conversion to occur near the pellet outer radius. Autoradiographs and other examination methods confirm that most of the plutonium is found in a thin layer near the pellet surface. Hence, a smaller pellet diameter yields a higher average concentration of plutonium than a larger diameter. This can be expressed as follows;

$$m_{Pu}/m_{fuel} = 4\,k_{Pu}\,a\,(d-a)\,d^{-2}+k_{Pu\text{-}avg}$$

where d is the pellet diameter, a is the zone thickness where most of the Pu is present, k_{Pu} is the excess concentration of Pu in the Pu-rich zone and $k_{Pu\text{-}avg}$ is the average Pu concentration in the pellet. The constants k_{Pu} and $k_{Pu\text{-}avg}$ depend of course on the reactor, operating conditions, burn-up, etc. If $a = 0.5$ mm and $k_{Pu} \gg k_{Pu\text{-}avg}$ are assumed and all other factors remain constant, a change from 10 mm pellets to 8 mm pellets would increase the concentration of Pu in the spent fuel by a factor ~ 1.23. However, a larger diameter most likely results in a fuel with a lower burn-up. This would result in more spent fuel for a given energy production, albeit with a smaller concentration of Pu, which might remove the gain. Careful calculations are needed in order to find an eventual optimum.

TABLE I. Comparison of calculated plutonium and minor actinide amounts $(g/GW_{th}d)$ in spent fuel from reactors operating at 1000 MW_e and a load factor of 0.8 (based on data from [1], [2] and [3]).

	BWR	PWR	LMFBR	THTR
Protactinium	2.6×10^{-6}	1.0×10^{-5}	–	**0.21**
Neptunium	11	14	7.7	**24**
Plutonium	288	334	**3030**	18.6
Americium	3.8	5.1	**28**	0.5
Curium	0.79	**1.3**	0.64	0.24

The LMFBR (U/Pu cycle) and THTR (Th/U cycle) are assumed to operate in breeding mode.

THE EFFECTS OF REACTOR TYPE AND FUEL

What about differences between reactor types and fuels? Table I gives calculated amounts of actinides in spent fuel from four reactor types normalized to 1000 MWe and a load factor of 0.8. As can be seen, the largest concentrations of protactinium and neptunium are expected for a THTR operated in breeding mode on the Th/U cycle, the largest plutonium concentration for a LMFBR operated in breeding mode on the U/Pu cycle, and the largest curium concentration for a PWR operated on enriched uranium fuel.

FUEL CYCLES

Figure 1 illustrates the current and future options for the U/Pu fuel handling. The left part shows the activities involved with the once-through fuel option, the center part shows the activities and options involved in U/Pu recycle, the right upper part indicates the additional activities for a breeder cycle, and the right lower part illustrates the additional operations needed in case a transmutation option for actinides would be established. The Figure also indicates where reprocessing and separation processes have to be used. In this context it is important to understand the possibilities and limitations of these processes. The following facts are important:

i) The fuel/target material after irradiation is the input to the separation process.

ii) The process separates chemical elements regardless of their isotopic composition (isotope separation is possible, but costly, and must be added after the chemical separation).

iii) The primary output of the process is either new fuel/target material in the proper chemical/physical form or raw material to a separate fuel/target fabrication process.

iv) The secondary output from the process is waste for direct disposal or input to a separate waste treatment process.

TABLE II. Some typical losses (as fractions of input and % of input) in separation and fabrication.

	standard PUREX		Reprocessing of spent fuel with modified PUREX process		TPu recovery process added	
Pa	1	100.%	1	100.%	1	100.%
U	0.005	0.5%	0.005	0.5%	0.001	0.1%
Np	1	100.%	0.05	5.%	0.01	1.%
Pu	0.003	0.3%	0.003	0.3%	0.001	0.1%
Am	1	100.%	1	100.%	0.01	1.%
Cm	1	100.%	1	100.%	0.01	1.%

MOX and target fabrication

All actinides	0.0001	0.01%	

TOTAL LOSSES TO WASTE

Reprocessing/separation processes always have some loss of actinides to waste streams. Table II summarizes some typical data for existing reprocessing operations and for future extended actinide recovery operations. The Table also gives some data on typical losses in MOX fabrication and in target fabrication for actinide burning. All values, except for standard PUREX and MOX fabrication, are estimates.

Recycling of actinides can in principle be divided into two cases; co-processing and separate processing. In co-processing the irradiated recycled material is mixed with spent un-recycled fuel and is treated in a common reprocessing plant. By following a unit mass of any actinide through the reprocessing plant, fabrication of targets from recovered material, new irradiation, and back to the reprocessing plant, cycle after cycle, until all material is transmuted after an infinite number of recycles it is possible to compute how much of the original material is lost as waste. This type of recycling is shown schematically in the upper part of Figure 2 for the first cycle. The total fraction of any element, originally present in the spent fuel, which is lost to waste, w, is given by:

$$w = a[\sum_{n=0}^{\infty}(1-a)^n(1-b)^n\{\prod_{m=0}^{n}(1-c_m)\}] + b[\sum_{n=0}^{\infty}(1-a)^{n+1}(1-b)^n\{\prod_{m=0}^{n}(1-c_m)\}]$$

where a, b, and c_m are the fraction of element lost to wastes in reprocessing, in target fabrication, and the fraction burned/transmuted in recycle pass m ($c_0 = 0$), respectively. As can be seen, it is important in this case to keep the losses to waste in reprocessing very low and the fraction transmuted per recycle high. Otherwise a considerable fraction of the initial amount of any recycled element will ultimately become waste.
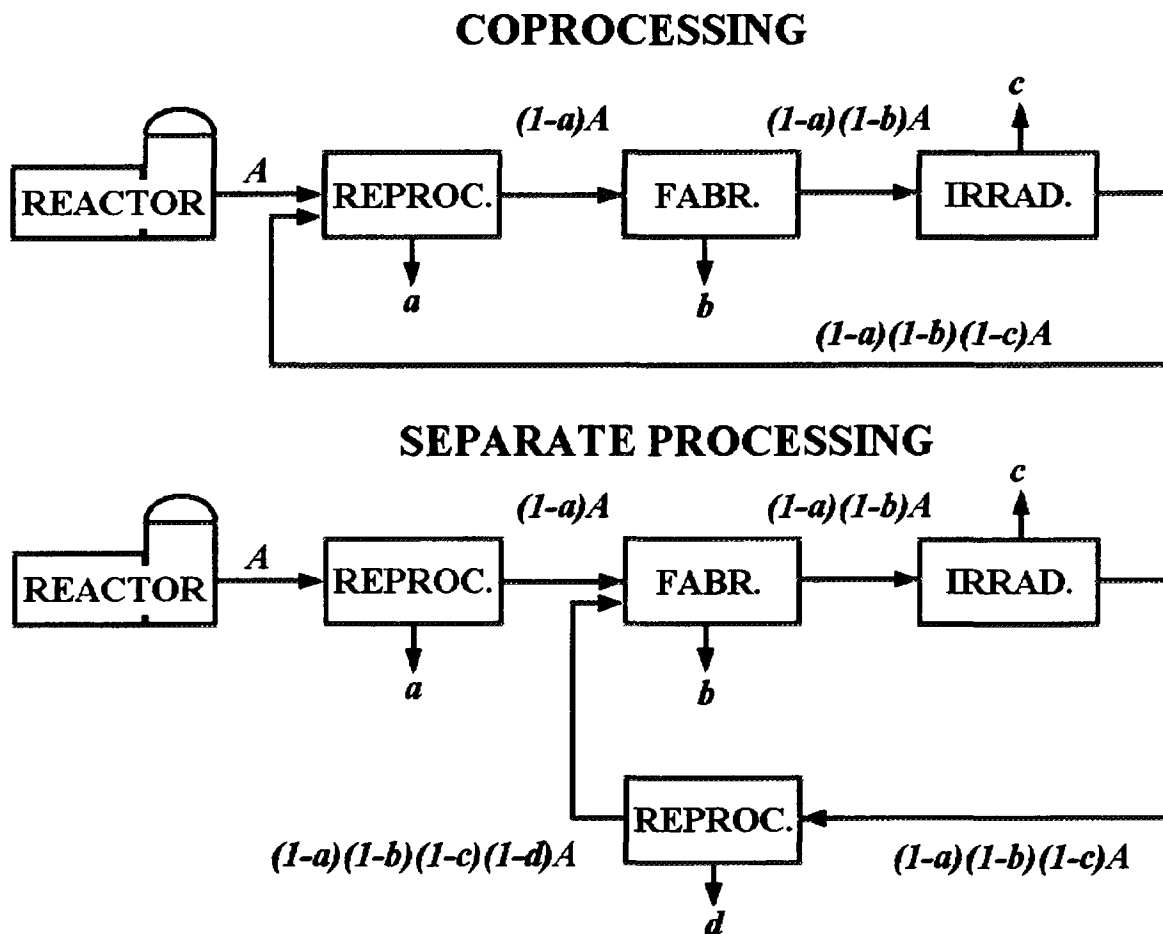
FIG. 2. Recycling options and fractions lost to waste in various operations.

When a separate plant is used to process/separate irradiated recycles material, as indicated in the lower part of Figure 2, the total fraction lost to waste after an infinite number of recycles is given by:

$$w = a + (1-a)\left[b\sum_{n=0}^{\infty}(1-b)^n(1-d)^n\left\{\prod_{m=0}^{n}(1-c_m)\right\} + d\sum_{n=1}^{\infty}(1-b)^n(1-d)^{n-1}\left\{\prod_{m=0}^{n}(1-c_m)\right\}\right]$$

where d is the fraction lost in the separation process. In this case the losses in normal reprocessing only occur once as an important term in the equation. Thus, slightly larger losses can be tolerated than in the case of co-processing. However, also in this case it is necessary to achieve low losses in the separate processing and manufacturing processes as well as a high burn-up in each recycle.

For a simple demonstration we can assume that the same fraction is transmuted in each irradiation, i.e. c_m = constant = c. After simplification with the assumption that a, b, c, and d are all $\ll 1$ we obtain the approximate equations:

$$w \approx (a + b)/(a + b + c) \qquad \text{for co-processing}$$

and

$$w \approx a + (b + d)/(b + c + d) \qquad \text{for separate processing}$$

For a meaningful destruction we must in both cases require that the denominator is dominated by c.

If we consider co-processing using a modified PUREX process with an add-on TPu separation process as an example, Table II gives the following data for neptunium: $a = 0.01$ and $b = 0.0001$. For a total loss to wastes of not more than 5% of all Np we must transmute at least 19.2% of Np in each irradiation.

Consider instead the case of separate processing after a modified PUREX with the same data as above. Furthermore, assume that the separation process only has a 0.5% loss of Np to wastes. In
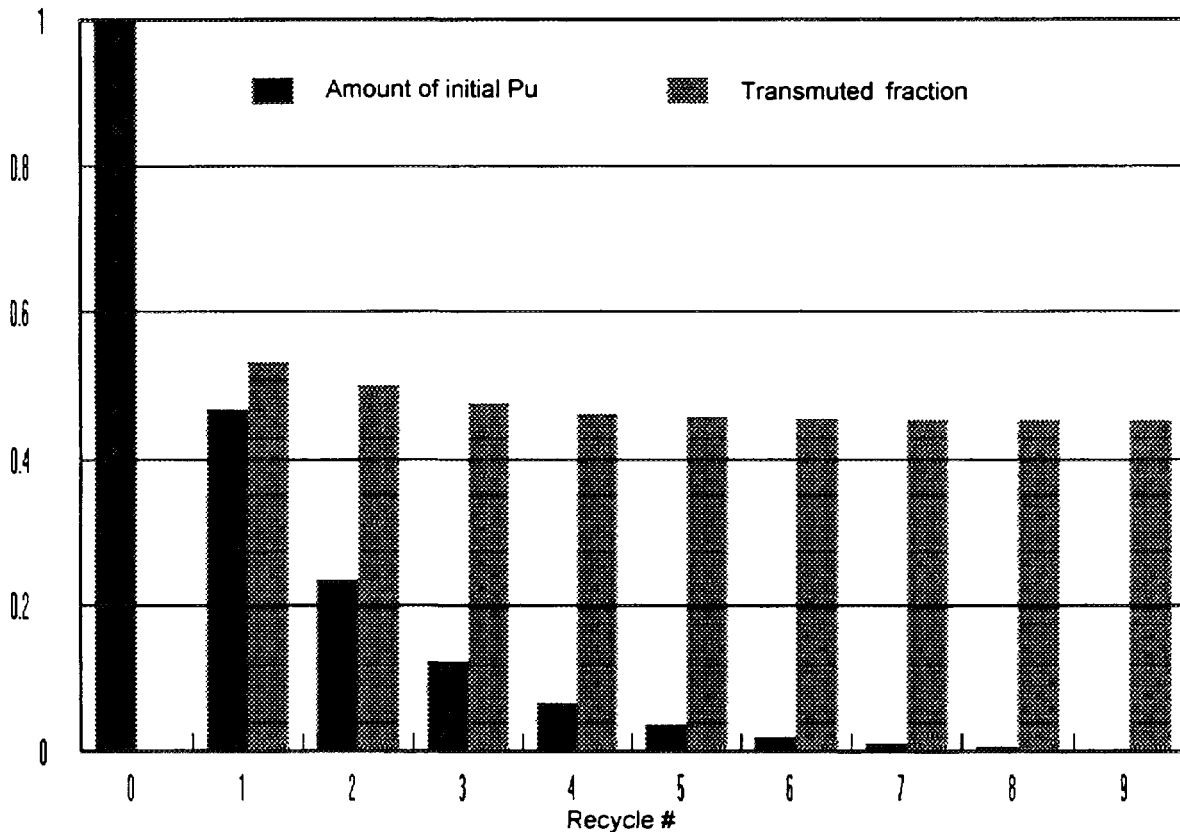
FIG. 3. Calculated transmuted fraction and amount remaining of Pu recycled as MOX at a burn-up of 33 MWd/kg.

this case we would require a transmutation of at least 12.2% to keep a loss of Np to all wastes of 5% or less.

Are such high transmutation fractions achievable? A simple calculation of the transmutation of plutonium when recycled to a light water reactor as MOX with 5% Pu and a burn-up of 33 MWd/kg gave the data shown in Figure 3. As can be seen, the transmuted fraction decreases somewhat with the number of recycles due to a changing isotopic composition of the recycled Pu. However, the single cycle transmuted fraction seems to be large enough to make a nuclear incineration feasible with rather small Pu losses to wastes.

THE TOTAL INVENTORY

If we denote the turn-around time of one recycle for the co-processing alternative by t_c, and the amount of an actinide fed to the process during t_c by A_c we can calculate the total inventory, M_c, of that actinide being circulated at equilibrium from the following equation.

$$M_c = A_c \left(1 + \sum_{n=1}^{\infty} (1-a)^n (1-b)^n \prod_{m=0}^{n-1} (1-c_m)\right)$$

If we now make the simplification that c_m = constant = c and use the data from the previous co-processing example ($a = 0.01$, $b = 0.0001$, and $c = 0.192$) the total circulating Np inventory would be about 5 times the amount of Np entering the recycling process during t_c.

CONCLUSIONS

The following general conclusions might be drawn regarding the available options to reduce actinide generation and actinide inventory:

1   - Countries who intend to store unprocessed spent fuel in a final repository.
-       Only marginal interest in actinide reduction methods which involve reprocessing.
-       Maybe BWRs are marginally better than PWRs with regard to actinide generation.

2   - Countries who accept reprocessing of spent fuel but desire to keep to current reactor types.
-       Only recycling/burning as MOX fuel or special target pins is of interest.

3   - Countries who accept reprocessing of spent fuel and "new" reactor types.
-       The Th/U fuel cycle has some benefits, but probably requires "new" reactor types and certainly requires new reprocessing facilities.
-       FBR technology is also of interest as such reactors can be used in burner mode.

4   - Countries who have political problems with poor acceptance of current light water reactors.
-       The idea of accelerator-driven systems might be more acceptable.

5   - Reduction of stock-piles of weapons-grade material.
-       High enriched uranium fuel could be used, which would lead to minimal production of transuranics. However, this may pose another safeguard problem.
-       ^{239}Pu could be used as MOX fuel in available power reactors or in special Pu burners, e.g. ATR or FBR.
-       Accelerator-driven systems may also have some benefits as Pu burners.

6   - Most important
-       To develop reprocessing and separation technology towards smaller losses to wastes.

- To be wary of new processes not proven on an industrial scale (remember the Mid-West Fuel Recovery Plant).
- To develop target materials for high burn-up and suitable for the separation process.

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THE FEASIBILITY OF MA TRANSMUTATION IN CEFR

ZHANG YUSHAN, YI XIAOYI, SU LIXIN
China Institute of Atomic Energy,
Beijing, China

Abstract

The feasibility of MA transmutation in CEFR(China Experimental Fast Reactor) is described. The nuclear characteristics of reference core and those of MA-loaded core are compared, the MA-transmutation amount is presented. Although the amount of MA transmutation in CEFR is limited, CEFR still has a significant role in MA fuel irradiation tests and MA transmutation technique studies.

1. INTRODUCTION

At the present time, the Chinese nuclear electricity generation rate is 15 TWh/a (2TWh/a for Qinshan and 13TWh/a for Daya Bay). It may be expected to increase slightly and to reach a new rate later on.This nuclear electricity is produced by PWR type reactors using uranium oxide fueled assemblies. With such type of fuel,the MA(Minor Actinides) Production rate is about 53 kg/a and long-lived fission product Tc is 55kg/a.In order to reduce the long term radiological hazard associated with the disposal of high-level radioactive waste, the problem of MA transmutation has been placed on the order of the day in China.

Various methods of MA transmutation have been proposed during the last 20 years. Among the possible schemes, one of the practical methods is to recycle MA in fast reactors [1] so as to incinerate them.

CEFR (China Experimental Fast Reactor)[2] is a 65 MWt (25 MWe) sodium cooled experimental fast reactor designed by CIAE (China Institute of Atomic Energy). In order to develop MA transmutation technique in China, It is necessary to consider the role of CEFR in MA transmutation. The first, some irradiation tests can be performed on CEFR .For example, irradiation of MA contained samples, irradiation of special fuel pins with various contents of MA. These tests can be used to study the properties of various MA-contained fuel samples and fuel pins for the examinations of some theoretical works. The second, CEFR can be used for MA transmutation. Although the amount of MA transmuted is limited, it is still significant in scientific sense and calculational demonstration. In the following sections, the feasibility of MA transmutation in CEFR is described. Parameter survey calculations are performed for CEFR MA-loaded core so as to investigate the basic nuclear characteristics of it. The comparison of nuclear characteristics between reference core and MA-loaded core is presented.The amount of MA transmutation in CEFR is presented and some comments are made.

2. CALCULATIONAL METHOD

The basic constant set used in calculations is 46 groups LIB-IV[3] modified library. This 46 groups constant set is condensed to 6 groups constant set by IDX code [4]. Various nuclear parameters of core were calculated by two dimensional diffusion and burnup code 2DB[5].

3. CEFR REFERENCE CORE [2]

The CEFR original designed core is defined as CEFR reference core. The main parameters of CEFR reference core are shown in Table I. The core layout is shown in FIG.1.

TABLE I DESIGN PARAMETERS OF CEFR REFERENCE CORE

Parameters	Data
Thermal power (MWt)	65
Gross electric output (WMe)	25
Operation length (EFPD)	71
Number of fuel batch	3
Fuel material (First Core)	UO_2
Enrichment of fuel	60%
Fuel pin diameter (cm)	0.6
Number of subassemblies	
Fuel subassemblies	82
Control Rods	8
Core height (cm)	45
Equivalent core diameter (cm)	58.5
Neutron absorber	B_4C
B-10 enrichment in B_4C	91%(at)
Assembly lattice pitch (cm)	5.85
Volume fraction	
Fuel	0.381
Sodium	0.207
Structure	0.397
Gap	0.015

◯	Fuel Subassembly	82
⊙	Blancket Subassembly	163
⊕	Reflector Subassembly	126
◎	Spent fuel storage position	54
◉	Control rods	6
◉	Safety rods	2
◯	Sheilding rods	216
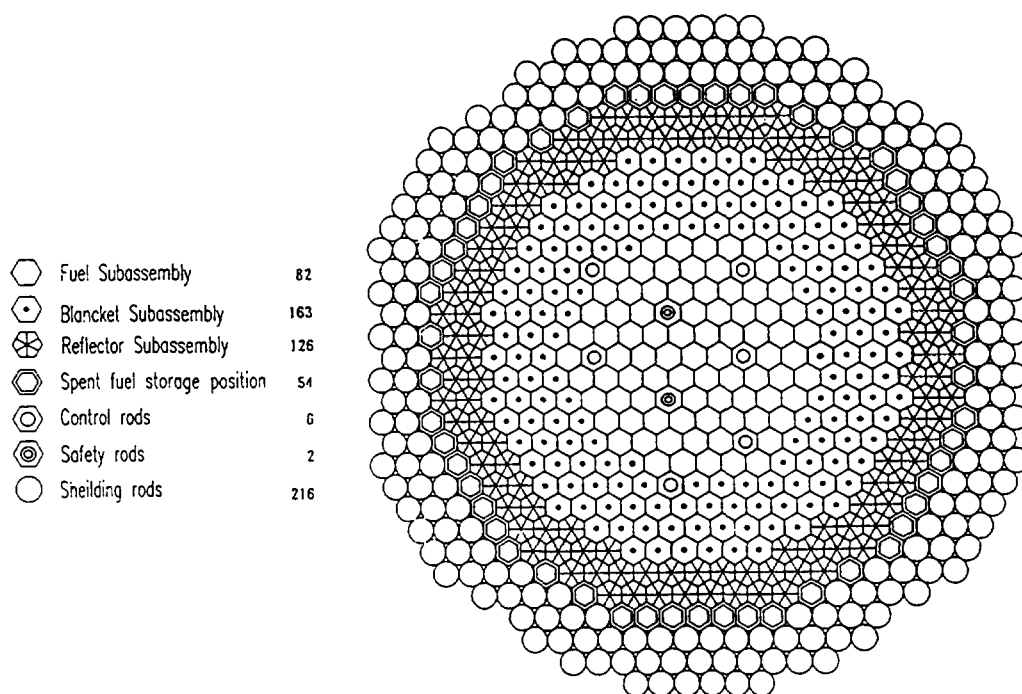
FIG. 1. Core layout of CEFR.

4. CEFR MA LOADED CORE

The MA fuel is assumed to come from PWR spent fuel which has been cooled for five years before reprocessing. The data of composition of MA fuel are cited from reference [6] (see Table II).

TABLE II. COMPOSITION OF MA FUEL * [6]

Nuclide	Weight Fraction(%)
Np-237	49.1
Am-241	30.0
Am-242m	0.08
Am-243	15.5
Cm-243	0.05
Cm-244	5.00
Cm-245	0.26

* Discharged from PWR (35GWd/t) and
Cooled for 5 Years Before Reprocessing

The MA is homogeneously dispersed throughout the entire core. The ratio of MA amount loaded in fuel is 50% and the enrichment of U-235 is 90%. The nuclear characteristics of CEFR MA-loaded core are shown in Table III and the reference core parameters are also listed for comparison.

TABLE III. THE CHARACTERISTICS OF CEFR MA-LOADED CORE

Item	CEFR Reference Core	CEFR MA-Loaded Core
U-235 Enrichment	60%(wt)	90%(wt)
Loading ratio of MA	0.	50%(wt)
Amount of MA loaded(kg)	0.	200
Number of fuel batch	3	3
Cycle length (days)	71	102
Burnup reactivity loss (% $\Delta k/k$)	0.71	0.49
Radial power factor	1.41	1.46
Axial power factor	1.17	1.17
Control rod worth (% $\Delta k/k$)	10.0	7.4
Doppler constant($Td\rho/dT$) ($10^{-4}\Delta k/k$)	-3.4	-0.8
Sodium void effect (% $\Delta k/k$)	-6.6	-2.6
β_{eff}	7.08×10^{-3}	5.39×10^{-3}
MA transmutation ratio (%/cycle)	--	1.57
Net amount of MA transmuted per year (kg/a)	-3×10^{-2}	9.4

As shown in Table III, the reactivity worth of all control rods in MA-loaded core is smaller than that of reference core This is caused mainly by the decrease of relative importance per neutron in MA-loaded core due to the increase of production rate of multiplication neutrons Although the control rod worth decreases, the total worth is still sufficient for reactor control.

The decrease of β_{eff} in MA-loaded core is due to the small value of delayed neutron yield of MA The decrease of sodium void effect in MA-loaded core is caused by neutron spectrum hardening which results in increase of fission cross-sections of MA The Doppler effect of MA-loaded core is smaller than that of reference core It is caused by the decrease of U-238 composition The burnup reactivity loss per cycle for MA-loaded core is smaller than that of reference core due to the production of Pu-238 from Np-237 It is advantageous to the extension of reactor operation period and the increase of fuel burnup

Although the parameters related to reactor safety of MA-loaded core are not as good as that of reference core, they are still acceptable from view point of reactor safety

5. CONCLUSION

1   It can be seen from Table III, the MA-transmutation rate in MA-loaded core is 9 4 kg/a It is nearly equal to the MA production rate in QINSHAN PWR nuclear plant (0 3GWe) Therefore CEFR MA-loaded core has the capability to transmute the long-living radiotoxic nuclides produced in QINSHAN nuclear power plant Although the amount of MA-transmutation in CEFR MA-loaded core is limited, it is still significant in China because this means that the problem of MA transmutation has been paid much attention at the start of PWR nuclear power development in China

2   CEFR also has its significant role in MA fuel irradiation tests and MA transmutation technique studies, for instance, irradiation test of MA contained semples, irradiation test of special fuel pins with various contents of MA These tests can be used to study the properties of various MA-contained fuel samples and fuel pins and to examinate some data and theoretical works

3   CEFR is an experimental fast reactor, the thermal power is limited (65MWt) Even though it has hard spectrum which is favorable for MA transmutation, the amount of MA-transmutation is still limited At present time, following the development of PWR nuclear electricity in China, the problem to alleviate the burden of the high level radioactive waste (HLW) depository has been placed on the order of the day An effective long term R&D program supported by authority for developing MA transmutation schemes is very necessary

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LIQUID FUEL CONCEPT BENEFITS

M. HRON
Nuclear Research Institute,
Řež, Czech Republic

Abstract

There are principle drawbacks of any kind of solid nuclear fuel listed and analyzed in the first part of the paper. One of the primary results of the analyses performed shows that the solid fuel concept, which was to 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, for public namely, way). On the background of this, the liquid fuel concept and its benefits are introduced and briefly described in the first part of the paper, too.

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 and currently having been under development in the Los Alamos National Laboratory [1], will be studied both theoretically and experimentally. There is a preliminary design concept of an experimental assembly LA-0 briefly introduced in the paper 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 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.

1. INTRODUCTION

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 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 U235 in natural uranium. Therefore, solid phase metallic uranium with highest as possible density was used and in the form of blocks with a specific definite size arranged in a heterogeneous lattice in a solid (graphite) or liquid (heavy water) moderator with a certain pitch determined by optimal neutronic conditions. This arrangement has remained nearly exclusive even in latter systems with fuels enriched by U235 content up to much higher levels. 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 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 are adopted. They all begin to form a magic circle of convention in which the short term and finally even long term operational behaviour of nuclear, namely power, reactor 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 more and a further operation of the reactor under original conditions is impossible. There is an unavoidable principle change in the operation and structure of the reactor which means an outage and exchange of at least a part of the 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 principally 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 violation 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 utilizes 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 so 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. PRINCIPLES OF DIFFERENT TRANSMUTATION SYSTEMS

The first candidate for this purpose is a nowadays called classical nuclear reactor itself. Its ability, to certain degree, to burn fission products is natural and well known. The first limitation and even barrier we certainly approach in this attempt is the neutron economy of conventional reactor systems. The next attempt could be an actinide fuelled (critical) reactor. Then another problems arise. First of all, the problem of control of such a system. It is well known that the fissile isotopes of Neptunium, Americium and Curium have a considerably smaller fraction of delayed neutron emitters in comparison to the common fuels based on Uranium isotopes and the essential role which the fraction of delayed neutrons is playing in the control of nuclear reactor in critical state is not necessary to recall here. Beside this, there are other problems with a small Doppler effect and a positive sodium void coefficient arising in this case as well.

To overcome these problems, there have been various concepts of subcritical systems driven by a suitable neutron source proposed in the recent past, aiming at the transmutation of actinides and long lived fission products. The suitable neutron source, it means the source with a sufficient yield of neutrons allowing to avoid the principle barrier of neutron economy, appeared in the concept of spallation reaction induced by highly accelerated charged particles, e.g. protons:

1) The idea of a direct exploitation of the spallation process to transmute actinides and fission products had to be given up because of a barrier of necessary particle currents which are much larger than the most optimistic theoretical accelerator designs show around 300 Ma. It had been reported [1,2] that the destruction rate of the largest possible proton accelerator would correspond only to a fraction of the amount of fission products

generated by one thermal reactor of 1000 MW$_e$ in the same period of time.

2) The idea of a direct use of spallation neutrons was tested. The fission products were supposed to be placed around a proton target to use only the spallation neutrons as they are generated in the target. In dependence on the characteristics of the material which is to be transmuted, either the fast neutrons would be used as they are emitted from the target or they would be slowed down by a moderator to energy levels with higher transmutation cross sections like the resonance or thermal energy ranges. The following simple estimate shows the substantial disadvantage of such a system. Assuming that it is possible to make all the spallation neutrons available for the transmutation process:

$$E_{fp} = q_{fp} \frac{P_b}{\eta_{sp}} \frac{1}{\eta_b \eta_T} \ [MW] \tag{1}$$

where E_{fp} is the amount of energy which is necessary to transmute the fraction

q_{fp} of radionuclides per fission process in a nuclear energy system,

P_b is proton energy,

η_{sp} is the number of neutrons generated by one proton,

η_b is the efficiency of converting electricity into proton beam energy (~0,5) and

η_T is the efficiency of converting thermal energy into electricity (~0.33).

For example, in the case of a 1,5 GeV proton beam emitting 50 neutrons per spallation in a lead target, the transmutation of ^{99}Tc, ^{129}I, ^{135}Cs, ^{90}Sr, ^{85}Kr and ^{93}Zr (constituting 28% of all fission products) would require 51,3 MeV to transmute the fission product fraction of one fission process. This is ~26% of the total power production of the energy system under consideration and the real percentage of energy required will be even higher due to the very optimistic assumptions made in this estimate. Together with the cost for reprocessing it would make this type of accelerator transmutation prohibitively expensive, at least in a commercial nuclear energy system.

3) The idea of a neutron source driven subcritical reactor-type assembly to improve neutron economy remained. Technically, this can be realized by surrounding a proton target region by fissionable material in a cooling system. For removing of the high specific heat released in the target, there is a liquid metal alloy mostly used. However, we should mention that the specific heat production per neutron is considerably lower than in a fission process (30 MeV against 80 MeV).

Let us quantify the power production P_{fi} of a subcritical assembly fed by spallation neutrons:

$$P_{fi} = \eta_{sp} \frac{a-k}{\mu(1-k)} \frac{i}{C} E_f \qquad (2)$$

where k is the multiplication factor, a is the importance of the target position (usually $a>1$ for a central target position), μ is the mean number of neutrons in a fission process, E_f is the power release per fission ($=3,2.10^{-11}Ws$), η_{sp} is a neutron yield from one proton, i is proton current, C is proton charge ($=1,6.10^{-19}As$). The dependence of the power production of an accelerator driven facility upon its subcriticality ($1-k$) assuming a proton beam of 1 GeV and 1 mA bombarding a Pb-Bi target releasing neutrons per spallation with an importance $a=1$ illustrates that near criticality an 1 mA current already generates a relatively high fission power and for $k=0,97$ more than 100 MW can be achieved.

In this case, the additional neutrons from the subcritical system as well as its fission power, which can be also transformed into electricity, can be exploited to run the transmutation process. The energy required to transmute a fraction q_{fp} of fission products in such a system can be expressed as follows:

$$E_{fp} = \frac{\eta_{sp} \dfrac{k}{\mu(1-k)} E_f - \dfrac{P_b}{\eta_b \eta_T}}{\eta_{sp} [(1-k/\mu) \eta_{fp} + \dfrac{k}{1-k} ((1-k/\mu) \eta_{fp} - \dfrac{q_{fp}}{\mu})]} \qquad (3)$$

where $\quad \eta_{fp} = \dfrac{\Sigma_a(FP)}{\Sigma(FP+fuel+constr.mats)}$

The positive sign of E_{fp} means that there is even a surplus of energy, while the negative sign indicates a need of energy which should be added to the system from outside.

In spite of these very promising features, the fact that the amounts of long lived fission products generated by currently operated thermal reactors are relatively very high and corresponding huge amounts of neutrons are required to their transmutation results to a certain stress in the overall neutron economy of those designed systems and requires a relatively high power of the accelerator for spallation neutron source. One of the possible improvements in this situation is a higher degree of multiplication of spallation neutrons from the source by the subcritical reactor. In the deeply subcritical transmuter with $k_{eff}=0.7$, the proton beam power of 300 MW is required to transmute ~17% of LLFP, such as ^{99}Tc, ^{129}I, ^{85}Kr and ^{93}Zr, created by a 1 GW_e LWR. When the spallation source neutrons are multiplied by a transmuter which is closer to criticality (with e.g. $k_{eff}=0.99$) and

when the ratio n_f of neutron capture by LLFP to the total is 20%, the proton power becomes 4 MW.

There has been a series of deep analyses and evaluations of various transmutation systems feasibility performed in the recent of few last years. In spite of the great variety of different concepts, most of them reached a consensus on the principal structure of a system being divided into two basic technological parts:

a) The source unit. (Mostly based upon neutrons yielded from either direct or spallation reactions induced by accelerated charged particles in nuclei of some heavy elements. There is , in general, an annulus of some high - Z material, such as lead, for further multiplication of the source through (n,xn) and (p,xn) reactions employed here or they may be multiplied by a fissile target core - so called active target.)

b) The subcritical reactor/blanket unit which contains a moderated heterogeneous lattice, using a flowing liquid which contains actinides. There may be separate regions in the blanket where long-lived fission products (LLFP) are transmuted. The structures of the heterogeneous core, which were competitive in the previous stages, were either a heavy water moderated aqueous slurry system or a graphite moderated molten salt system. The former has been connected with a demanding continuous chemical reprocessing which became its drawback. The latter can utilized a mechanical (e.g. centrifuge) separation of individual components of the fluid fuel and has also a higher thermal efficiency. Therefore, we will concentrate in our further analysis and a development of a design concept on such a type of systems.

3. LIQUID FUEL CONCEPT FOR NEUTRON SOURCE-DRIVEN TRANSMUTATION TECHNOLOGY (NSDTT)

3.1. The Los Alamos Molten Fluoride Salt Fuel for Accelerator-Driven Transmutation Technology (ADTT)

The LANL concept of an accelerator-driven subcritical blanket for a nuclear incineration of nuclear waste has been known for 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 of an approvement of 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°C and 650°C, respectively. The molten salt flows over the outside of a close-packed set of high-purity graphite blocks. One of the possible arrangements is graphite
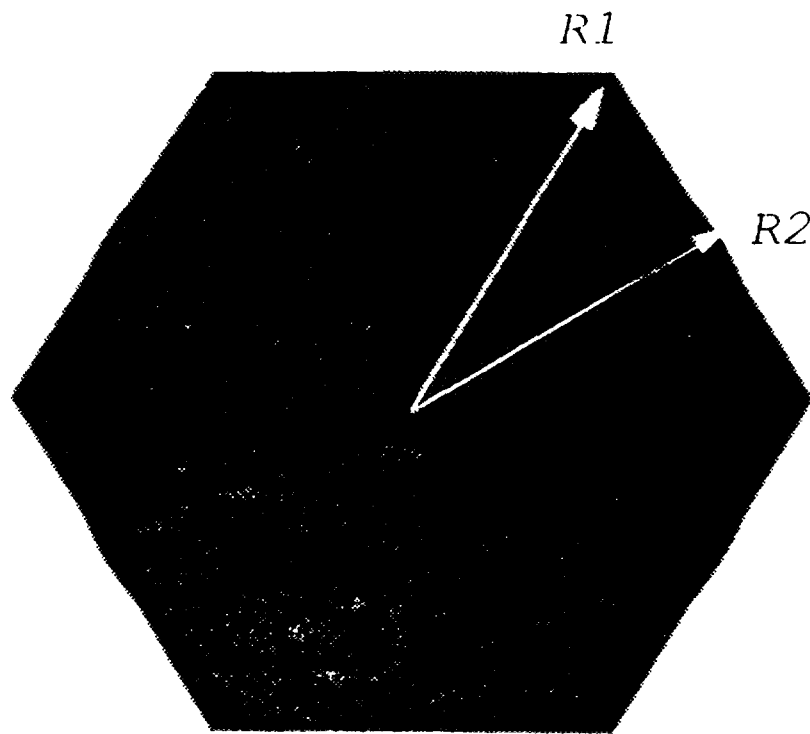
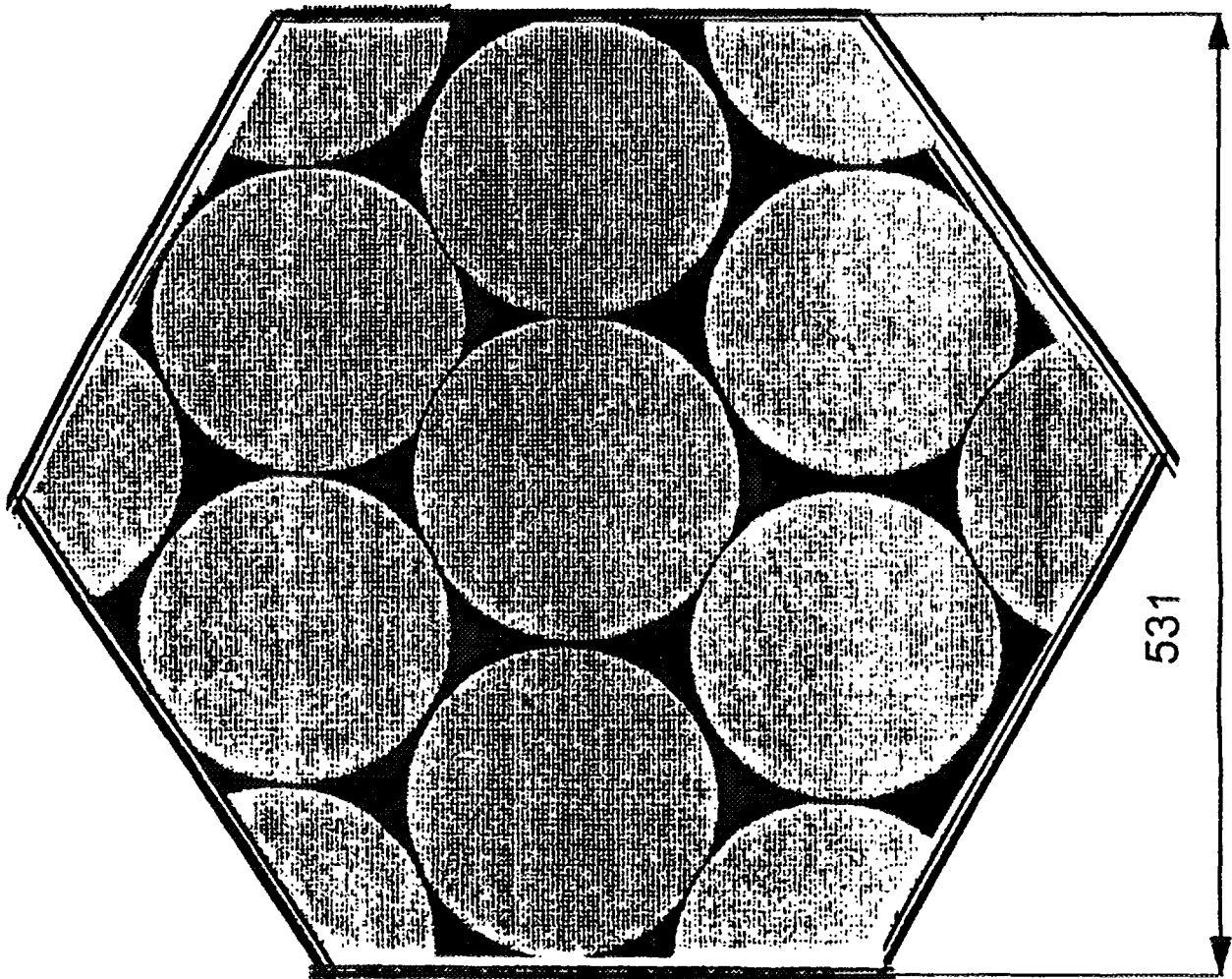FIG. 1. Experimental Assembly LA-O Elementary Cell.

FIG. 2. Experimental Assembly LA-O Elementary Module.

50

blocks in the form of cylindrical rods arranged in a regular triangular lattice. There were simple elementary analyses performed which has lead to a preliminary accepted set of optimal values of the graphite rod radius R1=8.66 cm and the elementary cell hexagon outer circle radius R2=10.0 cm (Fig. 1). There has been an experimental research system called LA-0 preconceptually designed by the author in LANL [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 behaviour. For the very first stage, the following scheme can be applied which will allow the first results to be reached very cheaply and relatively soon. There can be an elementary, however, a sufficiently representative sample of the investigated reactor blanket lattice (an elementary module - Fig.2) 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 modelling 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.

3.2. 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 the usual reactor control organs, negative sources like e.g. absorbers or decreasing of the dimensions of the system, etc.

The high level parameter proton accelerator with all its disadvantages (like e.g. the length ~ 1 km, the investments ~ U$ 1 billion, etc.) having been applied in the Los Alamos
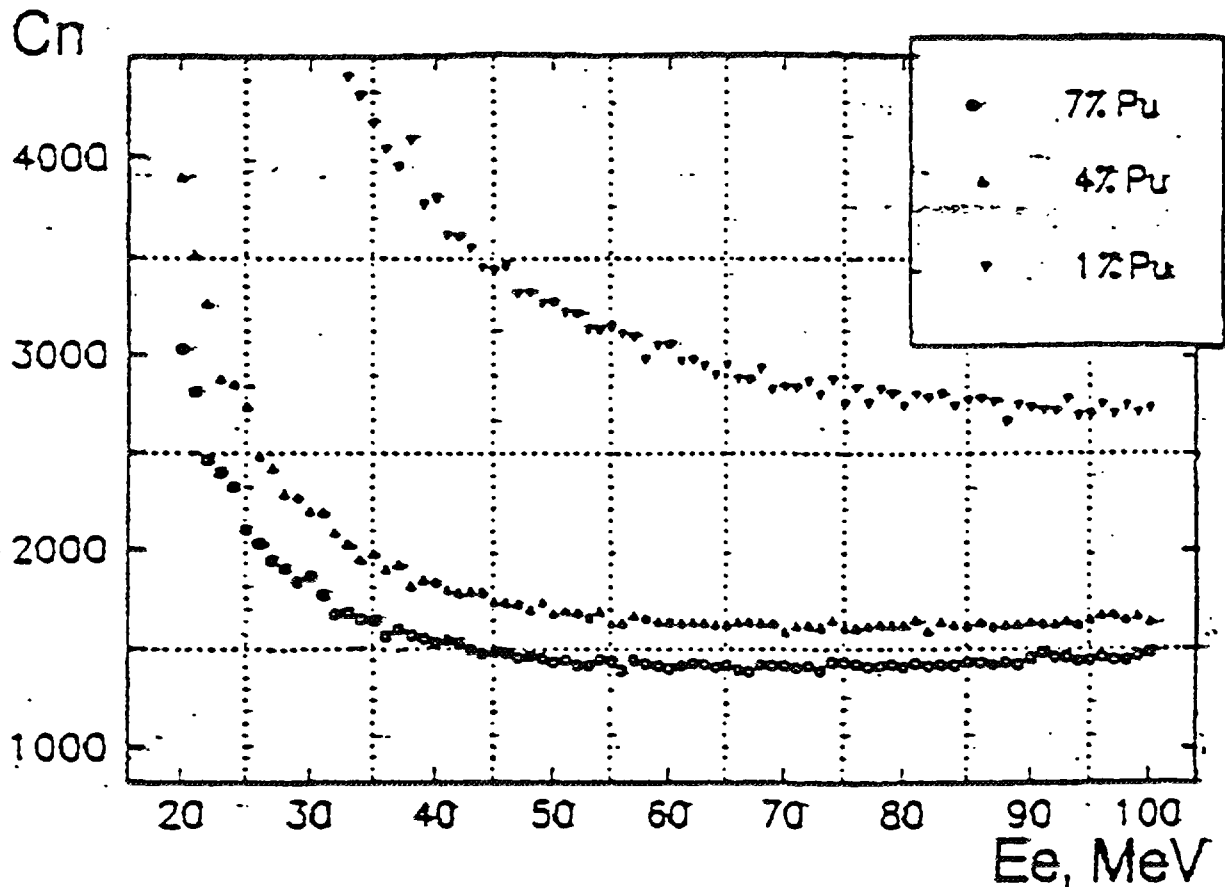
FIG. 3. Electron beam energy necessary for one photonuclear neutron generation.

concept is not necessary   in the system and a low level parameter accelerator can be employed. There has been
a very efficient concept proposed by Russian scientists [4,5] recently utilizing an electron accelerator. The electron beam is supposed to enter directly the molten salt environment in a homogeneous core where it generates photonuclear neutrons by
e - n - τ scheme. The Fig. 3 shows a calculated photonuclear neutron energetic cost in molten salt with equal numbers of Li, Be and F nuclei for compositions with 1, 4 and 7 mol.% of actinides. The energetic cost of a photonuclear neutron makes Cn ≈ 1300 ÷ 1400 MeV that is 25 times higher than that of a electronuclear neutron generated by a proton. However, the decision on the acceptability of a neutron source may not be taken only on the basis of the Cn value.

There may be a scheme for a significant and simultaneously safe neutron multiplication device   that would allow to apply electron accelerators effectively. One very convenient scheme of such a type has again been introduced in [4,5]. The main idea of this concept is a use of the principle of cascade neutron multiplication on subcritical cores known for a long time. The subcritical MSR is analogous to fast-thermal critical coupled

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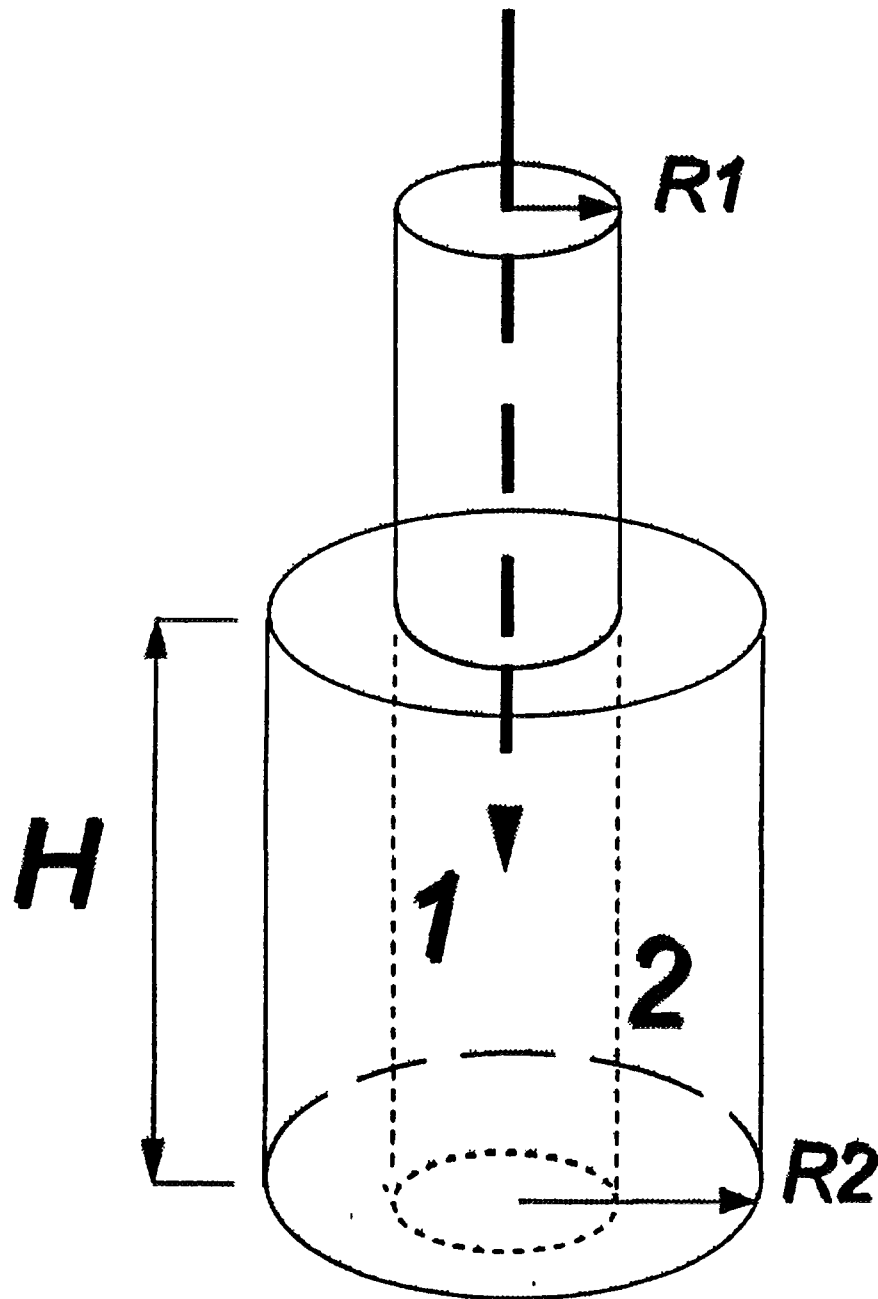
FIG. 4. Principal scheme of a cascade MSR.

reactors [6,7]. The theory of coupled reactors was developed and experimental results for such systems were described. Each reactor in a fast-thermal critical system is subcritical. The system cannot be critical only with fast neutrons. There was a good agreement between theory and experiment obtained. It shows that no unexpected and unknown phenomena occur in similar systems. There is a sufficient number of experimental results available which were performed on fast-thermal critical assemblies in USA (ZPR-III, ZPR-VI), Russia (SPECTR), France (ERMINE-2), Switzerland (PROTEUS) and Belarus (BTS).

The design introduced in [4,5] is a subcritical MSR of 1250 MW$_t$ which consists of two cylindrical cores (Fig. 4). The first core is a fast system with a fuel salt composition of 53NaF + 41ZrF$_4$+ + 6XF$_4$ which is separated by a cylindrical wall from the second pool-type thermal core with fuel salt composition of 69LiF + 28BeF$_2$ + 3XF$_3$ and with an external graphite reflector compatible with fuel salt at high temperatures. The principle scheme of the system's function is as follows:

Fuel salt 1 enters the core 1 bottom through a pipe and due to heating elevates in a 12-m raiser with the 30-m diameter, in which the external source channel is located, and then enters the separate part of the intermediate heat exchanger 1HX.

Fuel salt 2 enters the core 2 top, comes down in a peripheral annulus, elevates in a 12-m raiser with a 1.2-m outer diameter and then enters 1HX.

There is a secondary coolant of the composition of 92NaBF$_4$+ + 8NaF flowing through 1HX pipes. To avoid a fuel salt leak the reactor is placed into a 10-cm-thick double wall vessel. The gap between the inner and outer walls is filled with nitrogen. There is a gas volume above the fuel salt free surface. The pipelines are made of hastelloy-N - heat/proof material containing 15 - 18% Mo, 6 - 8% Cr and additionally alloyed with 1% of nitrogen to prevent interaction tellurium.

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LONG TERM POTENTIAL RISK DUE TO THORIUM MINING

J.P. SCHAPIRA, S. MENARD
Institut de Physique Nucléaire,
Orsay, France

Abstract

As in the case of uranium, thorium extraction produces a certain amount of radioactive wastes, the so called mining residues, in which natural long-lived natural decay products are mobilized. Monazites, the most likely ores to be exploited first, in case of an extensive use of thorium as nuclear fuel, contain a certain fraction of uranium as well. One has studied potential long term radiological impact of the residues originated from two typical monazites found in India and Italy, respectively poor and rich in uranium. In the case of the Italian monazite, uranium is supposed to be extracted at a level of 95%. For such monazites, the long term impact is completly governed by the residual uranium (presence of long-lived ^{238}U descendants, ^{230}Th (80 000 years) and/or ^{226}Ra (1600 years)). However, for the two extreme cases studied here, the potential risks remain much smaller (by a factor lying between 20 to 50) than in the usual case of uranium mining. The risk due to radon is also correspondly reduced by the same factors. These significant differences between uranium and thorium mining have to be considered in view of some estimated long term real radiological impacts due to uranium residues, which could reach a value of the order of 1 mSv/year, the dose limit recommanded for the public by the recent ICRP-60.

There is, since the beginning of the 90, a renew of interest for thorium as a waste management option, specially in relation with accelerator driven systems. The present study is part of a general effort to evaluate the radiological impact of thorium cycles, both in the short and long terms. It focuses here on the long term potential radiological risk related to thorium mining. The purpose is to compare it with uranium extraction, considering realistic thorium ores.

1. Brief review of thorium mineral composition

Thorium is widely spread around the world at very low concentrations (around 10 ppm in the earth crust) and is 3 times more abundant than uranium. The most important ore deposits are located in Brazil, Turkey, India and United States (see tab. 1). There are basically 6 minerals containing thorium at a significant concentration level (see tab. 2), among which thorite, thorogummite and thorianite could eventually present an economical interest. Beside these, most of the thorium is found in monazite, which is basically a cerium phosphate, containing thorium and uranium oxide in various proportions, as can be seen in tab. 3, which lists four typical monazites.

Tab. 1 - Estimated world thorium ressources in 1992 (market economy countries) in 10^3 tons, ref. [1]

Countries	RRA	RSE	Total	%
Europe				
Groenland	54	32	86	
Norway	132	132	264	6.4
Turkey	380	500	880	21.4
Total	566	724	1290	**31.4**
America				
Brasil	606	700	1306	31.8
Canada	45	128	173	
United States	137	295	432	10.5
Total	790	1125	1915	**46.6**
Africa				
Egypt	15	280	295	7.2
Niger	?	?	29	
South Africa	18	?	115	
Total	36	309	479	**11.7**
Asia				
India	319		391	7.8
Total	343	30	403	**9.8**
World Total	**1754**	**2188**	**4106**	**100**

RRA: resources reasonably achievable

RSE: ressources supplementary estimated

56

Tab. 2 - Minerals containing thorium as major constituent, ref. [2]

Mineral	Composition	Th concentration (% in mass)
Cheralite	(Th, Ca, Ce)(PO$_4$,SiO$_4$)	29.5% to 31.5%
Huttonite	ThSiO$_4$	81.5%
Pilbarite	ThO$_2$UO$_3$PbO 2SiO$_2$, 4H$_2$O	31%, variable
Thorianite	ThO$_2$	38.5 % to 93%
Thorite	Th(SiO$_4$)x	7.83% to 58.9%
Thorogummite	Th(SiO$_4$)$_{1-x}$(OH)$_{4x}$	24% à 58% and more

Tab. 3 - Composition of certain monazites (% in mass) , refs. [2, 3]

Country	UO$_2$	ThO$_2$	Lanthanides oxides	Other oxides	P$_2$O$_5$
Italy	15.64	11.34	35.24	6.76	31.02
Sri Lanka	0.10	14.32	53.51	5.03	26.84
California	6.95	4.22	?	?	?
India	0.29 [a]	9.80	?	?	?

a) As U$_3$O$_8$

2. Radioactive wastes generated from thorium mining

It is most likely that monazite would be the major ore deposit exploited, if thorium use in nuclear reactors was to develop. The isotopic composition, and therefore the radiological impact of the wastes generated by this ore exploitation will mainly depend on wether or not uranium is extracted as well. This depends chiefly on the uranium market situation and obviously on the uranium concentration. As an example, taken from tab. 3, it is most likely that uranium will be left with the wastes in the case of Indian monazites, whereas it might well be exploited together with thorium in the case of Italian monazites. We will study these two extreme situations.

Thorium mining wastes will contain the following isotopic composition :

a) The unextracted thorium with its daughters belonging to the 4n natural radioactive chain. We have taken an extraction efficiency of 95 % throughout this study [4].

b) The daughters of the extracted thorium, except ^{228}Th.

c) If uranium is not extracted, the totality of uranium with its daughters, except ^{221}Th, ^{227}Th (^{235}U daughters) and ^{234}Th, ^{230}Th (^{238}U daughters).

d) If uranium is extracted, the uncextracted part of it (5 %), with all the daughters of uranium, except again the four Th isotopes mentionned in c).

3. Radon exhalation

One of the difference between the residues of thorium and uranium extraction, is the absence of radon exhalation in the first case. This is due to the fact that the radon isotope ^{220}Rn, daughter of ^{232}Th, has a short period of 55.6 sec, whereas ^{222}Rn, daughter of ^{238}U, has a much longer period of 3.82 days. Therefore vitueally all the ^{220}Rn descendants are kept in situ and contribute to the residues radiotoxicity. On the contrary, only a certain amount is left within the residues, in the case of uranium, the other part contributing to the so called radon risk, due to the possibility of inhalating radon directly, or its descendants fixed on dusts or aerosols

4. Radiotoxicity calculations

In order to compare uranium and thorium mining, we have considered the radiotoxicity by ingestion as a measure of the potential risk (on the long term, the main route would probably residues transport by water). We have introduced the recent ICRP-68 dose factors [5] by ingestion in an evolution code, which solves the well known Bateman's equations, developped at IPN-Orsay [6]. By this, radiotoxicity is calculated at any time in the future, after mining has taken place

If a pure thorium ore would exist, the residues radiotoxicity would become negligible after sixty years or so, because all the short lived daughters decay at a pace defined by the 5.7 years period of ^{228}Ra, which is the longest period member of the 4n natural decay chain (beside ^{232}Th of course). This is the second major difference with uranium extraction, whose descendants ^{230}Th and ^{226}Ra have on the contrary very long periods.

In practice, the situation is more complex because of the extraction efficiency and the presence of uranium, as explained above in § 2. Two extreme cases of monazites (India and Italy) have been considered for radiotoxicity calculations The results are given in
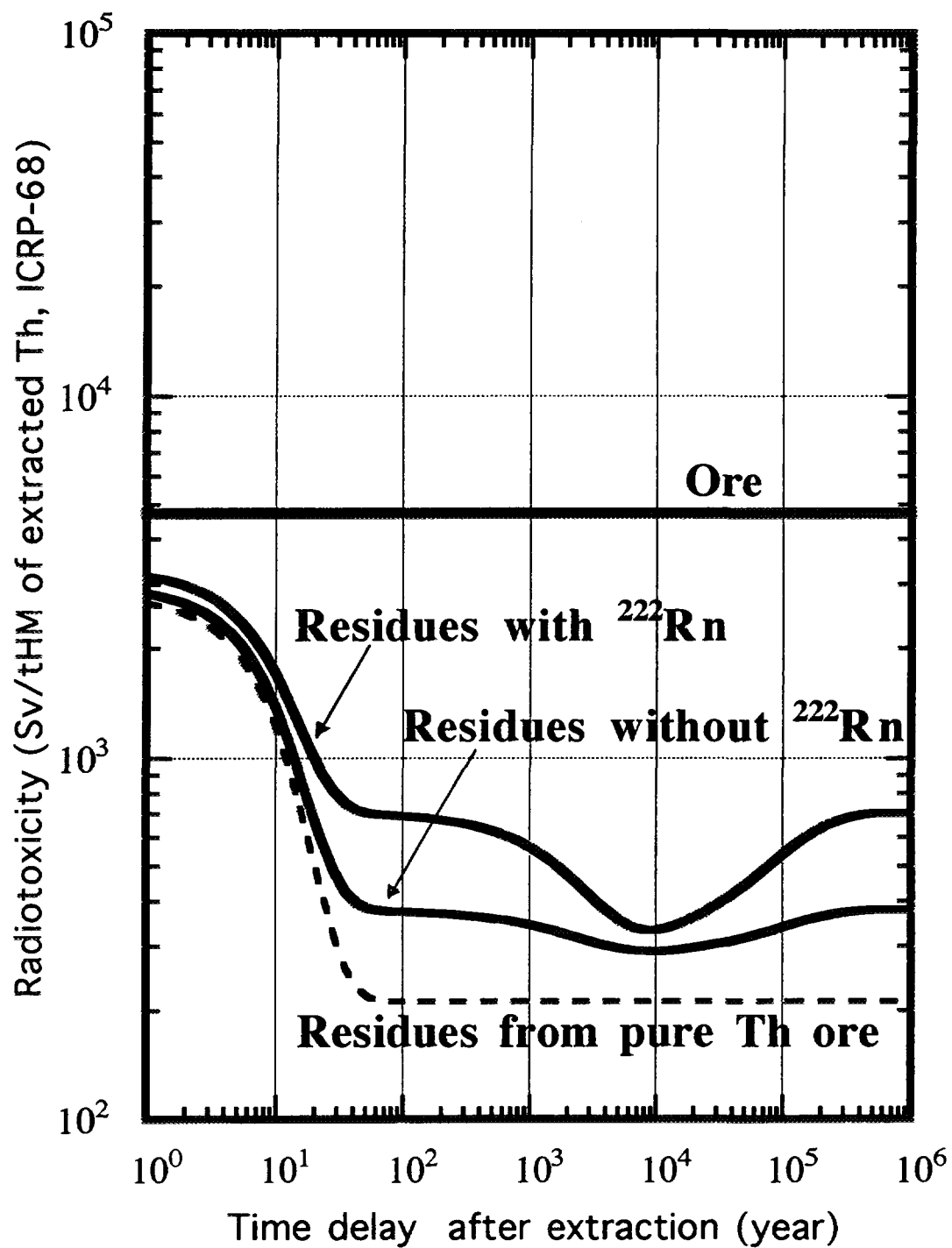
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Fig. 1

RESIDUES FROM INDIAN Th ORE
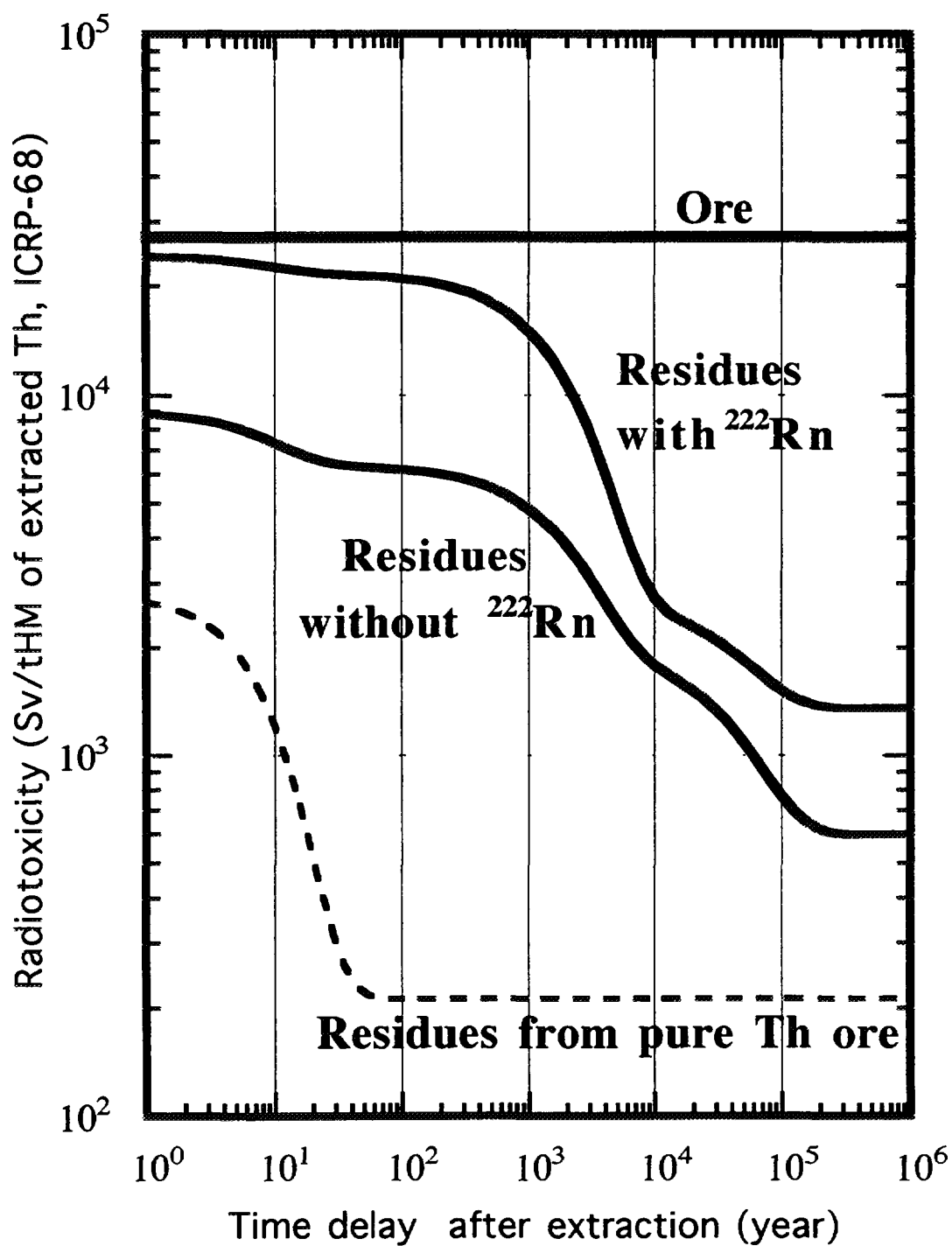(Only Th extracted with 95% efficiency)

Fig. 2

RESIDUES FROM ITALIAN Th ORE
(Th and U extracted with 95% efficiency)

figures 1 and 2, together with the radiotoxicity decay of an hypothetical pur Th ore. For both cases, we have considered the possibility of total ^{222}Rn exhalation and of no exhalation at all.

For the Indian case, the radiotoxicity evolution is governed by the ^{226}Ra decrease (^{230}Th has been extracted). After 10 000 years, the radiotoxicity increase to secular equilibrium is due to ^{230}Th building from ^{238}U. One observes that the radiotoxicity is due to the uranium left with these residues, and also to the unextracted thorium.

The Italian case is somewhat different, because 95 % of uranium is supposed to have been extracted together with thorium. The evolution of radiotoxicity is completly governed by the ^{226}Ra decay, because its precursor ^{230}Th has been removed. Of course, the radiotoxicity is completely governed by uranium residues.

5.   Comparison with uranium extraction

One observes that, for the same amount of heavy metal extracted, the ore radiotoxicity is roughly three times greater for uranium than for thorium (in the ratio of their periods). In figure 3 we represent the radiotoxicity evolution related to the extraction of 1 ton of uranium. This evolution is governed by ^{230}Th decay (periods of 80000 years), and by the unextracted uranium after 10^6 years. One observes that, whatever the origin of the Th ore is its mining residues show a much smaller radiotoxicity a factor than the corresponding one for uranium extraction by a factor bying between 20 to 50.

In conclusion, thorium extraction shows less long term radiological burden than uranium extraction, in terms of :

- radon exhalation ;
- long term residues radiotoxicity.

The differences depend on the amount of uranium mixed with the thorium ore, and on the economical decision to extract or not the accompanying uranium, together with thorium.

These significant differences between uranium and thorium mining have to be considered in view of some estimated long term real radiological impacts due to uranium residues, which could reach a value of the order of 1 mSv/year, the dose limit recommanded for the public by the recent ICRP-60 [7]
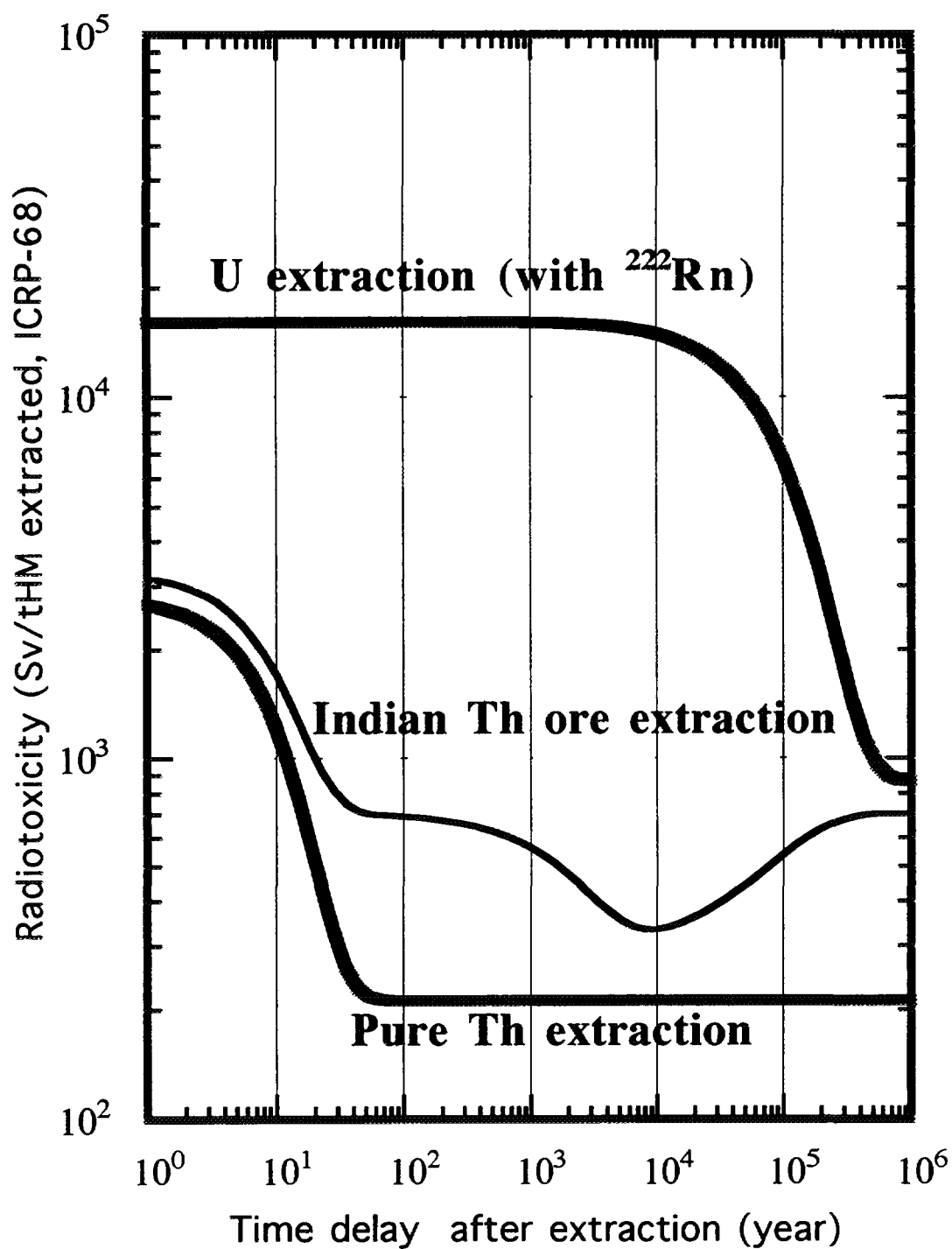
Fig. 3

COMPARISON BETWEEN VARIOUS RESIDUES
(95% extraction efficiency)

The presence of uranium may also have some incidence on the use of extracted thorium in reactors. For example, the contamination with ^{230}Th, which is a uranium's daughter, may increase the amount of ^{232}U, in irradiated fuels, by successive neutron captures. It is well known that ^{232}U is a very strong 2.6 MeV γ ray emitters (from ^{208}Tl) posing a serious radioprotection problem, especially at the Th-U fabrication stage.

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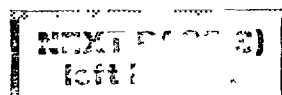
USE OF PLUTONIUM IN PEBBLE BED HTRs
IN A TWO BALL TYPE CONCEPT

M. KHOROSHEV, E. TEUCHERT
KFA Jülich,
Germany

Presented by P.-W. Phlippen

Abstract

Most of reactors can achieve only incomplete utilisation of plutonium. Insertion of plutonium as a mixed oxide with uranium (PuO2+UO2) brings only the partial burnup of Pu, as it is in parts compensated by the Pu-build-up process of U-238 breeding.

Comparing with other reactors the HTR has some unique features that can be used for the more complete burning of plutonium. The different fuel materials can be inserted into different coated particles. In pebble bed reactors the different coated particles can be loaded in different balls, and the balls can have different number of passes through the reactor until they reach the target burnup.

The two ball's concept proposes use of plutonium feed coated particles with a diameter of 220μ, packed in feed balls with heavy metal loading $3g_{HM}$/ball. Thorium with the admixture of high enriched uranium is inserted in coated particles with a diameter of 500μ and packed in breed balls with a content of heavy metal of $20g_{HM}$/ball.

It should be interesting to notice, that both types of balls can be exposed by 60% longer in the reactor without any additional requirements of plutonium or feed-uranium inserted into the fuel elements. The burnup of plutonium increases up to 90%, which corresponds the limitations for feed particles.

The study shows that pebble bed reactors, operated in Pu/Th-cycle provide a simple possibility for achieving very high destruction of plutonium. This is possible because Pu and Th can be inserted into different balls, and each type can be circulated through the reactor as often, as the constraints of target burnup allow.

Summary

Most of reactors can achieve only incomplete utilisation of plutonium. Insertion of plutonium as a mixed oxide with uranium (PuO2+UO2) brings only the partial burnup of Pu, as it is in parts compensated by the Pu-build-up process of U-238 breeding. The content of plutonium in the unloaded fuel is reduced only by factor 1/3, comparing with the inserted amount of it. Certainly, the fraction of denaturation Pu-fiss/Pu-tot reduces to less than 0.5. Considerably stronger diminution of the amount of plutonium can be achieved by its insertion as mixed oxide with thorium, but the burnup of Pu is limited in that case by maximal power and burnup of the fuel elements.

Comparing with other reactors the HTR has some unique features that can be used for the more complete burning of plutonium. The different fuel materials can be inserted into different coated particles. In pebble bed reactors the different coated particles can be loaded in different balls, and the balls can have different number of passes through the reactor until they reach the target burnup.

The possibility of the insertion of fuel material into different coated particles was originally developed for the high temperature reactors with block-type prismatic fuel elements. In Fort. St. Vrain reactor the thorium was inserted as ThO_2 in larger particles, so-called "breed particles", which can achieve the burnup up to 100 Mwd/kg$_{HM}$. The fissile material, high enriched uranium as UC_2
was inserted in "feed particles" where it burns up to about 90%.

Up to now in pebble bed reactors only mixed oxide coated particles have been applied, having the target burnup about 100-200 Mwd/kg$_{HM}$. Therefore, an initial study for the Pu/Th-cycle was performed with mixed oxide coated particles, having the target burnup 100 Mwd/kg. The isotopic composition of plutonium was taken Pu^{239}/Pu^{tot} = 70/30, which corresponds closely to the LWRs unloaded fuel. Addition of high enriched uranium is necessary in that case for achievement of criticality. Similar to other reactors such mixed-oxide concept results in an incomplete usage of the plutonium. Pu-239 burns by to 84%, total plutonium - up by 61%.

The two balls concept proposes use of plutonium feed coated particles with a diameter of 220μ packed in feed balls with heavy metal loading 3g$_{HM}$/ball. A limited study on such concept has been published by K. Yamashita et al. /1/ in 1994. Thorium with the admixture of high enriched uranium is inserted in coated particles with a diameter of 500μ and packed in breed balls with a content of heavy metal of 20g$_{HM}$/ball. The case 1 in Table 1 illustrates the representative calculational results following closely the known-mixed-oxide case. For both fuel types the possible target burnup is by far not reached here.

Table 1

Case NR.	1.	2.	3.
Heavy metal loading in balls feed/breed, g/ball	3.0/20	3.0/20	3.0/20
Burnup feed/breed, MWd/kg$_{HM}$	560/33	776/65	791/101
Average burnup , MWd/kg$_{SM}$	100	158	191
Use of plutonium Pu-239,%	84.6	99.5	99.9
Pu-tot, %	61.3	90.3	94.5
Power fractions in balls feed/breed, %	77/23	68/32	58/42
Temperature coefficients ΔKeff/ΔT Doppler, 10^{-5}/°C	-1.7	-1.7	-1.5
Moderator, 10^{-5}/°C	-4.3	-3.1	-2.1

It should be interesting to notice that both types of balls can be exposed by 60% longer in the reactor (Case 2) without any additional requirements of plutonium or feed-uranium to be inserted into the fuel elements. Obviously, the fissile material inventory remains well balanced by the thorium breeding process, and the increase of absorption by fission products is compensated by the burning of plutonium. The burnup of plutonium increases up to 90%, which corresponds the limitations for the feed particles. The burnup of breed balls doubles up to 65 Mwd/kg$_{HM}$, which is far below the allowable limit.

By the increase of the U-235 admixing in the breed balls it is possible to rise their burnup also to the admissable target value (Case 3). In that case the plutonium in the feed balls is burnt up to 95%, provided that the performance of the feed particles can be developed that far. Otherwise, feed balls must be unloaded earlier and fresh feed balls can be inserted instead into the core.

For the case no. 3 the burnup history of the plutonium is outlined in figure 1. The fresh elements contain 3.0g of plutonium comprising 2.1g of Pu-239 and 0.9g Pu-240. In the minitial phase the Pu-240. In the initial phase the Pu-240 reduces less strongly than Pu-239 because its annihilation is partly counterbalanced by the productions from the capture process in Pu-239.
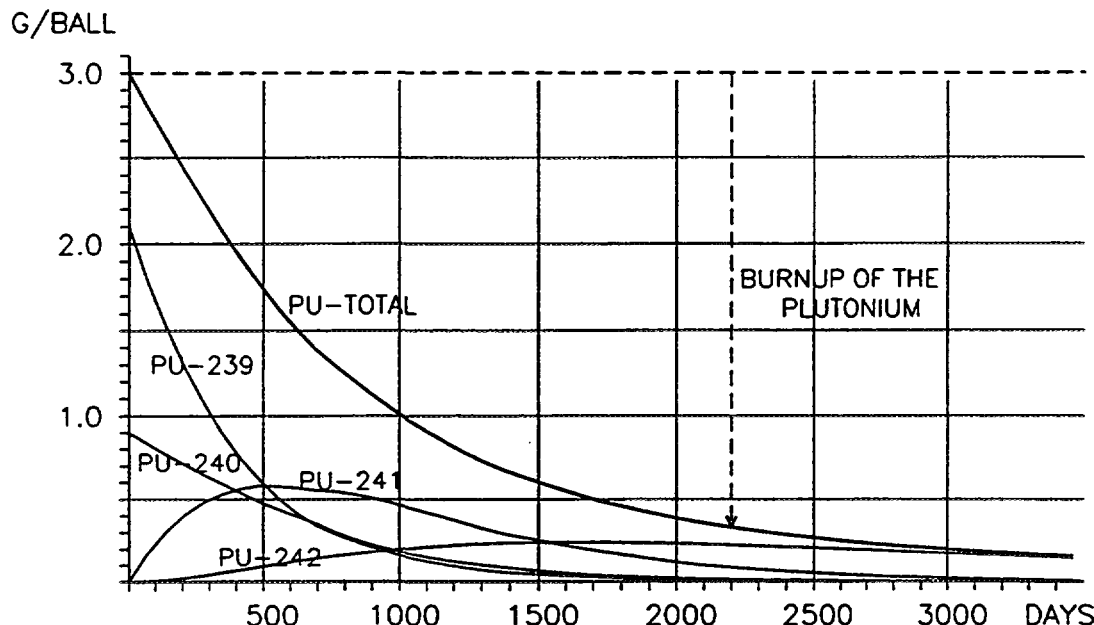
Fig.1 Plutonium burnup history

At the time of 700 days the isotopes become equal. Here, about 40% of the plutonium consists of the bred Pu-241, which is also fissile.

At ½ of the irradiation time, i.e. at 1730 days, the Pu-composition consists by 87% out of the bred higher isotopes Pu-241 (38%) and Pu-242 (49%). If the elements would be unloaded at that time, the Pu-content of the spheres would be reduced to 16%, compared with the initially loaded amount. Thereupon, over a longer storage time of 100 years, the content would reduce to 10.8% because of the decay of Pu-241 with the half life of 14.4 years.

During the second half of the irradiation time even the Pu-242 burns slowly down. At the end it is reduced to ½ of its maximum value, and it forms 94% of the total plutonium composition.

The dotted arrow gives the difference between the time dependent content of total plutonium and the loading at start-up. That range indicates the burnup. At the end the burnup is as high as 95%.

Clearly, in order to achieve reasonable burnup of the plutonium, the feed balls could be removed earlier from the cycle than in the case no. 3 (Table 1). That would bring the advantage that more fresh Pu-containing feed elements, will be loaded, i.e. more plutonium can be fed into the reactor at the cost of only very slight reduction of its burnup.

Table 2 shows the advantages of the flexibility of the HTR's shuffling scheme for burning plutonium using separate feed/breed fuel loading. Loading 3.0 gr Pu per ball was found to be most profitable by summing up all of the criteria considered in a pre-study. By changing the fraction of Pu-balls and number of their passes through the core three criteria were studied:

- increasing of the Pu per Gwd initially loaded;
- minimising of the discharged plutonium in spent fuel elements;
- minimising of the amount of U-235 inserted into the breed balls, having the target burnup 100MWd/kg_{HM}

As it is shown before, the insertion point of equal fractions of feed plutonium balls and breed thorium-uranium balls, travelling in parallel through the reactor (Case 5:5) allows to get almost zero amount of the rest Pu-239 - less the 0.1% - and minimal value of Pu-total in the unloaded fuel - about 5.5%. However, a relatively high amount of U-235 is needed as driver fuel in that case, - 0.413kg/GWd, and the amount of loaded Pu-239 per Gwd is relatively low in that case.

Table 2: Variation of the number of passes through the core for the plutonium balls

Number of passes feed :breed	2:8	3:7	4:6	5:5
HM , gr/ball feed/breed	3.0/20	3.0/20	3.0/20	3.0/20
Burnup, feed/ Mwd/kg $_{HM}$ breed	304/101	746/109	774/108	791/101
Fuel supply - discharge, kg/GWd				
U-235	0.5334-0.1929	0.1447-0.0181	0.3039-0.0407	0.413-0.0574
Pu-239	1.4538-0.6229	0.6500-0.0064	0.5478-0.0013	0.469-0.0003
Pu-tot	2.0795-1.3587	0.9297-0.1243	0.7835-0.0676	0.671-0.0368
Rest Plutonium in unloaded fuel elements				
Pu-239	42.840%	0.984%	0.237%	0.064%
Pu-total	65.40%	13.36%	8.62%	5.47%
Temperature coefficients, ΔKeff/ΔT				
Doppler, x10⁻⁵	-1.36	-1.54	-1.55	-1.52
Moderator, x10⁻⁵	-4.02	-3.30	-2.90	-2.08

Reducing of the feed elements residence time helps considerably in saving uranium and increases the plutonium demand A reasonable compromise is somewhere between cases 3 7 and 4 6, where the largest amount of Pu-239 - 0 65 and 0 55 kg/GWd, respectively - can be loaded into the reactor, and minimum of U-235 is required in the breed fuel elements The rest of Pu-239 is less than 1% in spent elements The rest of Pu-total is also rather low and does not exceed 14%

It is seen, that only two passes of the feed balls through the core (Case 2 8) are not enough for achieving the reasonable burning of plutonium Rest Pu is more than 65% in that case

Use of reactor-grade plutonium (30% Pu-240) brings negative temperature coefficients in all cases observed Other criteria, which are considered for the selection of the final reference case, such a power peaking and maximal power production in fuel elements are fully satisfied in cases 5 5 and 4 6 In case 3 7 the power fraction of the breed balls is relatively low Here, detailed analysis must find, whether the negative moderator coefficient will fulfill the demands of safety

The Doppler coefficient is due to the broadening of thorium resonances and so it works according to temperature changes of breed balls, whose power fraction varies from 23% to 41% The moderator coefficient is predominantly due to shifting of the thermal neutron spectrum in the energy range of the Pu-240 resonance at 1 eV being located in the feed balls

The study shows, that pebble bed reactors, operated in Pu/Th-cycle, provide a simple possibility for achieving very high destruction of plutonium This is possible because Pu and Th can be inserted into different balls, and each type can be circulated through the reactor as often as suggested by the optimisation and by the allowable target burnup

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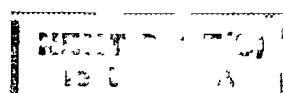
REDUCTION REQUIREMENTS FOR ACTINIDES WITH SPECIAL REGARD TO THE ISOLATION TIME AT FINAL DISPOSAL

P.-W. PHLIPPEN
Institute for Safety Research and
Reactor Technology,
Jülich, Germany

Abstract

The additional knowledge acquired about the metabolism of the actinide elements, and the experience with the development of cancer risk have resulted in several realignments of the ALI limits by the ICRP. This needs a re-evaluation of the toxicity potential inherent in the nuclear fuel cycles of nuclear reactors and a re-evaluation of drawn conclusions. The radiotoxical evaluation of actinides and long-lived fission products are presented and discussed with special regard to Partitioning and Transmutation (P&T) issues. Detailed information about balancing the toxicity potential flow and its growth during the nuclear fuel cycle is given in order to determine a reference value for the comparison of natural and man-made toxicity. Calculations for different fuel types are exhibited and the resulting toxicity potentials are compared to these reference values in order to solely quantify in an idealized way the consequences of human action. The long-term toxicity potentials of discharged PWR-fuels in case of direct disposal as well as Pu-recycling within MOX elements using U and Th are presented.

The inherent drawbacks lead to the conclusion that with respect to a modified goal of final disposal only a full-scale P&T scheme is able to achieve long-term toxicity potentials on the same level as that of fresh fuel decaying naturally. Thus, the storage in a repository can relay more heavily on engineered barriers.

INTRODUCTION

All nations using nuclear energy intend to dispose their high level nuclear wastes (HLW), i. e. spent nuclear fuel or vitrified high level liquid waste from reprocessing, in geologically stable and hundred millions of years old ages, which are to be qualified for this task by detailed examinations. The retention of radioactivity from the biosphere is to be achieved by a multi-barrier system, which is dominated by geological transport mechanisms and not by engineered barriers. The release from those geological ages may only rise by transport in water. But the delay caused by this transport mechanism is expected to be extremely high, so that in accident conditions dose risks on the surface my occur at earliest after some ten thousand years since disposal [1,2]. In the normal evolution scenario, the long-term risk is dominated by ^{237}Np. Nevertheless, the calculated annual effective dose rates are some orders of

magnitude lower than the natural dose rate of about 2.4 mSv per year; this yields as well, if one includes the spread over the globe.

Despite these very positive results, especially if they are compared with risks raising from the operation of nuclear power plants or coal fired plants for electricity generation [3], this way of final disposal is not accepted by a large fraction of the population in many nations. This lack of acceptance is argued for as follows:

- The long-term presence of radiological risks is not acceptable because of the unpredictable evolution of the earth and of human beings.

- It is not believed in the long-term stability of the geological age after human intrusion and decay heat deposition.

- The valuation of radiological risks changes with increasing knowledge about the metabolism of elements. Thus conclusions, which are valid today - on the basis of incomplete understanding of the metabolism - may become invalid in future.

- From the ethical point of view, today's generations are not allowed to take the advantages of nuclear energy utilization without solving the risks in an accepted way.

In nuclear technology, usually experiments help to clear raising problems or questions. But especially the second argument mentioned cannot be refuted by experimental means. On the other hand, it is neither necessary nor recommendable to dispose today's HLW immediately. A long-term intermediate storage helds open the application of further developments in reprocessing and recycling; on the other hand mining requirements are hardly reduced by recycling residual fissile isotopes. Applying the last argument verbatim for fossil energy consumption, we would have to stop it immediately until the climatic problems have been solved conveniently.

HISTORICAL DEVELOPMENT OF RECOMMENDATIONS

The influence of increasing knowledge about the metabolism of special elements as well as about the significance of radiation to humans may be demonstrated comparing the regulations denoting the limits on intake of radioactive isotopes. A comparison between the twentyfive years old limits given with the Code of Federal Regulations (CFR) [4], the 1980 and 1988 (ICRP-30) [5,6] recommendations of the International Commission on Radiological Protection (ICRP), on which the German Radiation Protection Ordinance of 1989 (StrlSchV) [7] bases, and the newest recommendations of the ICRP [8,9] is given in Table 1, showing data of the ingestion case. For comparison, the annual limits on intake (ALI) and the corresponding reference annual dose which they are basing on have been converted to Bq/Sv; in the conversion of the CFR concentrations a water intake of 1 m^3/a is assumed.

For fission products, the shifts in the evaluations of ^{90}Sr and ^{137}Cs should be noted for medium-term projections; owing to the raising of the ^{90}Sr limit value, these now lie at about the same level. For long-term projections, the decrease in the evaluation of

Table 1: Comparison of the maximum annual radioactivity intake per Sv via ingestion for major nuclides with view of evaluating the toxicity potential of radioactive waste [4-9]

Regulation Year Nuclide	Half-life	ICRP-61 1991 Bq/Sv	ICRP-30 1980 Bq/Sv	ICRP-30 Part 4 1988 Bq/Sv	10 CFR 20 1971 Bq/Sv	StrlSchV 1989 Bq/Sv
Sr-90	28.5 a	3.0E+7	2.0E+7		2.2E+6	6.0E+6
Cs-135	2E+6 a	5.0E+8	6.0E+8		7.4E+8	0.0E+0
Cs-137	30.17 a	5.0E+7	8.0E+7		1.5E+8	8.0E+7
Tc-99	2.1E+5 a	1.5E+9	2.0E+9		1.5E+9	8.0E+8
I-129	1.57E+7 a	1.0E+7	4.0E+6		4.4E+5	2.0E+6
Pb-210	22.3 a	1.0E+6	4.0E+5		7.4E+5	2.0E+5
Po-210	138.38 d	4.5E+6	2.0E+6		5.2E+6	6.0E+5
Ra-226	1600 a	4.5E+6	1.4E+6		2.2E+5	8.0E+5
Pa-231	3.28E+4 a	5.0E+5	1.4E+5		6.7E+6	8.0E+4
U-233	1.59E+5 a	3.5E+7	8.0E+6		2.2E+8	6.0E+6
U-234	2.45E+5 a	3.5E+7	8.0E+6		2.2E+8	6.0E+6
U-235	7.04E+8 a	3.5E+7	1.0E+7		2.2E+8	6.0E+6
U-236	2.342E+8	3.5E+7	1.0E+7		2.2E+8	0.0E+0
U-238	4.47E+9 a	4.0E+7	1.0E+7		3.0E+8	6.0E+6
Np-237	2.14E+6 a	1.5E+6	6.0E+4	4.0E+5	2.2E+7	2.0E+5
Pu-238	87.74 a	2.0E+6	6.0E+6	6.0E+5	3.7E+7	4.0E+5
Pu-239	2.41E+4 a	2.0E+6	4.0E+6	6.0E+5	3.7E+7	4.0E+5
Pu-240	6550 a	2.0E+6	4.0E+6	6.0E+5	3.7E+7	4.0E+5
Pu-241	14.4 a	1.0E+8	2.0E+8	2.0E+7	1.5E+9	1.8E+7
Pu-242	3.76E+5 a	2.0E+6	6.0E+6	6.0E+5	3.7E+7	0.0E+0
Am-241	432.6 a	1.5E+6	1.0E+6	6.0E+5	3.0E+7	4.0E+5
Am-243	7370 a	1.5E+6	1.0E+6	6.0E+5	3.0E+7	0.0E+0
Cm-243	28.5 a	2.5E+6	1.4E+6	8.0E+5	3.7E+7	4.0E+5
Cm-244	18.11 a	3.0E+6	1.8E+6	1.0E+6	5.2E+7	6.0E+5

[129]I by a factor of ten should also be noted. New recognitions about the metabolism of transuranium elements (TRU) [10] triggered a partial revision of the ALIs in ICRP-30 in 1988 [6]. The limit value of [237]Np was raised by a factor of seven, while the limits for Pu were reduced by around one order of magnitude and those for Am and Cm were approximately halved. Now the limits for TRU-elements in CFR and ICRP-30(4) differ by nearly two orders of magnitude! Updated cancer mortalities and the adaptation of a new dosimetry system lead to the 1990 recommendations (ICRP-60/61), which reset the more restrictive limits of ICRP-30(4) nearly to those of ICRP-30.

Figure 1 shows the timedependence of the toxicity potential for actinides and fission products, comparing ICRP-61 and 10 CFR 20. Basically, as a result of the changed classifications of the TRU, the total curves for fission products and actinides have shifted in such a way that the latter determine the toxicity potential practically from the very beginning. The familiar development pattern of the total toxicity potential, where it reduces by approximately two orders of magnitude over the first 300 years as result of fission product decay, is no longer in evidence. The development over
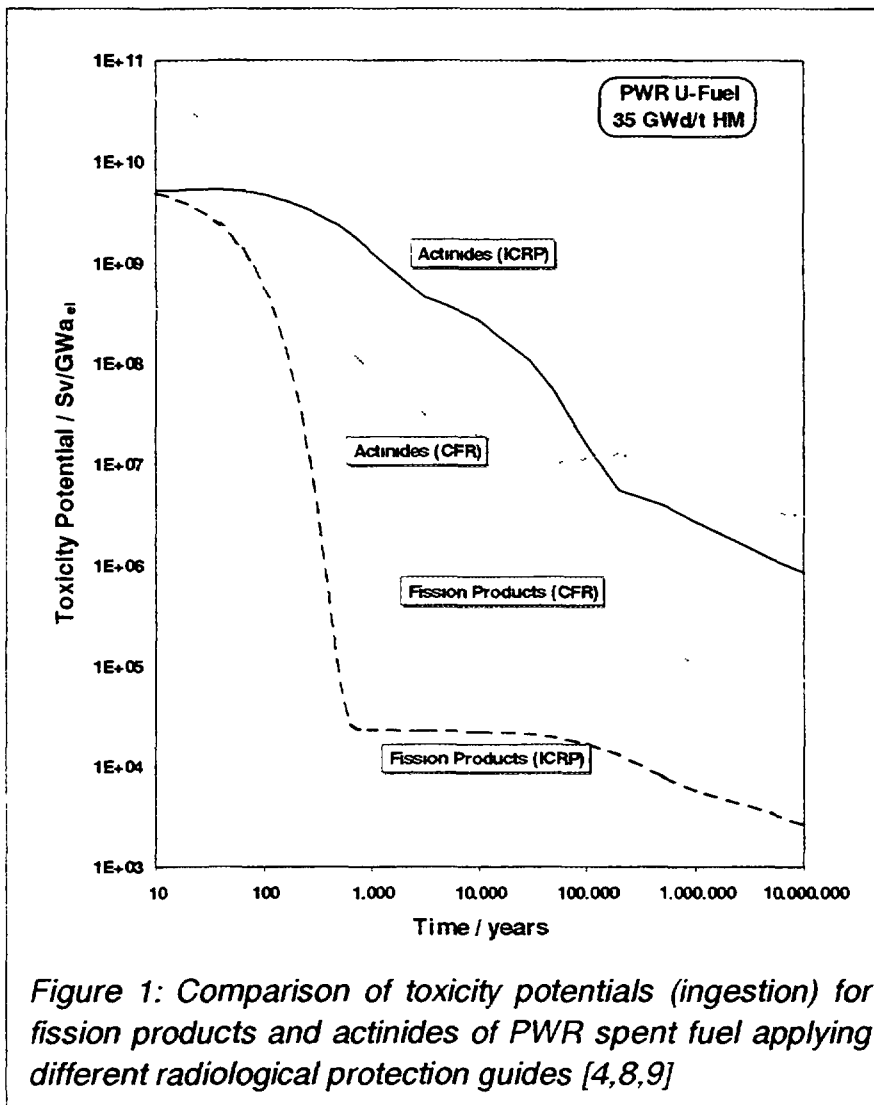
Figure 1: Comparison of toxicity potentials (ingestion) for fission products and actinides of PWR spent fuel applying different radiological protection guides [4,8,9]

time is, in effect, determined by the decay of Pu. The long-lived fission products ^{99}Tc and ^{129}I play only a subsidiary role in this scenario. The long-term view shows, that the ranking between fission products and actinides does not change, but the distance increases by one order of magnitude due to the decreased ^{129}I influence. Discussions in more detail are given in [11, 12].

BALANCE OF TOXICITY POTENTIAL

In order to get a reference value for that toxicity potential to achieve with the decaying spent fuel, usually the toxicity potential of the amount of natural Uranium - in equilibrium with its decay products - needed for the production of the initial fuel is used. Afterwards the timedependent toxicity potentials are compared and passing the crossing point, the additional risk of the spent fuel is taken as vanished. This is not the correct story, as *Figure 2* shows for two different balance schemes.

Regarding first the reference value of uranium ore, we easily see, that we have to balance the amounts of toxicity potential taken from the ground (uranium ore in equilibrium) with those ones returned to the

Figure 2: Scheme to balance the human influence on the increasing toxicity potential by nuclear energy usage

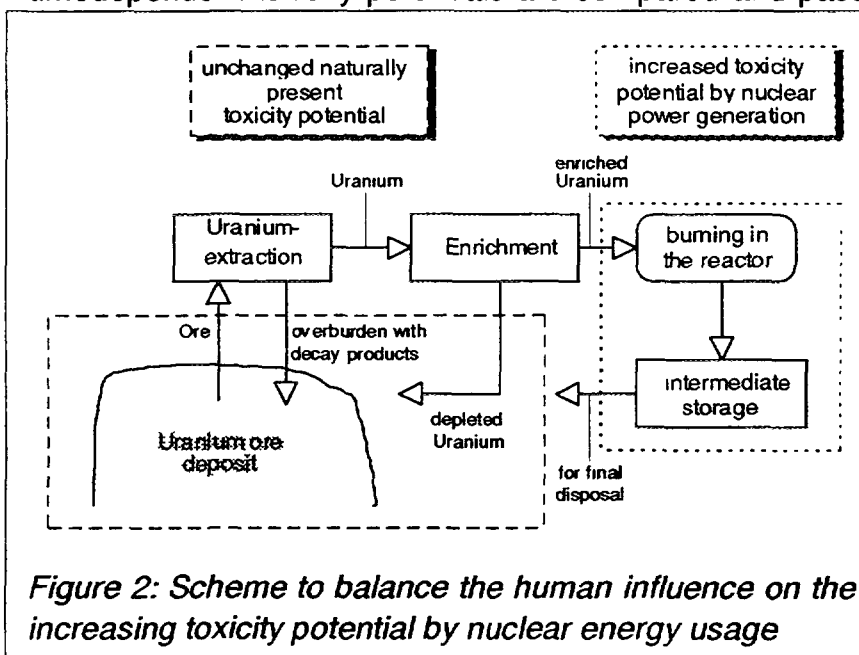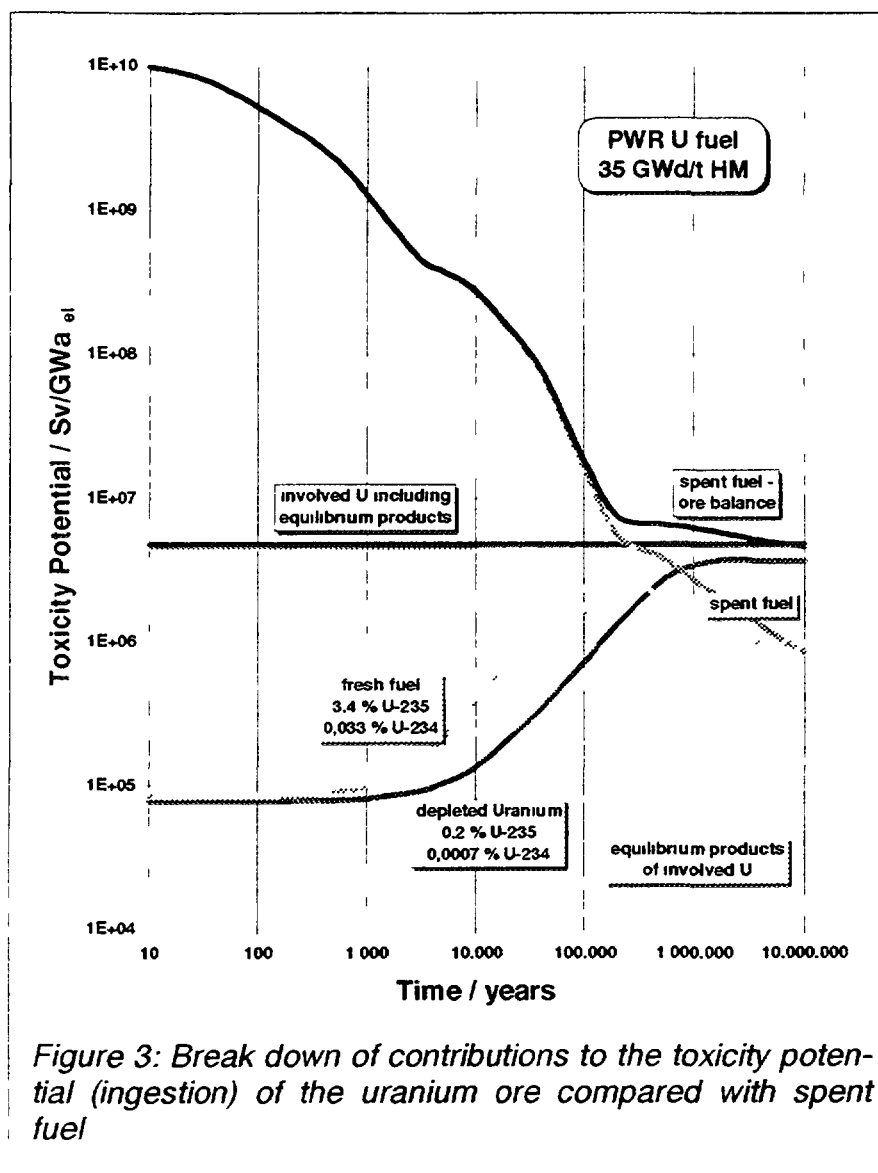ground (overburden with decay products, depleted uranium and spent fuel). To simplify the picture, I've assumed a direct disposal of spent fuel; additionally the depleted uranium is not used in today's nuclear power plants, so that it is to be seen as „returned toxicity potential" to the ground. After disposal, the equilibrium decay products determine the toxicity potential of the ore with 96 % of the total value; the uranium metal fractions contribute approximately 2 % each (*Figure 3*). But some hundred thousand years later, the depleted uranium becomes dominant. In fact, this usually neglected fraction determines the long term toxicity potential after nuclear energy usage!

Chart labels: PWR U fuel 35 GWd/t HM; involved U including equilibrium products; spent fuel - ore balance; spent fuel; fresh fuel 3.4 % U-235 0,033 % U-234; depleted Uranium 0.2 % U-235 0,0007 % U-234; equilibrium products of involved U. Y-axis: Toxicity Potential / Sv/GWa el. X-axis: Time / years (10, 100, 1 000, 10.000, 100.000, 1 000.000, 10.000.000).

Figure 3: Break down of contributions to the toxicity potential (ingestion) of the uranium ore compared with spent fuel

Regarding the second much smaller balance boundary depicted in *Figure 2*, we recognize easily, that only the fresh uranium fuel feed into the nuclear reactor can act as reference value. The overburden of mining including the decay products as well as the depleted uranium are toxicity potentials which are not influenced by burning fuel in a nuclear reactor. They have only been moved out of the deposit and of course they have to be buried to avoid dispersion into the environment and to avoid an intake by humans. Thus, comparing PWR spent fuel with decaying fresh fuel, we must draw the same conclusions as before (ref. to *Figure 3*): The increased toxicity potential of the spent fuel does not meet the reference value before some million years of decay. Thus, the effect of nuclear energy usage on the toxicity potential vanishes only after a very long time. In case of direct disposal and, as we see later, for today's reprocessing residiuals, the only way to exclude them from the biological environment is the disposal in a geological age.

REQUIREMENTS FOR ACTINIDE REDUCTION

Referring to the mentioned lack of acceptance and the afore depicted isolation requirements for final disposal, we should try to find a way to overcome most of the raised problems. Doing so, a critical review of the goal for final disposal is absolutely necessary, aiming to reduce the exclosure time requirements drastically. The newly defined goal may become as follows:

Exclosure of radioactive material from the biosphere by engineered barriers until the increased toxicity potential caused by nuclear energy usage has vanished the natural level.

Thus, comparing PWR spent fuel with decaying fresh fuel, we must draw the same conclusions as if we compare with natural uranium deposits (*Figure 4*): The increased toxicity potential of the actinides vanishes only after long decay times and needs a geological age for its long-term enclosure. Assuming today's reprocessing with 0.5 % U and Pu-losses, this conclusion holds as well. The reader should be aware, that the reprocessing curve implies a fission of the extracted U- and Pu-fractions, an implication which is not realized! *Figure 7* gives an imagination of the timedependency assuming uranium to be disposed too. Regarding HTR-fuel with a burnup of 80 MWd/t HM, which is more than twice as high as for the shown PWR-fuel, these conclusions are also valid [12].

Hence, to meet the modified goal for final disposal, additional measures have to be taken which are discussed as „Partitioning and Transmutation" (P&T) and may follow four steps:

1. Partitioning of long-lived elements from the spent fuel,

2. Transmutation of the partitioned elements into short-lived ones,

3. Encapsulation of the residuals by corrosion- and/or leach-resistant containers and

4. Disposal in geological ages.

The needed duration of the exclosure from the biosphere (1,000 years give an idea of the timespan in mind) must be guaranteed by engineered barriers and determines the tolerable level of fuel cycle-losses. By this, the geological age serves only as storage area to protect the stored waste against external impacts. Passing the lifetime of the waste containers the age includes the „surviving" toxicity potential which is less toxic than the reference value. Thus the residual toxicity potential remains in a geological environment with comparable retention capabilities as the primary deposit.

Corrosion rates in salt-brine or groundwater are well known for container materials from experiments in the field of direct disposal. Dependent on the corrosion regime, wall thickness between 5 cm and 15 cm are large enough to withstand corrosion attacks in salt-brine at least for 1,000 years [13,14]. Results of investigations at poten-
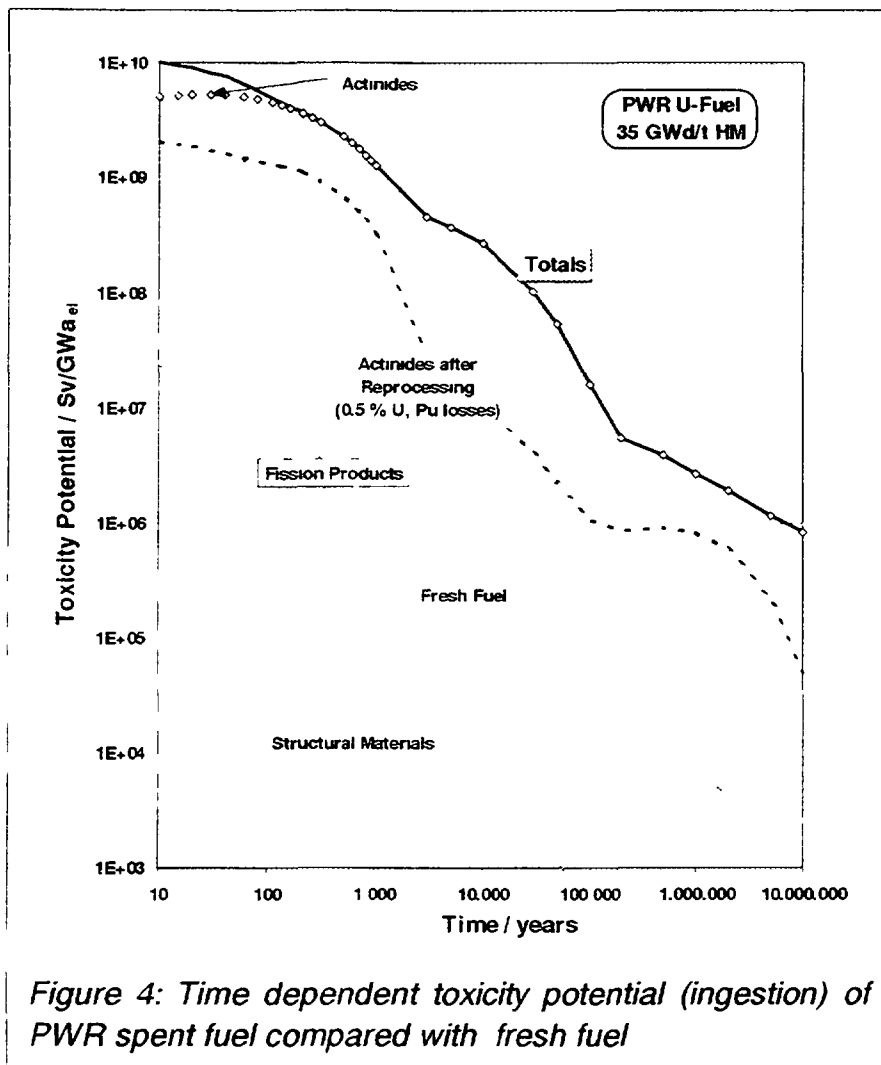
Figure 4: Time dependent toxicity potential (ingestion) of PWR spent fuel compared with fresh fuel

tial final disposal sites are suitable as well. Finally attention must be drawn on P&T in order to show what's necessary to achieve the afore mentioned goal.

As *Figure 4* shows very clear against the reference of fresh fuel, fission products and structural materials do not determine the toxicity potential significantly after some hundred years of decay. The structural material of the fuel elements is less significant than the fission products and passes the reference line after approximately 200 years of decay. Thus relaying on decay times of at least thousand years to diminish the toxicity potential to the level of fresh fuel, only actinides are to be cared for[1].

P&T-REQUIREMENTS

Setting up requirements for P&T applications in order to achieve the (new) goal of final disposal, *Figure 5* shows the contribution to the toxicity potential of each element-chain. These graphs allow the assignment of partitioning needs to the significance of each element. Thus it looks very clear, that Pu and Am are most significant, but U, Np and Cm have less significance. This conclusion differs somewhat from requirements often raised in papers dealing with P&T.

Relating to frequently discussed much higher decontamination ratios for [237]Np, the mobility in the geosphere and the long half-life time influence this stronger requirements. But makes it sense to reduce the accidental individual dose rates of final re-

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[1] The toxicity potential as defined here does not include geo-chemical transport mechanisms in the age. It is well known, that some fission products are much more mobilizable than actinides. Thus they determine the potential dose rates after some ten- or hundred thousand years after repository failure. Because of the recommended shortened decay time such long periodes need not to be cared for.

positories by another order of magnitude, although today's final disposal procedures ensure already some orders of magnitude less individual dose rates [1] than natural ones? I think, that makes no sense.

Regarding ^{244}Cm decaying to ^{240}Pu, it influences the total toxicity potential only marginally, so that a higher decontamination seems not justifiable. This conclusion must be reviewed in case of very high burnups with Pu-recycling, lets say as U/Pu-MOX-fuel, resulting in increasing Cm ratios in spent fuel.
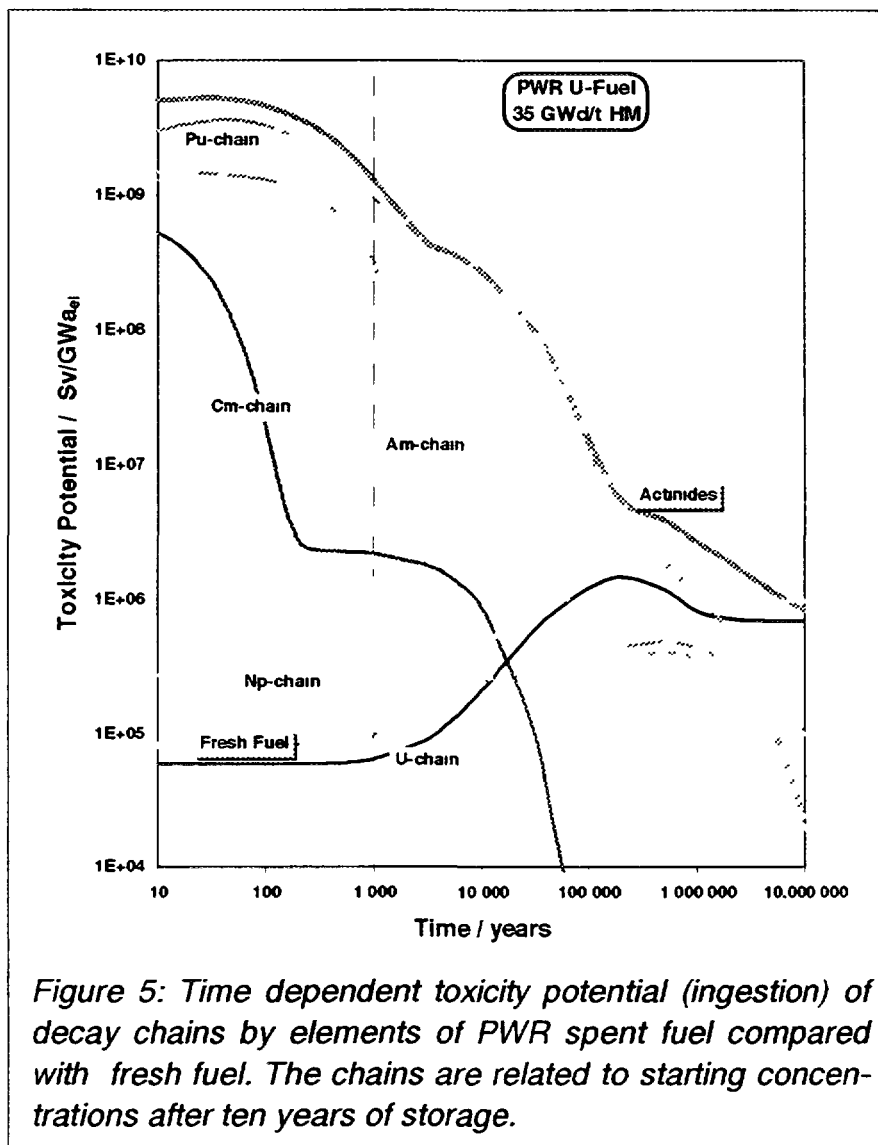
Figure 5: Time dependent toxicity potential (ingestion) of decay chains by elements of PWR spent fuel compared with fresh fuel. The chains are related to starting concentrations after ten years of storage.

Setting up requirements, Figure 7 shows today's reprocessing losses against a sophisticated P&T scheme, assuming total fuel cycle losses of 1 % for U, Np and Cm as well as 0.01 % for Pu and Am. Only these drastic reductions allow to meet the reference values of either uranium ore or fresh fuel within the mentioned timespan. This picture shows, that it makes no difference which balancing scheme is applied if complete balances are established.

PU-RECYCLING IN PWR

Today, a recycling of separated Pu is performed using U/Pu-MOX-fuel elements in PWR. Regarding that as a transmutation step, we must judge the resulting toxicity potential applying the raised goal. Balancing the involved mass flows and the toxicity potential with fresh fuel, one can setup a rough flow sheet as shown in Figure 6, assuming no losses during the fabrication step. Approximately five tons of spent fuel are needed to produce one ton of MOX fuel containing natural uranium as diluent for Pu. As today's recycling modes do not address reprocessed uranium, it is to be dealt like waste, even if no final disposal is foreseen. Therefor reprocessing losses include all actinides except the recycled Pu amount. The result is shown in Figure 8, separated into the contribution of the initial U-based fuel and that of spent MOX-fuel.

Figure 7: Balancing different separation quantities applying two balance schemes
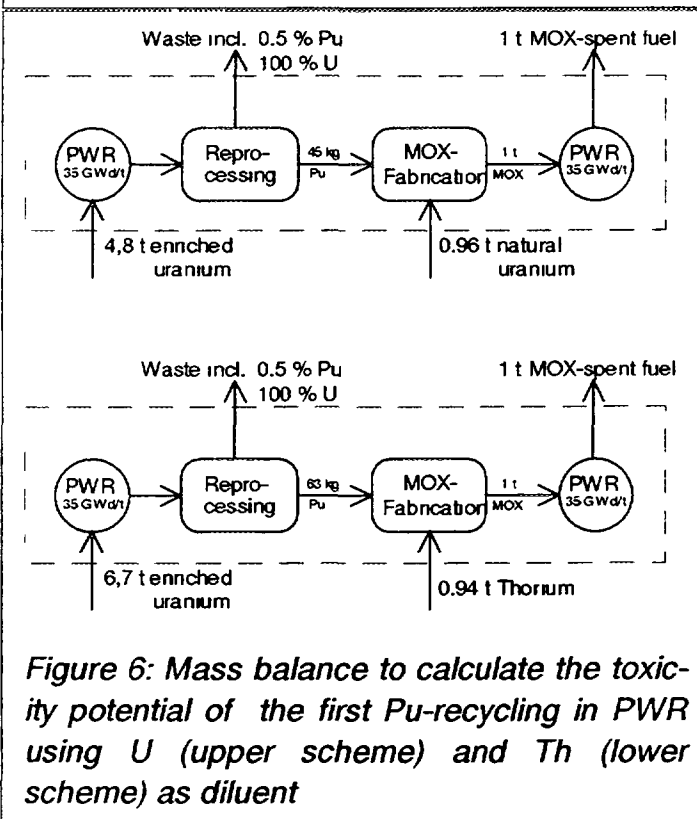
Figure 6: Mass balance to calculate the toxicity potential of the first Pu-recycling in PWR using U (upper scheme) and Th (lower scheme) as diluent

Comparing the total toxicity potential with that of the once-trough-cycle (see *Figure 4*) no reduction has been achieved and thus with respect to an intended transmutation, the goal is not meet. Up to thousand years of storage, the toxicity potential of the Pu-recycle is slightly higher (app. +25 %) than for the once-through-cycle; up to hundred thousand years it is slightly lower (app. -25 %). Of course today's Pu-recycling is not dedicated to transmutation and therefor arguing positively, the additional gained electrical work has not produced more toxicity potential per electricity unit than a once-through-cycle.

Often a recycling of Pu using Th as diluent is discussed. This way avoids breeding of new TRU from the diluent; instead of Pu, ^{233}U is breaded. Thus applying the same burnup, a much higher net consumption of Pu is obvious. Analyzing the depicted alternative Th-recycling scheme in *Figure 6*, we get the toxicity potentials shown in *Figure 9*. The reference line of the initial fuel including enriched uranium and thorium looks like that one of the U-based MOX fuel, caused by the low toxicity potential of natural Th. The behavior of the

79

Figure 8: Time dependent total toxicity potential (ingestion) after Pu-recycling in a PWR with a burnup of 35 GWd/t HM at each step using U as diluent (balance of Figure 6)

Figure 9: Time dependent total toxicity potential (ingestion) after Pu-recycling in a PWR with a burnup of 35 GWd/t HM at each step using Th as diluent (balance of Figure 6)
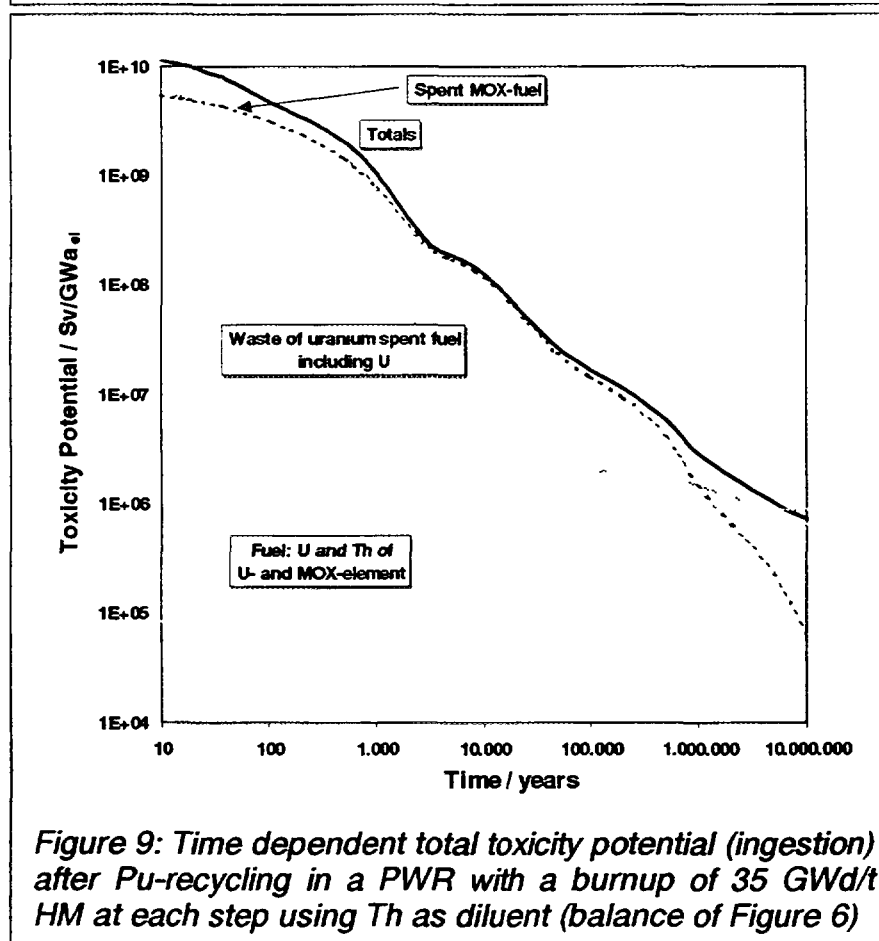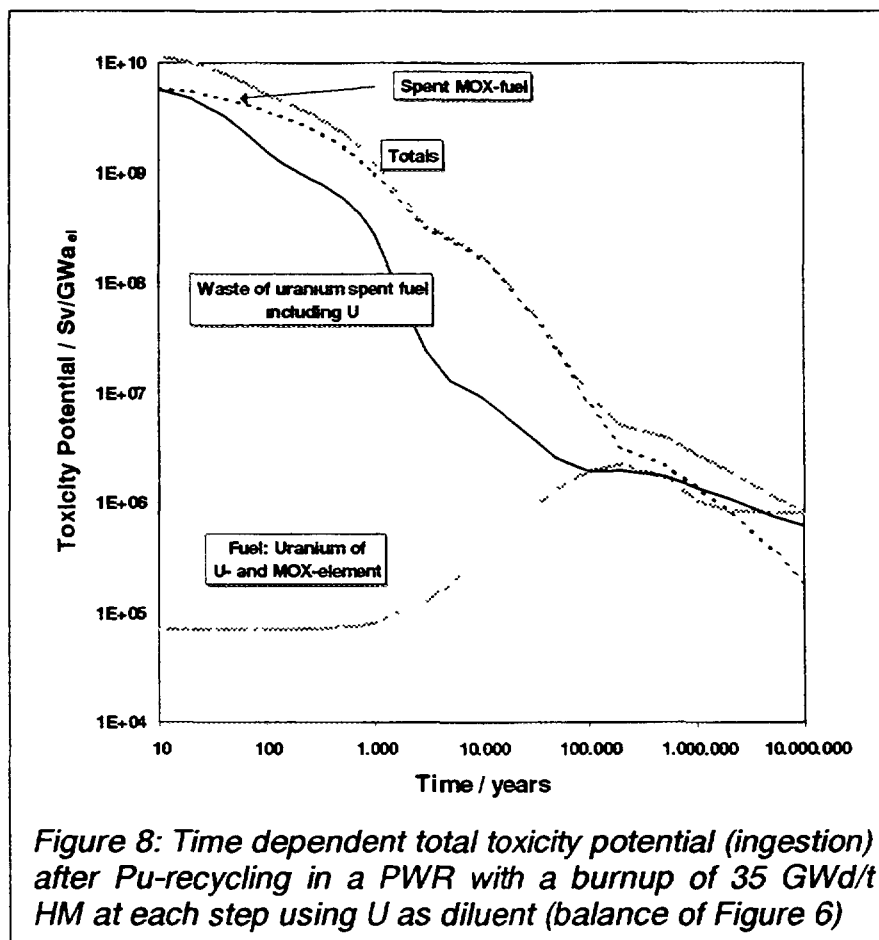
spent Th/Pu-MOX-fuel seems to be the same as U/Pu-MOX-fuel. Referring to the toxicity potential per kWh this conclusion is correct. But during the time-span between hundred thousand and one million years the Th-case shows a higher toxicity potential because of the contribution of the U-chain. Beside this, Pu and Am are dominant even in this case.

Valuing this result, attention must be focused on the initial Pu content. To survive the burnup period in the reactor, the Th based MOX-fuel contains 6.3 % Pu initially which is to be compared with 4.5 % of the U/Pu-MOX one. The burnup of the latter one decreases the Pu-content to 75 % and the TRU-content to 80 % respectively. Regarding the Th/Pu-MOX fuel the initial Pu-content is reduced to 46 % and the TRU-content to 50 % respectively. This different consumption behavior results in the same remaining TRU-masses in the spent fuel and thus both toxicity characteristics must be similar.

Nevertheless, the total TRU-consumption of the Th-based fuel is 3.5 times higher than applying the U-based one. Including newly generated U-isotopes as a drawback, the reduction decreases to 2.8 times. Although using Th-based MOX fuel does not show advantages in terms of the discussed toxicity potential in a first Pu-recycle, however it consumes much more Pu and therefor is favorable in achieving a significant reduction of the Pu stock-piles.

CRITICAL JUDGMENT

The presented way of applying the criteria „toxicity potential" to judge fuel cycles with respect to the potential of P&T to „clean" the residuals of nuclear energy utilization, is an incomplete view of the problem. To get a more complete view, drawbacks of recycling TRU including fuel refabrication, reprocessing and reactor-characteristics under accident conditions are to be added. Doing this, the task of judging potential risks of the final repository vs. real man doses applicated during the additional steps of the nuclear fuel-cycle has to be solved. Some questions and first suggestions within this area my be found in [15].

On the other hand retention capabilities of the geological ages, containing a final repository, are not taken into account applying toxicity potentials for ingestion on the spent fuel instead on the released residuals from the age. But trying to reduce only the potential accidental individual dose rates from repositories, estimated to stay some orders of magnitude below the natural dose rate, in my opinion are no substantial argument for P&T. Thus a new quality of final disposal as depicted for example may be accepted as a weighty argument for implementing P&T to the nuclear fuel cycle. In this sense, neglecting the retention capabilities of the repository are logical within the frame of the proposed definition of the goal for final disposal.

CONCLUSIONS

Most objections against today's envisaged final disposal procedures are based on the unpredictable future behavior of the geological age and the validity of today's judgment of radiation doses received by human beings, which partwise had undergo drastically changes during the last decades. Respecting these objections, the way of final disposal has been redefined to the application of engineered barriers which retain the residuals for at least 1.000 years.

The presented way to get a reference level coming from a balance of the uranium circuit identifies the fresh fuel feed into the reactor as an adequate reference value. It is shown, that simply comparing the toxicity potential of the uranium ore with decaying spent fuel neglects the influence of unused depleted uranium. In order to achieve the new goal, the implementation of P&T requires a reduction of the disposed TRU-residuals by some orders of magnitude relative to their content in spent fuel; reductions by 10.000 for Pu and Am as well as 100 for U, Np and Cm seem to

be necessary to meet the newly defined goal of final disposal. Especially the required reduction ratios for Pu and Am are far away from the actual state of the art. Today both, partitioning- and transmutation-facilities, are not available. To realize them, it needs considerable changes in reprocessing and fuel fabrication. Possibly, this goal can only be meet going another way of nuclear energy utilization.

The assessment of the first recycling of separated Pu into PWR using U- and Th-based MOX fuel shows, that the generated toxicity potential per kWh does not differ significantly for both MOX-cycles as well as compared with the once through cycle. Hence an implementation of these ways do not meet the goal at all. But with respect to a commonly mentioned reason for the implementation of P&T, the reduction of Pu stock-piles, the Th-based cycle reduces the initial TRU-content of the fuel by 3.5 times with respect to U/Pu-MOX fuel. Thus Pu-burning using the diluent Th may be a way to switch to the nearly TRU-free Th-based closed fuel cycle.

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BUILD-UP OF PLUTONIUM ISOTOPES IN HTR FUEL ELEMENTS; COMPARISON BETWEEN COMPUTED PREDICTION AND CHEMICAL ANALYSIS

H. WERNER
Research Center Jülich,
Institute for Safety Research and
Reactor Technology,
Jülich, Germany

Abstract

Irradiation experiments are reported with HTR fuel elements within the AVR reactor. It is shown how to charge fuel elements onto the core to detect them by the burnup measurement facility exactly after passing the inner part of the core one time. So these fuel elements could be discharged directly.
The method of chemical dissolution of the fuel elements is presented and the results of the mass spectrometric analysis are reported.
The analysis by calculation with the reactor simulation program HTR-2000 is described. The results are listed and discussed.
The comparison between the experimental and computed outcomes gives a good agreement

Introduction

The decision to change the HTR fuel cycle from high enriched uranium with thorium (HEU) to low enriched uranium (LEU) made the question important for the validation of calculated build-up of the plutonium isotopes.
This problem was not so urgent using high enriched uranium because of the low amount of U-238 a major source for the plutonium isotopes. The usage of low enriched uranium however led to an increase of the expected quantity of plutonium. So the validation of the calculated results became more important. It was necessary to gain measured data by experiments, in order to compare the results with the calculated numbers.
The principle difficulty to compare measured data gained from chemical investigation of fuel elements from the pebble bed reactor type with computed results is the flowing fuel during power operation.
The fuel elements dont remain standing at the same position during operation rather they are moving through the core.
That causes the uncertainty to assign a life history to a fuel element discharged from the core. So the boundary conditions for the calculation are uncertain and the results are not directly comparable with the measured data obviously.
The direct comparison between measured and calculated dat requires the knowledge of the environment of the fuel element during irradiation exactly. That means the neutronflux, temperature and the residence time have to be so well noted as possible.
In a test reactor the fuel elements are inserted in a certain position stationary. The neutron flux and the temperature of the environment are adjust by the reactor power and measured during the irradiation experiment.
In the pebble bed of this reactor type it is not possible to insert a measurement facility during power operation, to observe the fuel elements or control the environment. Only leaving the core the fuel elements can be controlled by a special equipment. Therefore experiments have to be performed in such a way, that a comparison between measured and calculated results makes sense.
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A detailled knowledge of the reactor facility and the operation running is necessary. Without an opportunity to get any data of inner pebble bed by measurement calculations have to to be performed to describe the environment.

Description of the reactor

The AVR reactor is a high temperature gas cooled and graphite moderated reactor. The thermal power was 46 MW with an electric gross output of 15 MW. The reactor core was cooled by a helium flow rate of 13 kg/sec and pressure of 10.9 bar. The core inlet helium temperature varied about 270 C. The averaged allowed maximum outlet temperature was 950 C. The steam generator worked under a main steam pressure of 72 bar, a main steam temperature of 505 C and main steam flow rate of 56 t/h. Core, steam generator and ist components are placed in a double-walled pressure vessel. The outer vessel has a diameter of ca 6 m and a hight of 25 m. The core itself consits of a loose filling of abbout 100.000 fuel element spheres with a diameter of 6 cm. The loose spheres are placed in a cylindrical vessel, which is coated by 50 cm thick graphite blocks which are used in the same time as a reflector. These graphite blocks themselves are coated by a coal stone because of shielding and heat isolation purpose.

A longitudinal section of the reactor illustrates ist construction Fig. 1. The reactor core ends downwards in a funnelshaped bottom consisting also of graphite blocks. The cooling gas suplied from underneath the core by two circulators, flowing throug the core, passing the ceiling and the steam generator and turned again to the circulators. The cooling gas direction is shown in Fig 1 by arrows.

The fuel elements are given from above by five charging tubes to the core and leaving it through a central sphere discharge tube. This means a continuous refueling during full power operation is possible. Leaving the sphere discharge tube the fuel elements have to pass a control equipment. By the help of gamma spectrometric measures the burn-up is determined and the fuel elements will be discharged or recycled onto the core. This decision depends on the grade of burn-up which has been measured.

Through the arrangement of the charge tubes Fig. 2 , four tubes end at the reflector wall and one tube is directed to the centre. The outer part of the core and the inner region can be charged differently.

The horizontalshown in Fig 1 by arrows.

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The horizontal cross section in Fig 2 shows the arrangement of the charge tubes mentioned above and the control rods guided in four protuding graphite noses. The metalic rods with boron carbide as neutron absorber are inserted from the bottom of the core. They are used for the reactivity control during power operation.

Experiments in the AVR

During power operation of the AVR experimental reactor a lot of different fuel element types has been charged and their qualification investigated. Until the decision to change the fuel cycle the fuel consits of high uranium mixed with thorium except a little test charge of aboutb 3.000 elements with low enriched material.

At the time of changing seven fuel element types weren in operation with different heavy metal loading and particle design. In Tab 1 the main design data of the fuel element types are listed. All of them

thermal power L_{th} ——— live steam pipe

——— steam generator

——— inner reactor vessel

——— outer reactor vessel

He-pressure p ——— biological shield No.1

He-outlet temperature $\bar{T}_G$ ——— top reflector

——— thermal shield

——— shutdown rod nose

reflector temperatures ——— structural jacket

——— reactor sleeve

neutron flux $\phi(t)$ ——— reflector jacket

——— pebble bed core

——— bottom reflector

——— sphere discharge tube

——— main gas valve

He-inlet temperature $\bar{T}_E$ ——— circulator

He-pressure difference Δp
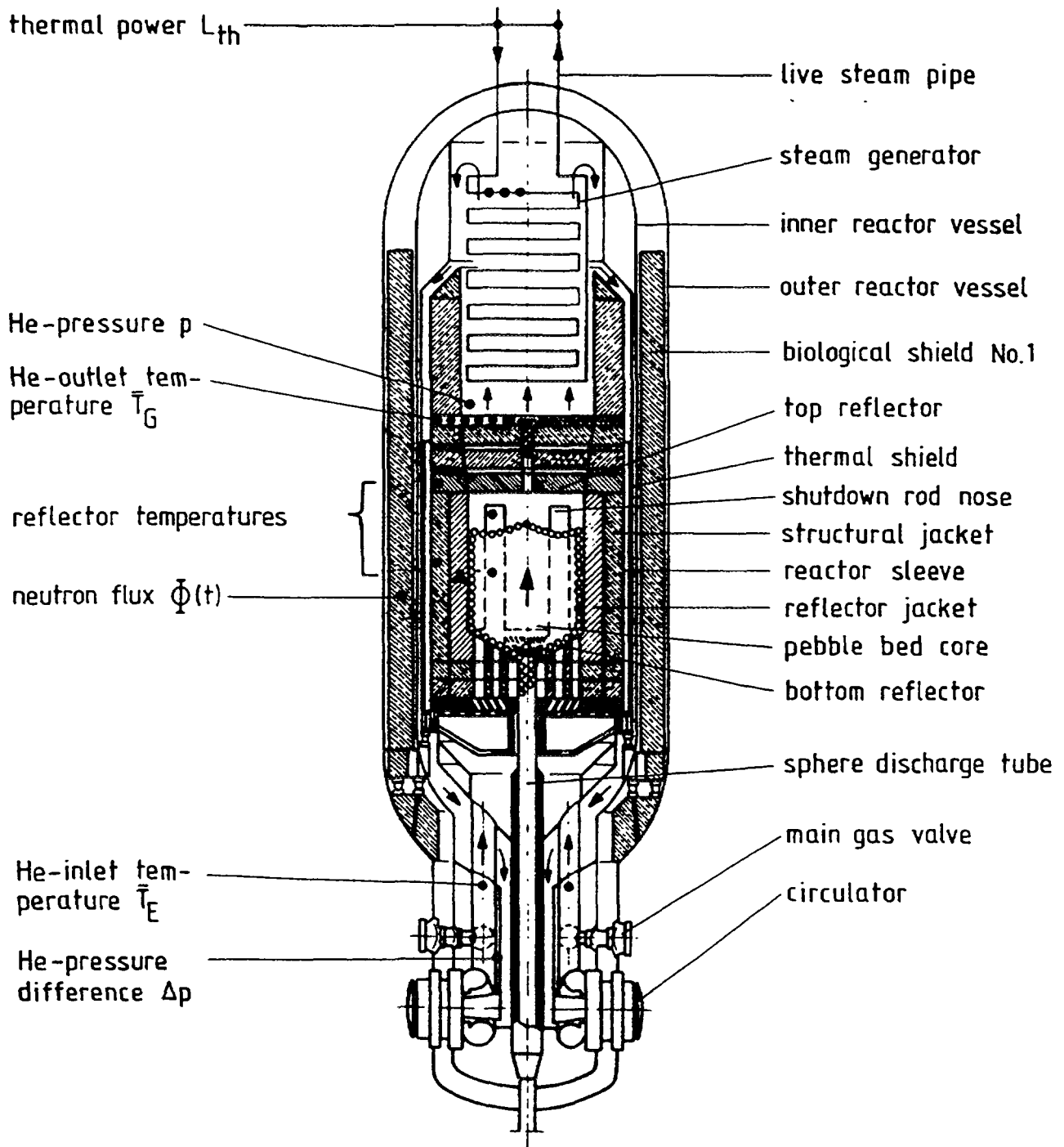
Fig 1 Longitudinal Cross Section of the AVR

except the test charge, mentioned above, contain 1g U235 as fissile material. The mixture of thorium however is in one case 5g and in the other 10g. The major amount of fuel elements in the core was the group with 6g heavy metal loading about 80 %. Beginning with loading the LEU fuel elements with the denotation GLE-3 (Tab. 1) it was possible to distinguish three groups of fuel elementt types by the help of the burn-up measurement equipment.

AS 4

0°

FR 2

FR 5

Outer Core

Thermal shield

R2520

Inner Core

R2960

R1000

AS 1

R1500

AS 3

Control rod insertion hole

AS 1

Graphite Reflector

FR 1

FR 4

180°

AS 2

Neutrons measuring column

central Fuel charge pipe

FR 3

Fig 2 Axial Cross Section of AVR

Tab 1 Fuel Element Types in the AVR

Brennelementtyp		GK	GO	GLE-1		GFB-1/2	
Anzahl der CP-Typen		1	1	2		2	
Protaktiniumklasse		2	2	1		3	
Brennstoff		$(U/Th)C_2$	$(U/Th)O_2$	UO_2	UO_2	UO_2	ThO_2
Anreicherung	%	93	93	15	U_{nat}	93	-
Kerndurchmesser	μm	400	400	600	600	200	600
Masse U_{235}	g	1	1	1,32	0,08	1	-
Masse Th_{232}	g	5	5	-	-	-	10
Masse U_{ges}	g	1,075	1,075	20		1,075	-

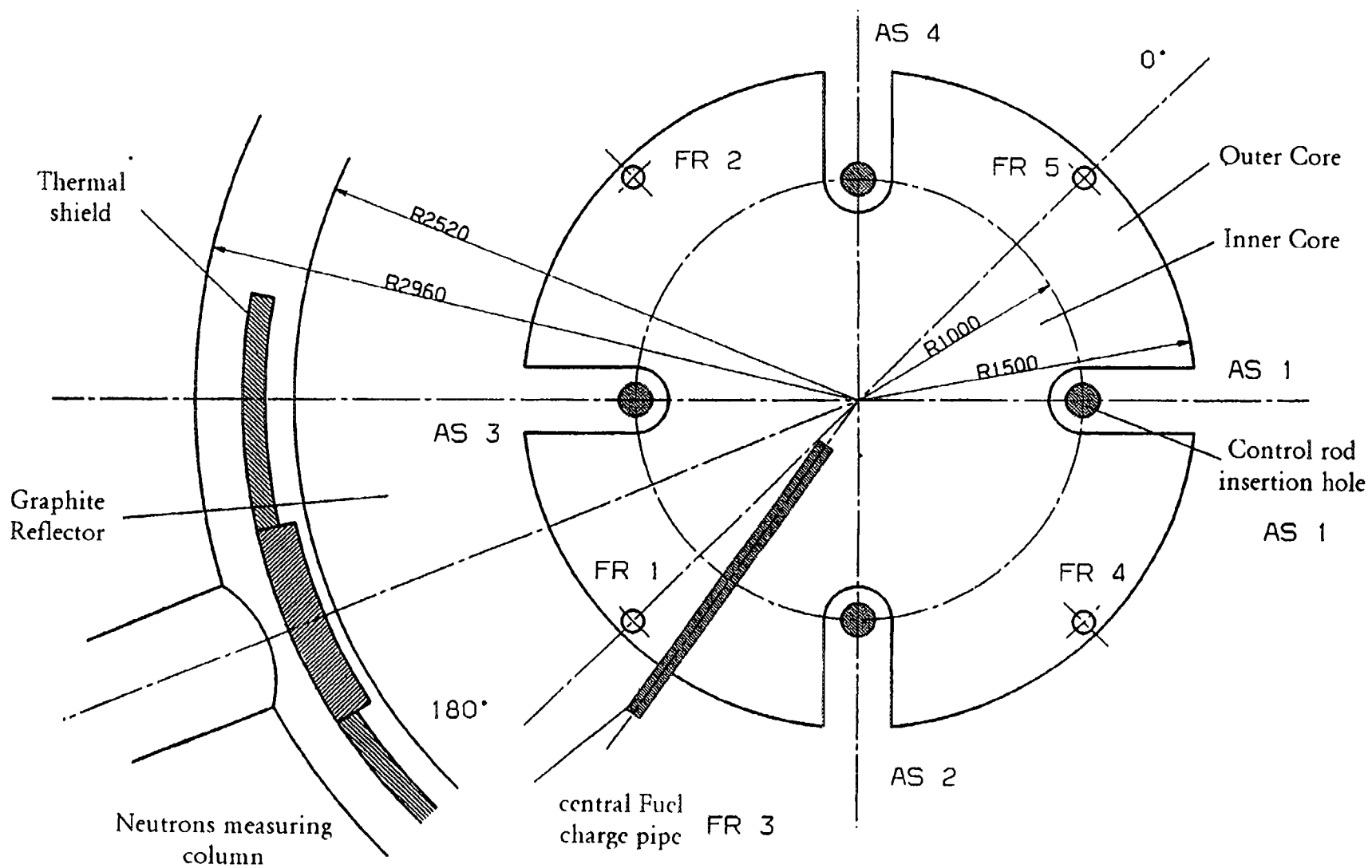| Brennelementtyp		GO(THTR)	GFB-3		GFB-4/5		GLE-3	GLE-4
Anzahl der CP-Typen		1	2		2		1	1
Protaktiniumklasse		3	2		2		1	1
Brennstoff		$(U/Th)O_2$	UO_2	ThO_2	UC_2	ThO_2	UO_2	UO_2
Anreicherung	%	93	93	-	93	-	10	16,7
Kerndurchmesser	μm	400	198	500	198	500	500	500
Masse U_{235}	g	0,96	1	-	1	-	1	1
Masse Th_{232}	g	10,2	-	5	-	5	-	-
Masse U_{ges}	g	1,033	1,075	-	1,075	-	10	16,7

Burn-up Measurement

The burn-up measurement deals in principle with gamma spectrometry. Guiding a fuel element after leaving the discharge tube to the measurement equipment a coherent spectrum of all photon emitting nuclides is detected. Through the intensity of some selected and qualified nuclides characteritical properties of a single fuel element can be detected by the help of the spectrum.

So the nuclide Cs137 with a half life time of 30 y is in combination with his daughter Ba137m a feasible indicator for the fission events in the fuel element. That means with the evaluation of the peak at the energy of 661 kev a measure of burn-up can be determined. This procedure is limited by very short decay time after irradiation. This difficulty is not important at the AVR, because the residence time for elements in the discharge tube is from 14 up to 29 days.

An other nuclide Pa233 with a half life time of 30 d and a peak at 300 kev is feasible to distinguish the elements with 5g and 10g Thorium load. This differentiation fails for long residence times in the discharge tube, for example after a shut down break.

A differentiation of fuel elements with Thorium and with not is not touched by this limit.
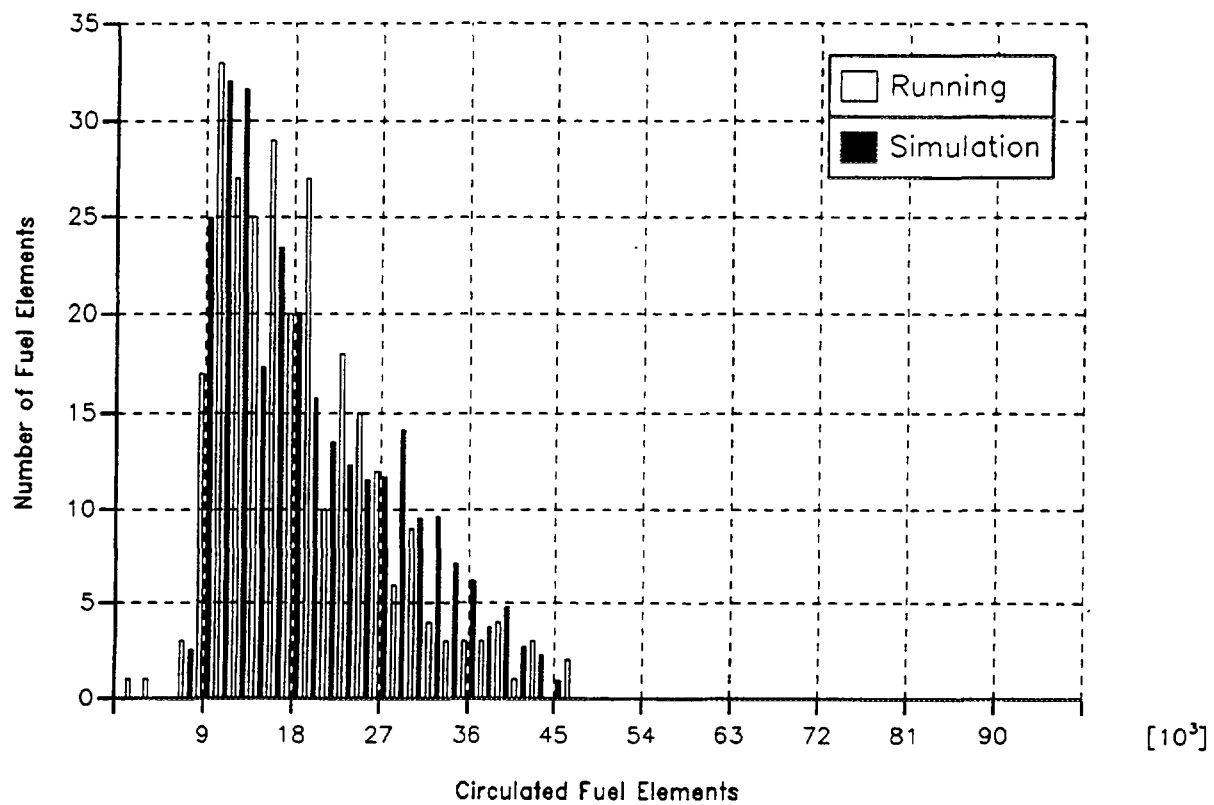
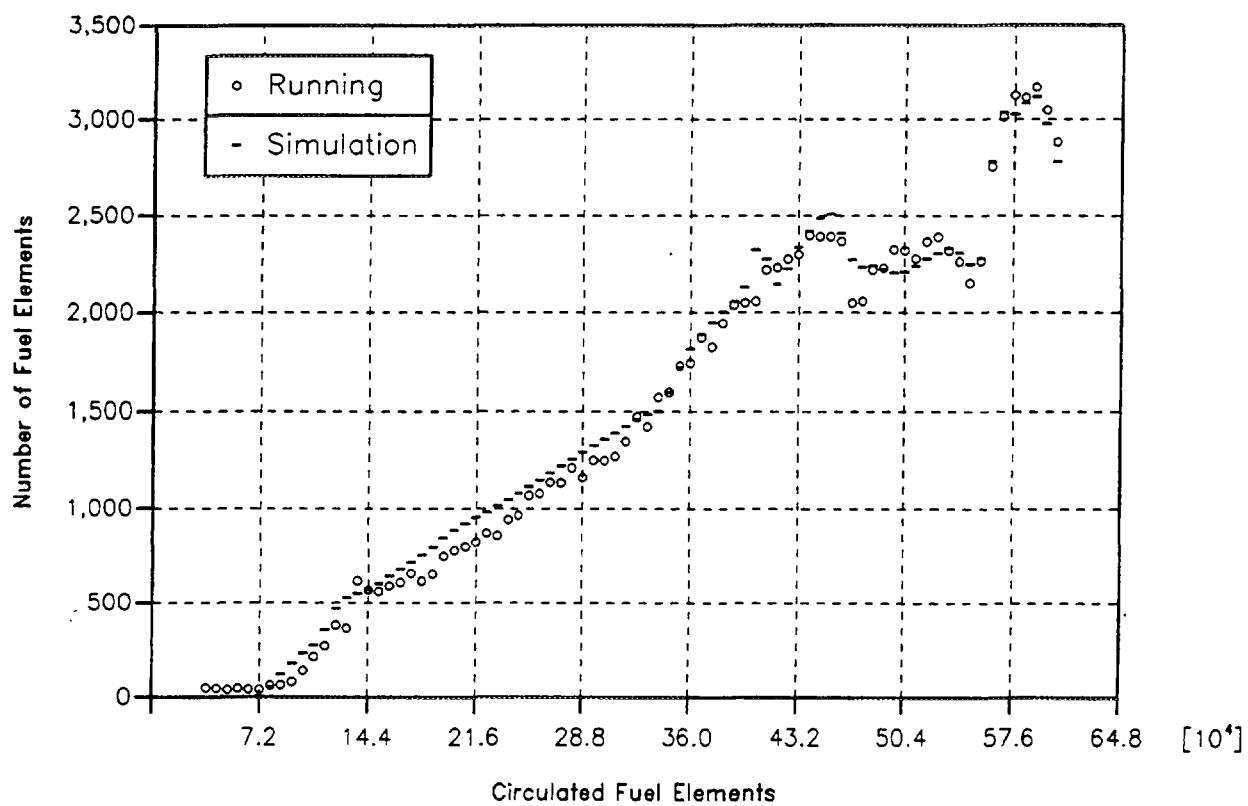Fig 3 Fuel Element Residence Spectrum; Core Inner Part

Fig 4 Fuel Element Residence Spectrum; Core Outer Part

Normaly it is possible to distinguish three element types regarding the amount of Protactinium. (Tab 1). A classification of al types into burn-up classes, regarding the amount of Ba137/Cs137 activity is feasible without any limitation.

By the help of these distinguishing earmarks experiments could be performed during full power operation to investigate the real fuel element moving through the core and after knowing the residence time to discharge fuel elements directly.

To perform this experiments it was necessary to choose the suitable fuel element type and the right moment of charging.

Flow Experiments

Flow Behaviour of the Pebble Bed

After a operation time of about three years since the beginning of the first LEU-FE (Low Enriched Uranium-Fuel Element) reload there was the following situation of core design.

Until this moment about 42.750 LEU elements (types GLE-3/GLE-4) were loaded. The total loading of the core consits of 45% LEU-FE about 55% HEU-FE. In this situation 300 HEU-FE (type GO Tab 1) were charged by the central charging tube onto the inner part of the core. After moving one time through the inner core they could be distinguished exactly from the other HEU-FE because of low burn-up and from the LEU-FE through the existing protactinium peak. The drawing in Fig. 3 shows the measured residence spectrum in white columns. This spectrum is the result of the flow characteristic of the fuel elements in the inner part of the core.

Because of directing all LEU-FE fresh and recycled only by the outer loading tubes, there was a possibility to get a result for the flow characteristic of the outer part of the core. Since all detected LEU-FE leaving the discharge tube were moving through the outer region. Fig. 4 shows the measurement points marked by circles.

The evaluation of these experiments gaves the full flow characteristic of the fuel elements in the core. Now it was possible to perform an experiment for feasible comparison between measured and calculated results, because of the exact knowledge of the flow behaviour.

Irradiation Experiment

After charging an amount of about 45.000 LEU-FE the reloading was continued by charging HEU-FE (type GO). In this situation after power operation of about three month 100 LEU-FE (type GLE-4) were charged onto the inner core.

In addition these fuel elements were marked by hand with a circle in order to identify these test elements after disloading in the hot cells by visual selection without any doubts.

By the help of this procedure five of these test elements could be identified after discharge and have been prepared for further analysis.

Now the boundary conditions for the calculation of the burn-up of heavy metal within the fuel elements were fixed. At the moment of the loading and the date of discharging the irradiation time was given. The power and the gas inlet and outlet temperature were given in the operation protocol. The other boundary conditions, like neutron flux and the fuel temperature have to take out of calculation by the reactor life program HTR2000.

Description of HTR2000

To describe the physical behaviour of the HTR core of a pebble bed typ it is necessary to simulate feasibly the pebble flow, the loading and discharging of the fuel elements and the controlling by the burn-up measurement equipment. The burn-up within the fuel elements has to be simulated. That needs a sufficient detailed calculation of the neutron flux and temperature. The interaction between different fuel element types has to take in consideration.

Rezyklierung von
bestrahlten
Brennelementen

Beschickung
frischer
Brennelemente

Beladestrategie, Kugelfließen
Netzgenerator, Transformation
(Flüsse+Nukliddichten)

Resonanzabsorption
für Brennelementgruppen
und zonengemittelt

Spektrumsberechnung
Wirkungsquerschnitte für
breite Energiegruppen

Abbrandrechnung

Abbrandmeßsimulation

thermische Leistung
Gaseintrittstemperatur
Gasaustrittstemperatur

Spektrumsberechnung (zeitgem. Daten)
Wirkungsquerschnitte für
breite Energiegruppen

Buckling-
iteration

3-D Diffusionsrechnung
Neutronenfluß, Leistung, k-eff
Temperaturen
(Brennel., Moderator, Gas)

HTROGEN: Schwermetallaufbau und
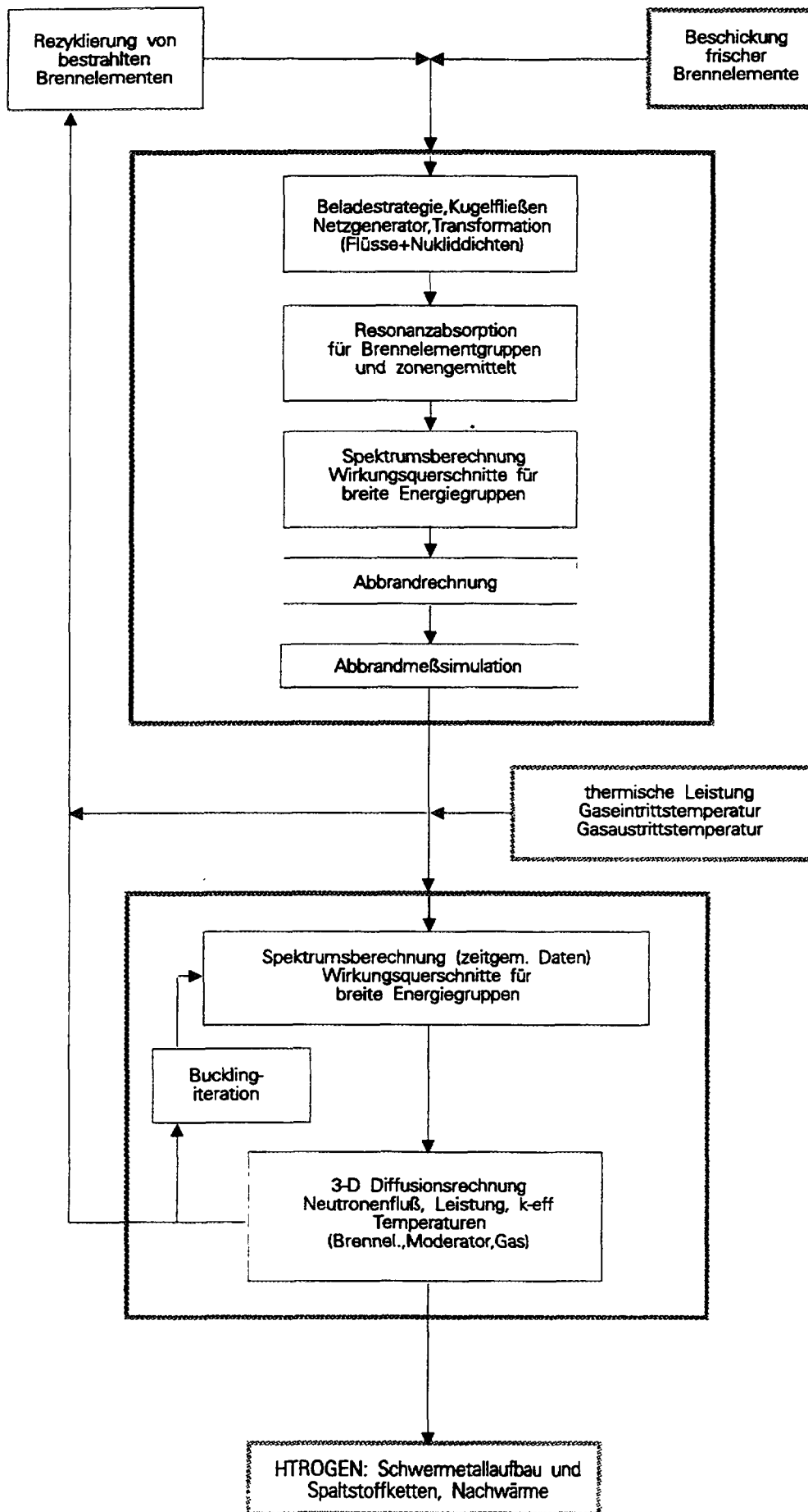Spaltstoffketten, Nachwärme
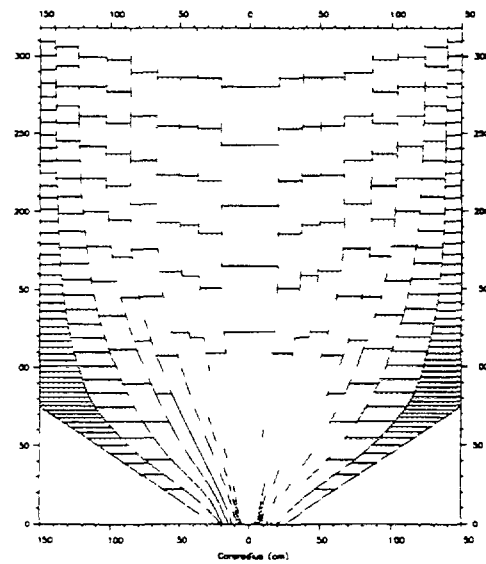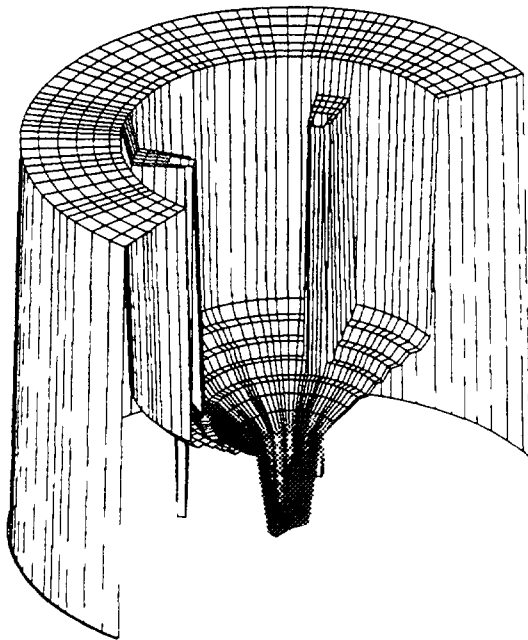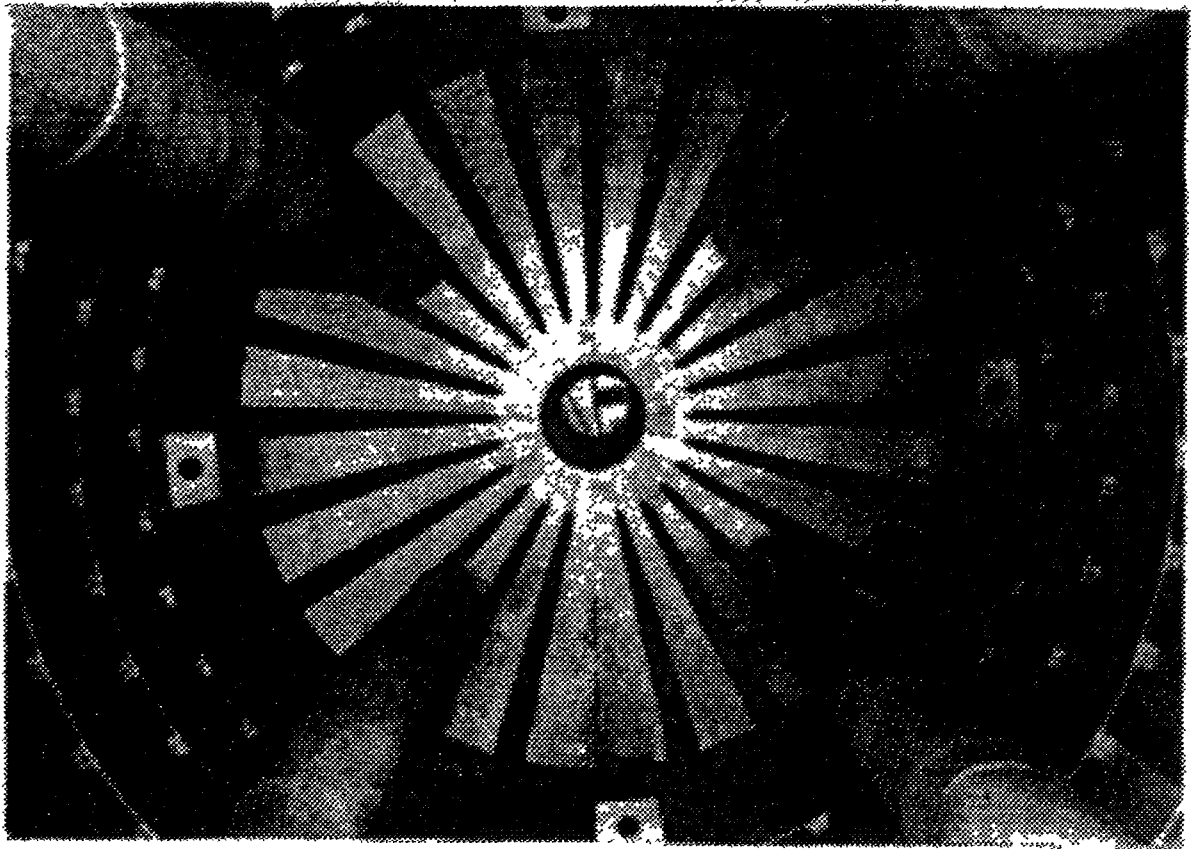
Fig 5 Flow Diagram of HTR2000

90

Fig 6 Real Geometry of AVR. Spatial Lattice. Flow Grid

Further on it is necessary to trace the geometry of the core and its structure. This is especially important for structures that are not rotational summmetrical like the AVR core.

In the program system HTR2000 [1] the points mentioned above are realized. The specific proportions at the AVR have been taken into consideration.

Fig 5 shows in a flow diagram the main function of the HTR2000 system within two blocks.

In the first block operation informations about the present fuel element loading, the power and power operation are used to simulate the moving of the fuel element and to calculate the bum-up.

By the help of the experiments mentioned above a model to describe the fuel element moving was gained. Simulating calculations were performed to evaluate the experiments. These calculations are based on a stream tube model. The pebble bed is treated at this theory as a slow flowing incompressible fluid. The fluid particles are moving along stream lines.

In Fig. 3 and 4 a comparison between simulation and measurement is depicted. The flow grid gained by the calculations is shown in Fig 6. Every volume depicted in the grid is a bum-up region with fuel elements different by type and bum-up. The influence of the different fuel with the resonance absorber (U238,Th232) is taken into consideration by a collision probability model [2] and a transport theoretical model [3]. The fuel element motion is simulated by shifting the volums in a stream tube step by step. The depicted grid is subdivided into 139 subregions.

The simulation of bum-up measurement transforms the information about the fuel elements detected by the measurement equipment into bum-up and protactinium classes. That gives a matrix of 50 bum-up and 3 protactinium classes tranformed to 9 fuel element types. The charge and discharge is simulated by removing the bottom regions and by filling the top regions.

In the second block the geometry of the reactor core is traced in a three dimensional lattice. In Fig 6 the chosen grid is compared with the real geometry. Within this lattice the neutron flux distribution is calculated by the use of a finite difference method [4]. The necessary macroscopic cross sections are calculated by transport theoretical methods as in part one.

The neutron flux distribution gives the power distribution that are needed for the temperature calculation. A buckling iteration takes into consideration the leckage out of the spectrum zones.

For normalisation the neutron flux the thermal power of the reactor has to be input. The gas inlet and outlet temperature is needed for the calculation of the temperature field.

These results are recycled into block 1.

To follow selected fuel elements moving through the core and calculate the bum-up history the HTR2000 system has been merged with the Isotope Generation and Depletion code ORIGEN [5] to a system named HTROGEN. So it is possible to solve the bum-up equation system for 1156 nuclides including such transmutations as (n,), (n,), (n, p), (n, 2n) and(n, 3n) processes. The code works with three neutron energy groups. The group constants have to be fitted to conserve the reaction rate in HTR2000. This is done by delivering the group cross sections and neutron fluxes from HTR2000 system to HTROGEN for each time step. So it is possible to select fuel elements and stream channels to caculate the bum-up history in detaile.

Chemical Analysis

As mentioned above five fuel elements of type GLE-4 have been discharged after moving through the reactore core one time. They have been identified in the hot cells and prepared for further analysis. The chemical analysis was performed in the Austian Research Centre Seibersdorf.

Characterising of the Unirradiated Material

The five investigated pebble fuel elements are descended from the 21 refueling charge of the AVR with the denotation AVR 21/GLE-4.

The fuel elements consits of A3-27 graphite. They have been pressed cold isostatically. The finish glow were performed by a temperature of 1950 C. The fuel enrichment of 16,76% U235 leads to an amount
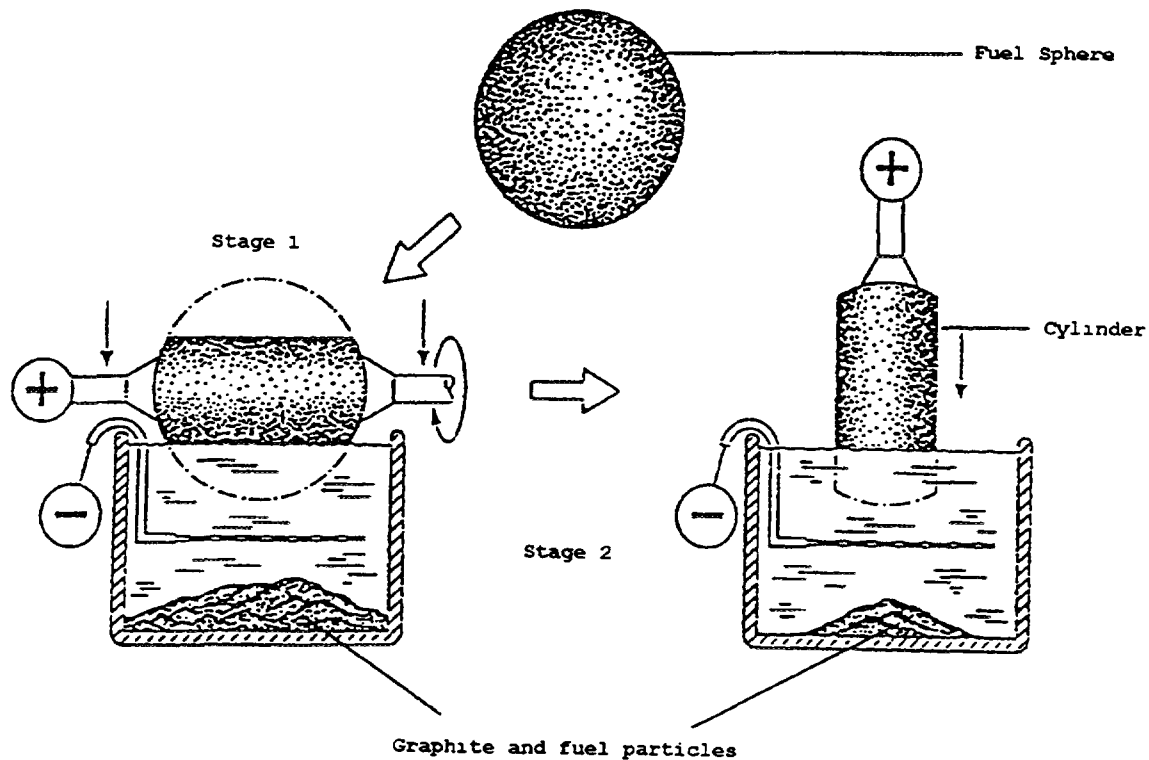
Fig 7 Desintegration of the Spheres

of 1g U235. That gives a sum of uranium of 6,0g. The composition of uranium isotopes is as follows: U234 0,14%, U235 16,67%, U236 0,09%, U238 83,10%.

The fuel element contains averaged over all 9.560 TRISO coated particles. The cernel consists of UO2 with a averaged diameter of 502 m. At first a buffer layer with a thickness of 96 m, followed by an inner pyro carbon layer of 41 m, the the SiC layer of 35 m and finished by an outer pyro carbon layer of 40 m leads to an averaged diameter of a particle of 926 m.

Desintegration of the Spheres

To analyse a particle out of the graphite matrix of a pebble fuel element chemically an electrolytical desintegration method has been developed.

Because of an expected gradient of fission product and heavy metal concentration within the pebble, a so called profile desintegration was performed. That means removing the fuel material layer by layer. In Fig 7 a principle sketch of such a procedure is shown.

The fuel element is taken apart in two steps. At first a cylinder is created by permanent turning in a solution of electrolytes (2n HNO3) and by an aniodic oxidation. The dissolved material is not needed further on. In the second step the cylinder is taken apart layer by layer. The particles out of each layer can be selected for the next analysis.

At Seibersdorf a more fashionable procedure has been developed. That is a profile drill along the pebble axis [6].

To eliminate disturbing influences by the contamination of the fuel free zone, this material is removed at each of the five testelements mechanically by a special constructed lathe. After this cleaning of the surfes the electrolytical profile desintegration has been performed in 18 steps. The fuel particles have been separated from the matrix graphite by sifting. All of the particles have been measured gamma spectrometrically and sorted by position. After that fuel particles out of the edge position, the middle and the centre position have been selected for the investigation.

93

Chemical Dissolution

Once more they have been measured gamma spectrometically and then they were separated in cemel and coating by mechanical cracking. Subsequently the cemels have been weighed by a microbalance. Five cemels have been sumed up to a batch, one batch for each position in the fuel element. Each batch was dissolved chemically in a mixture of Nitric acid and Hydroclorid acid quantitatively. Little individual differences of inventory are compensated because of this grouping. Then the dissolutions have been prepared for further chemical analysis.

Out of this parent solutions defined aliquots were removed by weighing. This weighing is essential more exactly then volumetric methods. A part of these aliquots were mixed with well known amounts of Uranium, Plutonium, Americium and Neodymium spike solutions.Losses of efficiency during later performed selection and weighing of samples can be neglected, because of refering to the added spike. That is a great advance of this method of isotpe dilution. Important is however the right estimation of the added amount of spike, in order to meet all interesting peaks by the mass spectrometric evaluation. By the help of freezing extraction Uranium and Plutonium were separated. In addition Neodymium was separated by ion exchange chromatographics. In the separated samples as well the frequency of the isotopes as the absolute amount of the isotopes were determined by mass spectrometry at the end of the chemical analysis.For Pu238 the results were in addition ensured by alpha spectrometric controlling measurements to avoid possible disturbances by U238 [7].

Burn-up Determination

To compare the measured data with calculated results the determination of the burn-up stage is necessary. The burn-up fima is defined as the rate of fissions occured during irradiation to the initial heavy metal loading. That means the fissions occured during the irradiation have to be evaluated. Because of the unability to measure this value directly only an indirect method has to be applied. By the help of feasible isotopes monitoring the fissions the burn-up can be determined. The Neodymium isotopes are interesting for the burn-up calculation based on mass spectrometric data. Well known as burn-up monitor is the Neodymium isotop Nd148, because of the neutron capture cross section is very low and the fission yield for Uranium and Plutonium fissions are nearly the same. The disturbing source out of the activation of Nd147 is neglectible.

For LEU fuel the knowledge of the Nd148 is sufficient to evaluate the total fission rate with sufficiant accuracy. Including the inventory of Nd150 the value is improved, because of the estimation of the fission fraction out of U235, Pu239 and Pu241. The sum of Nd143 and Nd144 or the sum of Nd145 and Nd146 seems to be more feasible.

To estimate the effective fissions neutron depended cross sections are used. For the iterative calculation it is necessary to choose the controlled variable to fit the calculations with the experimental data [7].

Discussion of the Results

In Tab. 2 the measured data are compared with the calculated results.

For the calculation it was assumed, that the test elements have been running in the third channel counted from the centre to edge (Fig 6). In this calculation the accordance between simulated and real loaded elements was the best. The power operation was simulated by HTR2000 and the single element was calculated by HTROGEN.

The measured data are averaged over all results gained from particles and have been extrapolated to an averaged fuel element.

Two measured data are listed for burn-up. The first has been obtained from the neodymium data measured mass spectrometrically. The second is an interpretation of the Cs137 activity by a function of Cs137 activity and fima gained by HTROGEN calculation. The difference of 2.5% is low.

The calculated value by simulation is 9% higher than the measured. This means the irradiation time in the model is to long. This effect is affirmed comparing the uranium data.

The comparison between measurement and calculation shown in Tab 2 is satisfactory. All differences are lower than 10%. Only for Pu238 the difference is 14%.

Tab 2 Comparison Between Measured and Calculated Data
of GLE-4 Elements

	Averaged - Measurement -	Standard-deviation - Measurement -	Calculation
Burnup (fima) Mass Spectrometry	3.48 %	± 0.03 %	3.8
Burnup (fima) Gamma Spectrometry	3.57 %	± 0.11 %	3.8
U-234 (mg/BE)	7.68	± 0.12	7.77
U-235 (mg/BE)	761.17	± 12.34	744.10
U-236 (mg/BE)	44.41	± 0.87	47.30
U-238 (mg/BE)	4,912.47	± 51.99	4,921.10
Pu-238 (mg/BE)	0.0184	± 0.0003	0.02081
Pu-239 (mg/BE)	18.5	± 0.4	17.79
Pu-240 (mg/BE)	4.04	± 0.05	3.909
Pu-241 (mg/BE)	0.73	± 0.02	0.657
Pu-242 (mg/BE)	0.056	± 0.001	0.0559

Further Investigation of Fuel Elements

Motivation Back Ground

The burn-up reached in this experiment of about 3,6% fima shown in Tab 2 is low in comparison with the aim of 15% to 18% fima in the reactor by power operation.

In order to get a better standart of comparison more investigations were carried out. In these experiments the restriction of low enriched uranium fuel has been canceled. The investigations were extended to fuel elements with high enriched uranium. Here the main question was the build-up of the plutonium isotope Pu238.

As shown in Fig 8 this isotope has two sources on one hand U235 on the other U238. In both pads there are a lot of steps to gain Pu238. Therfore the increasing of this nuclide concentration is slowly. But when the burn-up of fuel elements reached a high level, the amount of Pu238 becomes significant.

This process is especially important for the high enriched uranium cycle.

Predicting calculations by HTROGEN have shown, that the estimated values for expecting amount of plutonium, obtained from equilibrium calculations were to low.

The calculated values by HTROGEN have been higher than the double of the estimated data.

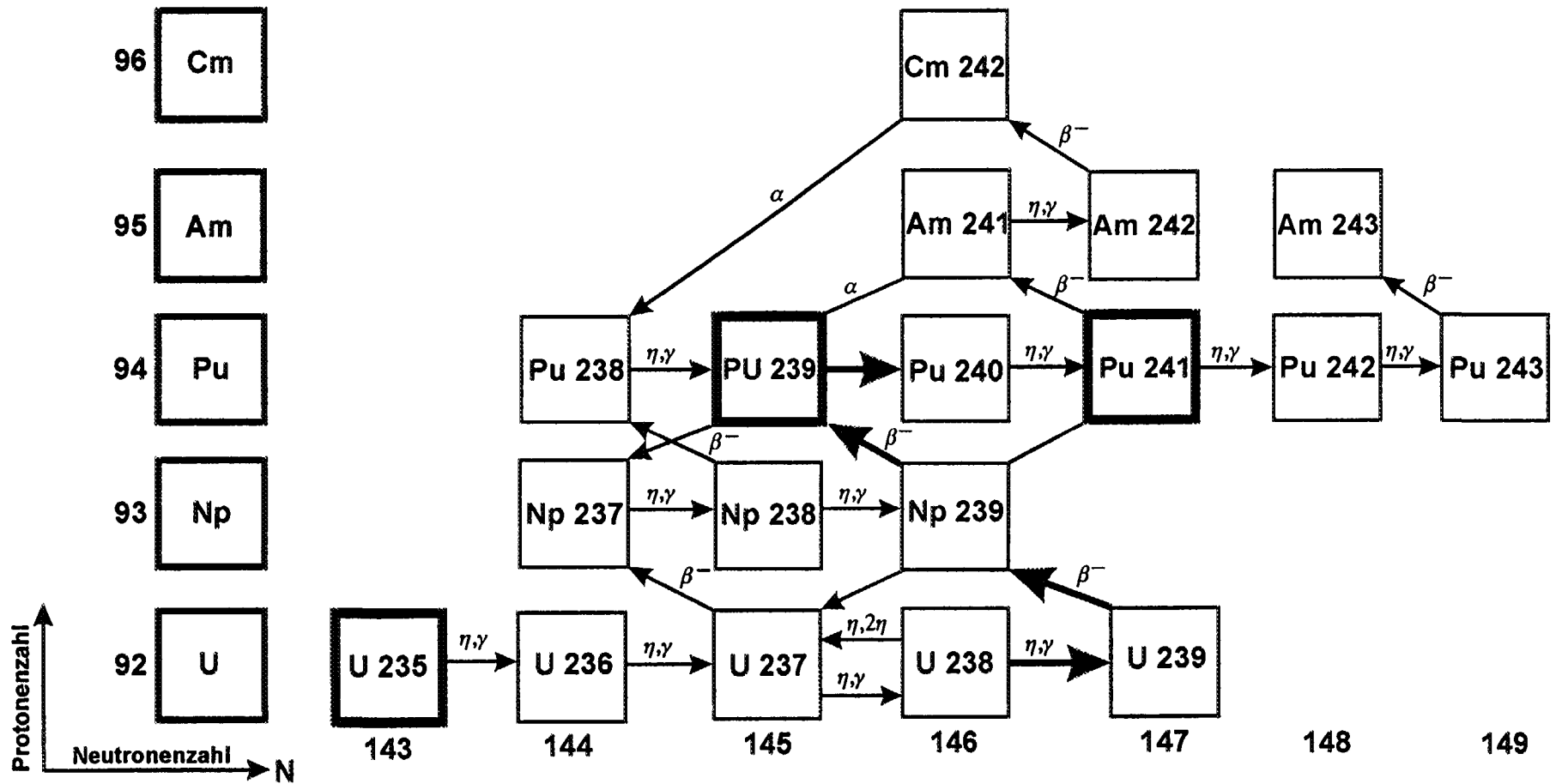This result is very sensitive for the declaration of the spent fuel.

Fig 8 Build-up of Plutonium and Other Trans Uranium Nuclides

To answer this question in addition six samples of the type GO were investigated by following institutes.
OEFZ Seibersdorf and Process Servis Division Harwell UK.
To ensure the results obtained from calculations for LEU fuel elements two samples of the GLE-4 type were investigated. It was the aim to gather a burn-up spectrum so wide as possible.

Calculation Strategy

The test elements should be of a high burn-up stage. Therefore the fuel elements have to be recycled several times. A flat assign between the life history moving through the core and the simulation were not possible. So an other strategy was applied for calculation.
The most probable way for a fuel element moving through the core during power operation has to be defined.
Normaly all of the fresh and low burned-up fuel elements were loaded by the outer loading tubes. The recycling by the outer tubes were finished at a minimum of burn-up. From that point the recycling were performed by the central tube. This loading scheme was simulated.
After moving along the outer channel the test element has been led to the inner part step by step. These calculation was performed at a defined operation situation of the reactor.

Discussion of Results

HEU Fuel

In Fig 9 - 20 the results of the simulation are drawn as solid line and the measurements as symbols. As characteristic for the HEU fuel the uranium isotopes U233, U234, U235 and the plutonium isotopes Pu238, Pu239, Pu241 have been chosen.
The concentration of the U233 in Fig 9 shows the influence of the way that has been chosen. Up to 10% fima, this is a time space of about two years, the test elements are moving in the outer part near the reflector.
The strong increase after moving through the core one time is caused by the decay of Pa233 in the disloading tube. So each cycle is shown definetly.
The measurements of both institutes meet the simulation quite good. The interpretation of U234 gaves some difficulties. In the simulation a value of 93% was assumed for uranium enrichment. That leads to a definit initial value of U234. The attachment of all measurements to calculated curve would give a too high starting value for the calculation.
The characteristic of the unirradated test material gives the answer. The first four samples , two of them were investigated at Seibersdorf and two at Harwell, consits of fuel with an enrichment of 92% but not 93% enrichment as assumed.
The comparison between the so calculated curves and the measurements gives a sufficient agreement.
The prognostic calculation resulted in a high expecting value of Pu238. These prediction is confirmed by the measurement impressively shown in Fig 12.
A strong increase is starting from a fima value of 12% as shown in the picture. The developement of Pu239 (Fig 13) is caused by the increasing of Pu238. Here also a strong increase is shown at 12% fima, because of the neutron capture in Pu238. However the birth rate is somewhat over estimated.
The curve of Pu241 (Fig. 14) is also verified [8], [9], [10].

LEU Fuel

For the LEU fuel the uranium isotopes U234, U235, U236 and the plutonium isotopes Pu238, Pu239, Pu241 has been selected as representative.

Fig 9-14 Measured and Calculated Data for GO Element

Brennelementtyp: GO
U-233
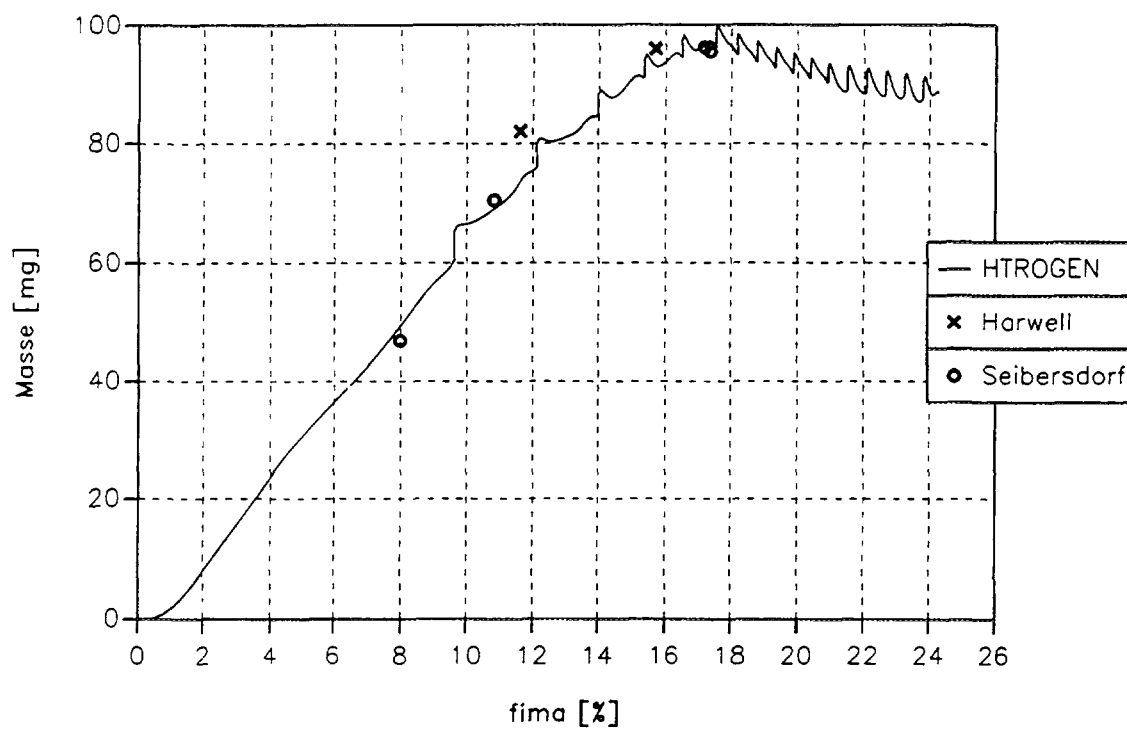
Fig 9

Brennelementtyp: GO
U-234
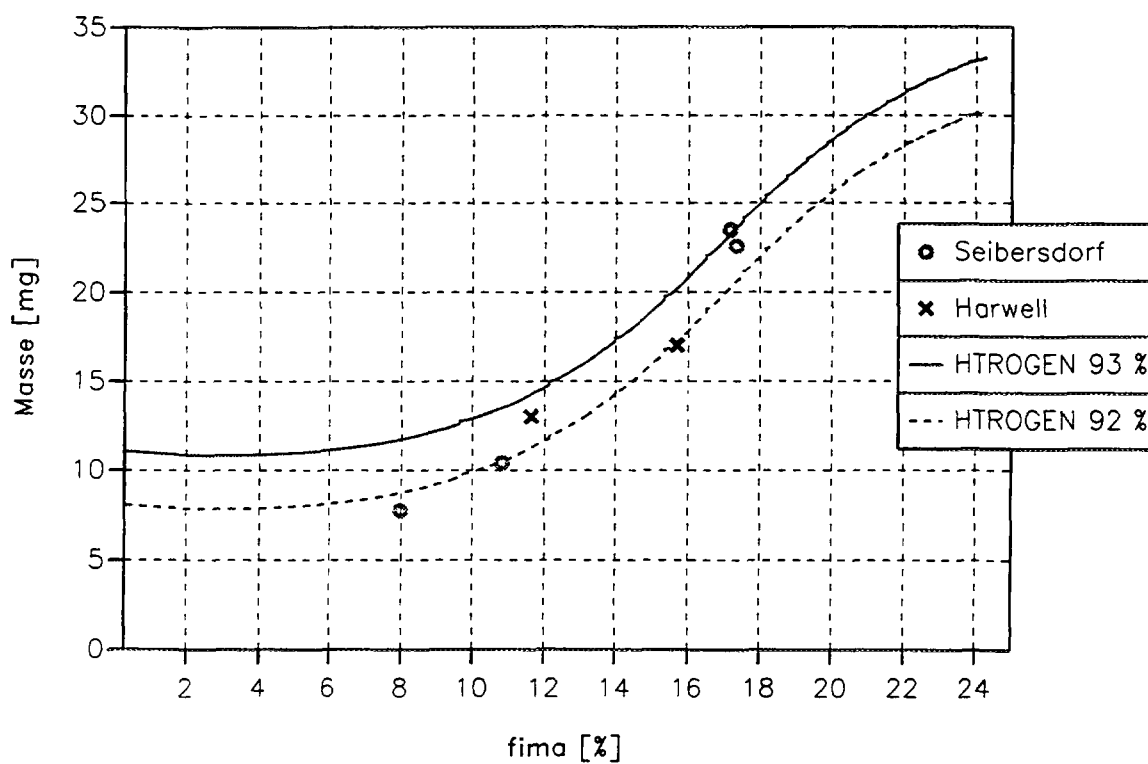
Fig 10

Brennelementtyp: GO
U-235
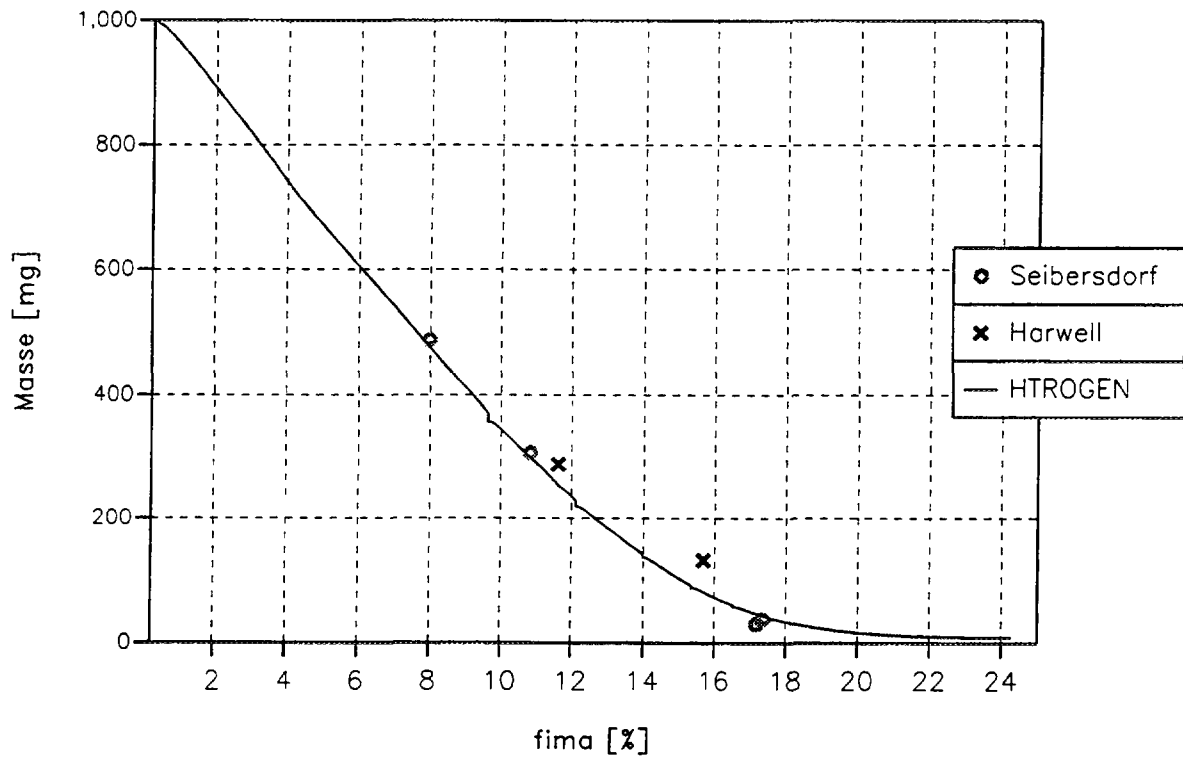
Fig 11

Brennelementtyp: GO
Pu-238
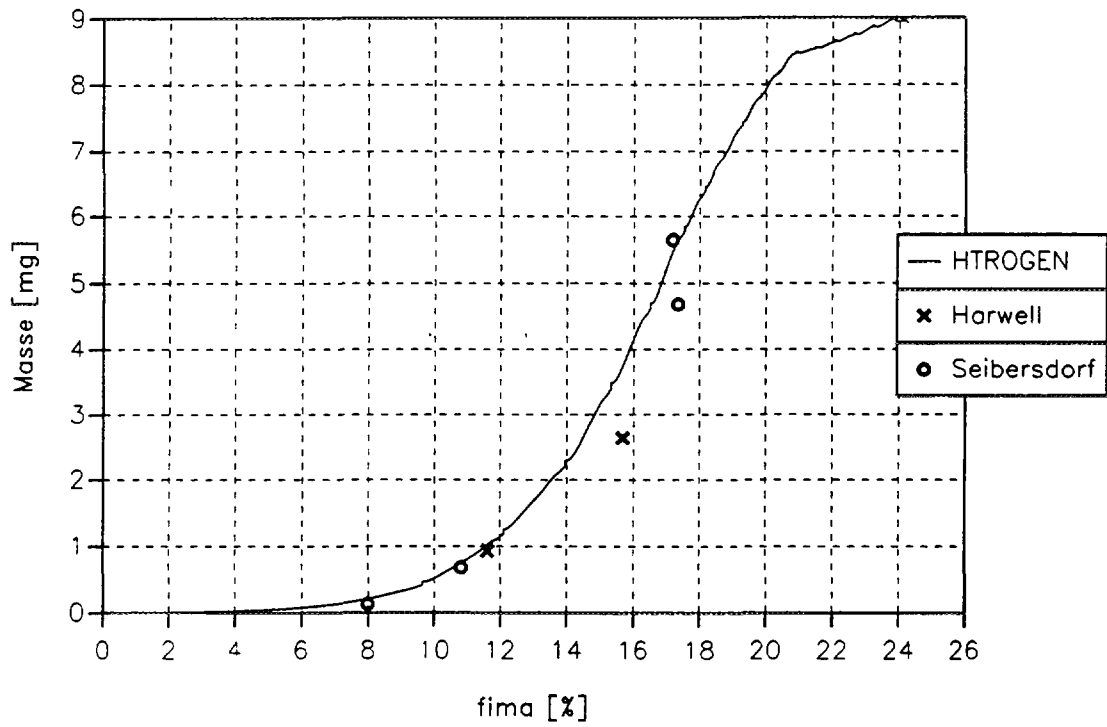
Fig 12

Brennelementtyp: GO
Pu-239

fima [%]
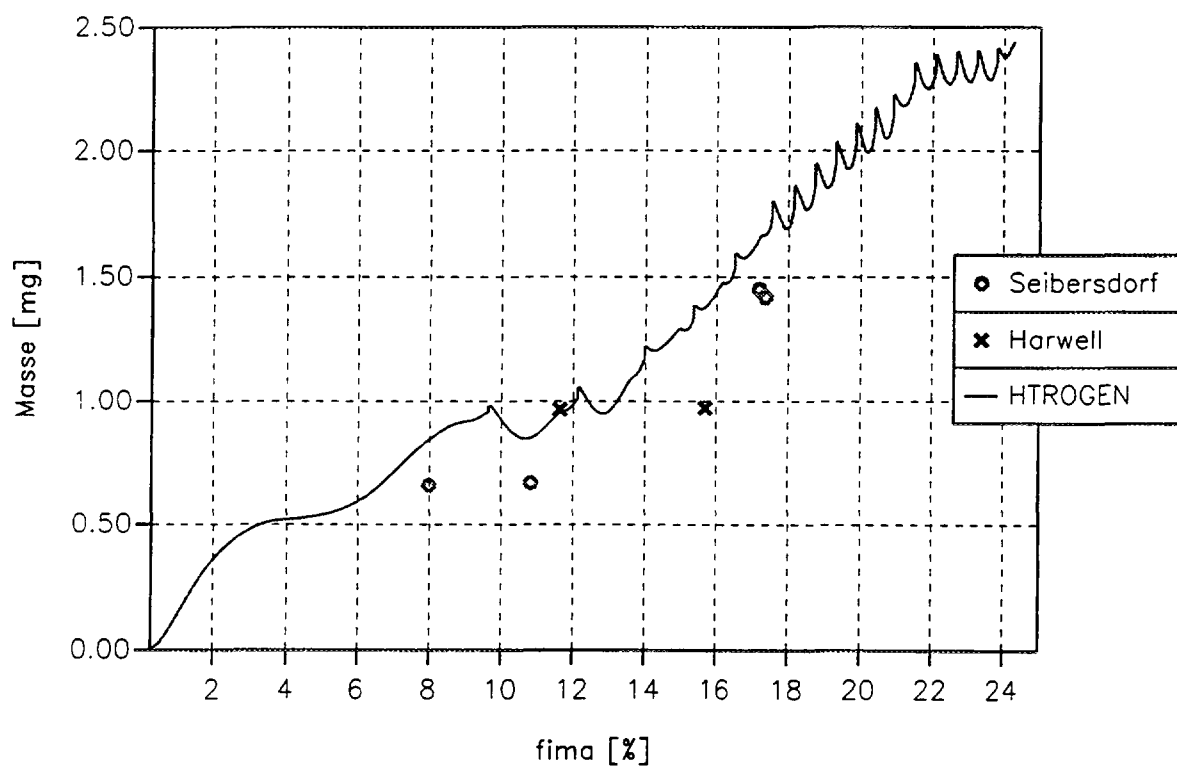
Fig 13

Brennelementtyp: GO
Pu-241

fima [%]

Fig 14

100

Fig 15-20 Measured and Calculated Data for GLE-4 Element

Brennelementtyp: GLE4
U-234

Fig 15

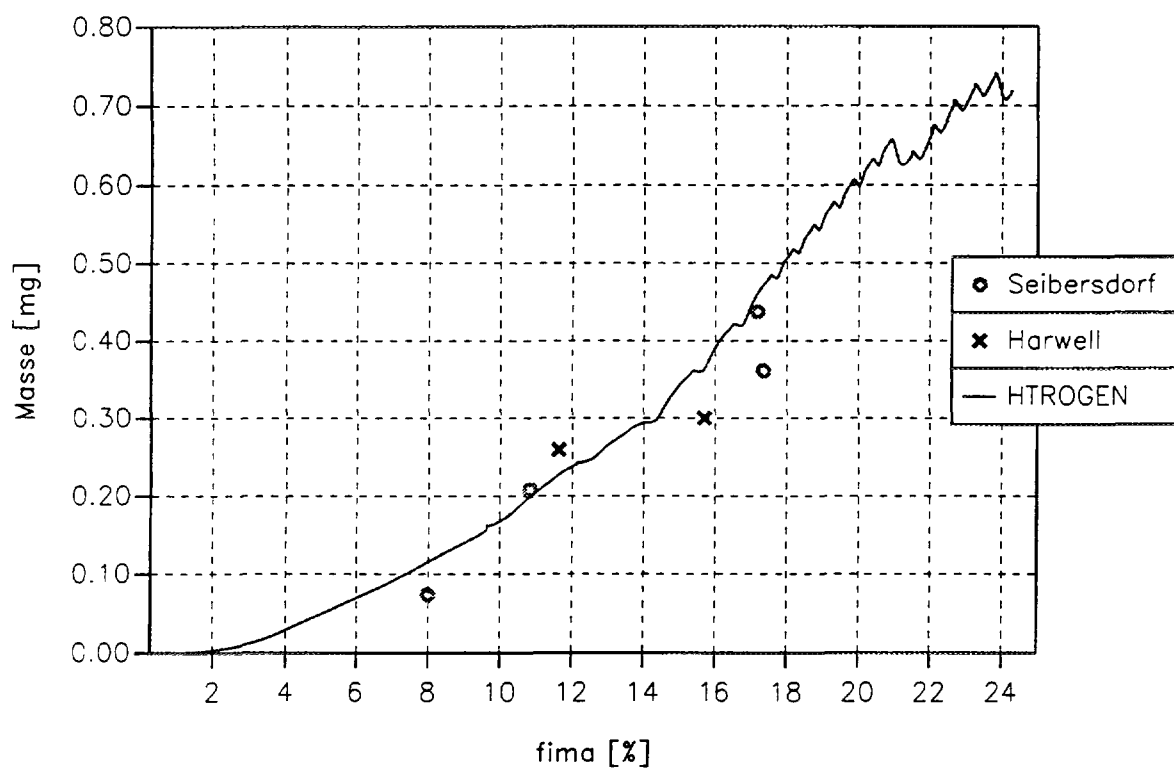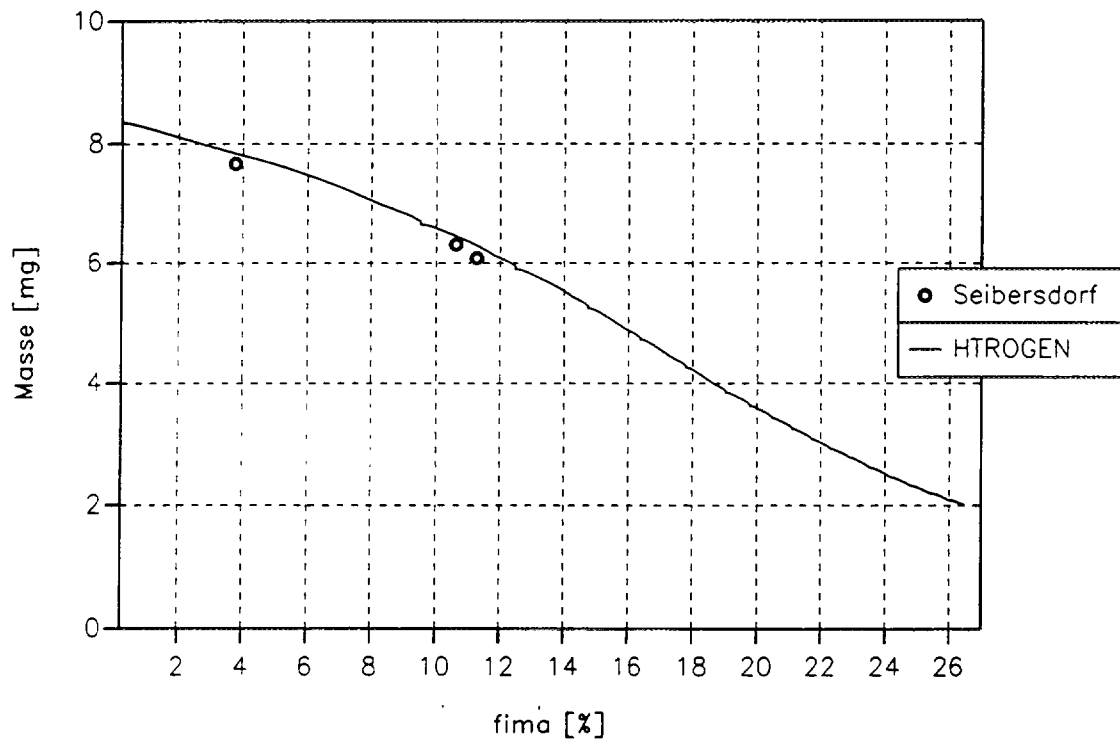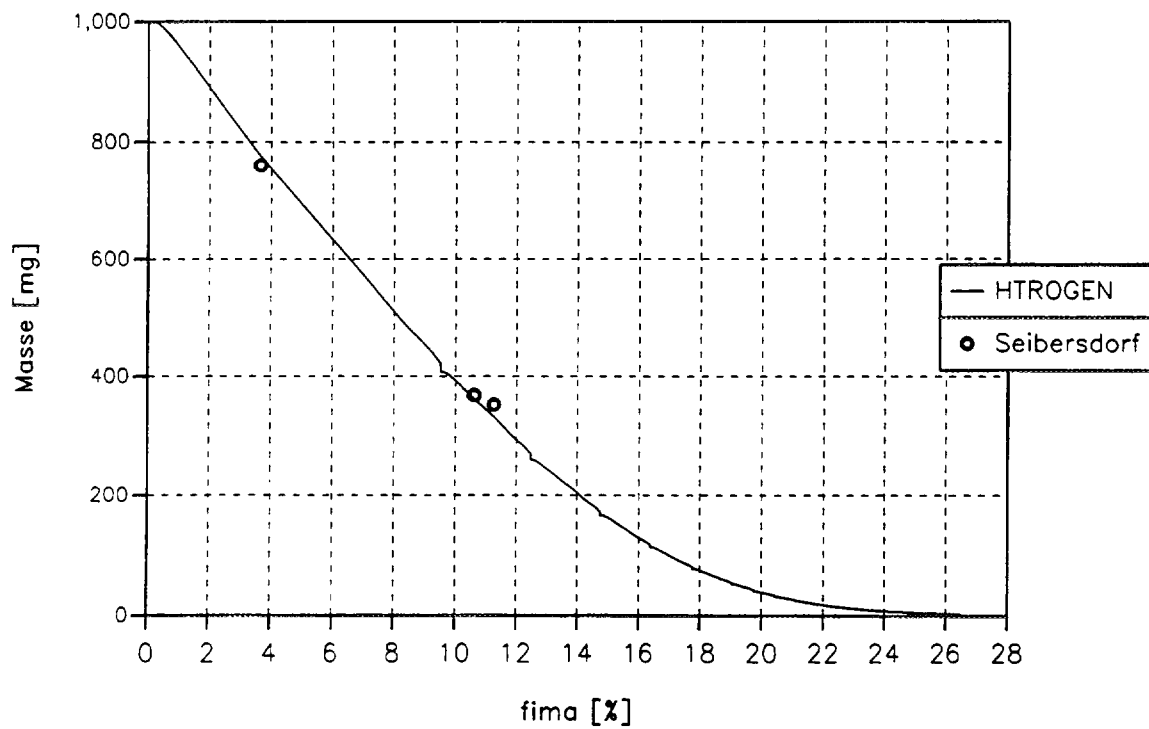## Brennelementtyp: GLE4
U-235

Fig 16

Brennelementtyp: GLE4
U-236
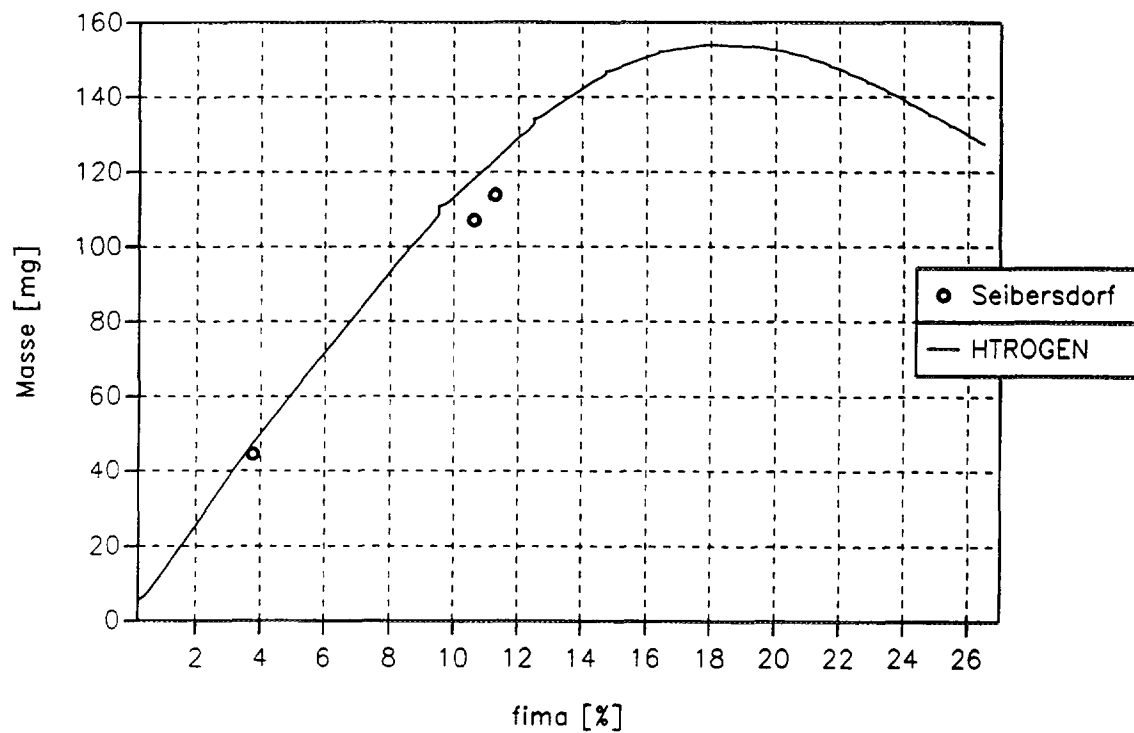
Fig 17

Brennelementtyp: GLE4
Pu-238

Fig 18

102
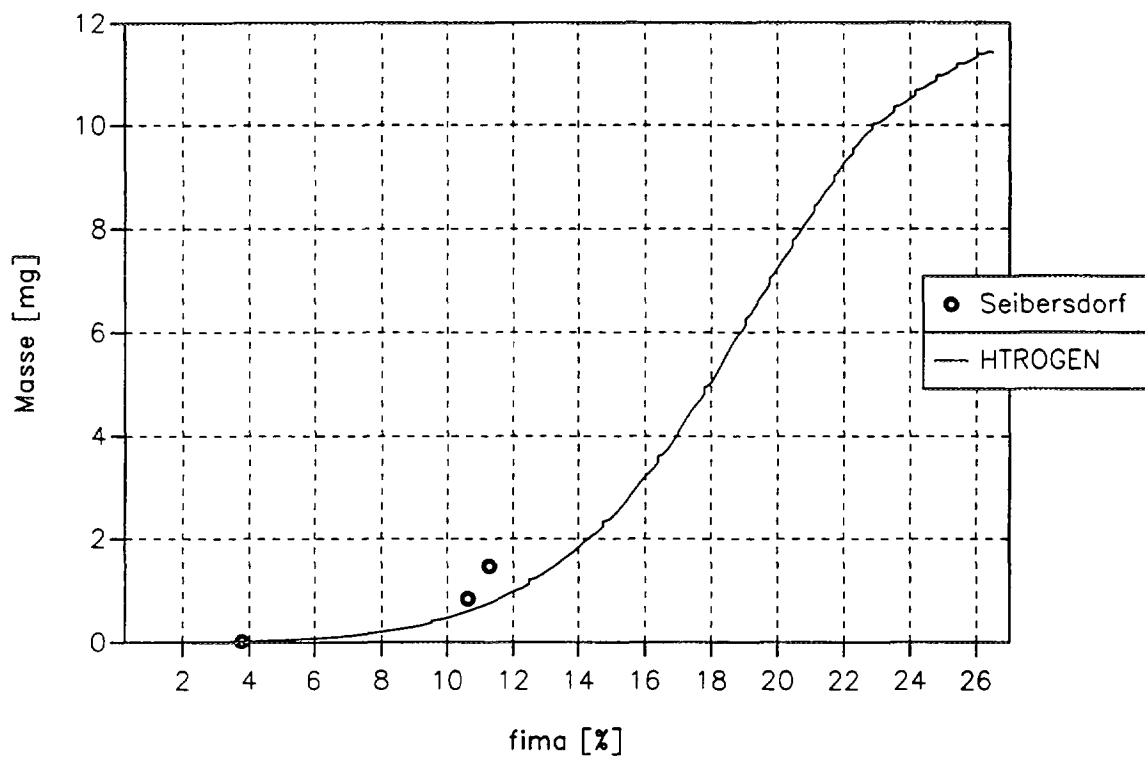
Brennelementtyp: GLE4
Pu-239
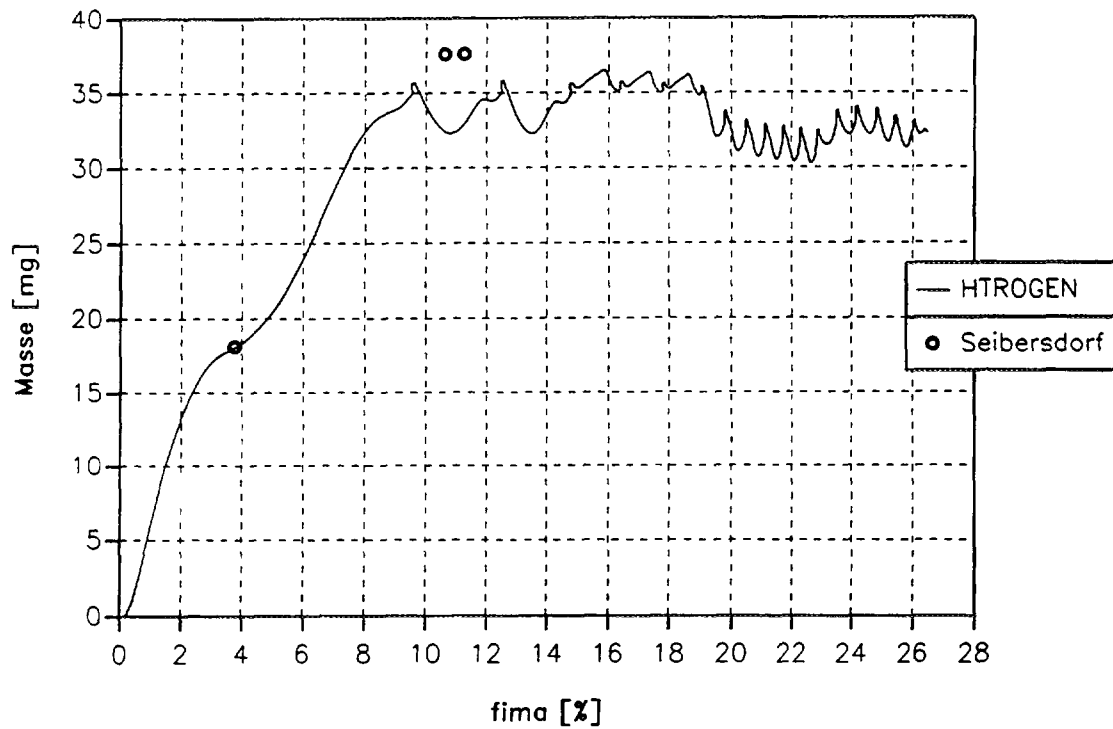
Fig 19

Brennelementtyp: GLE4
Pu-241
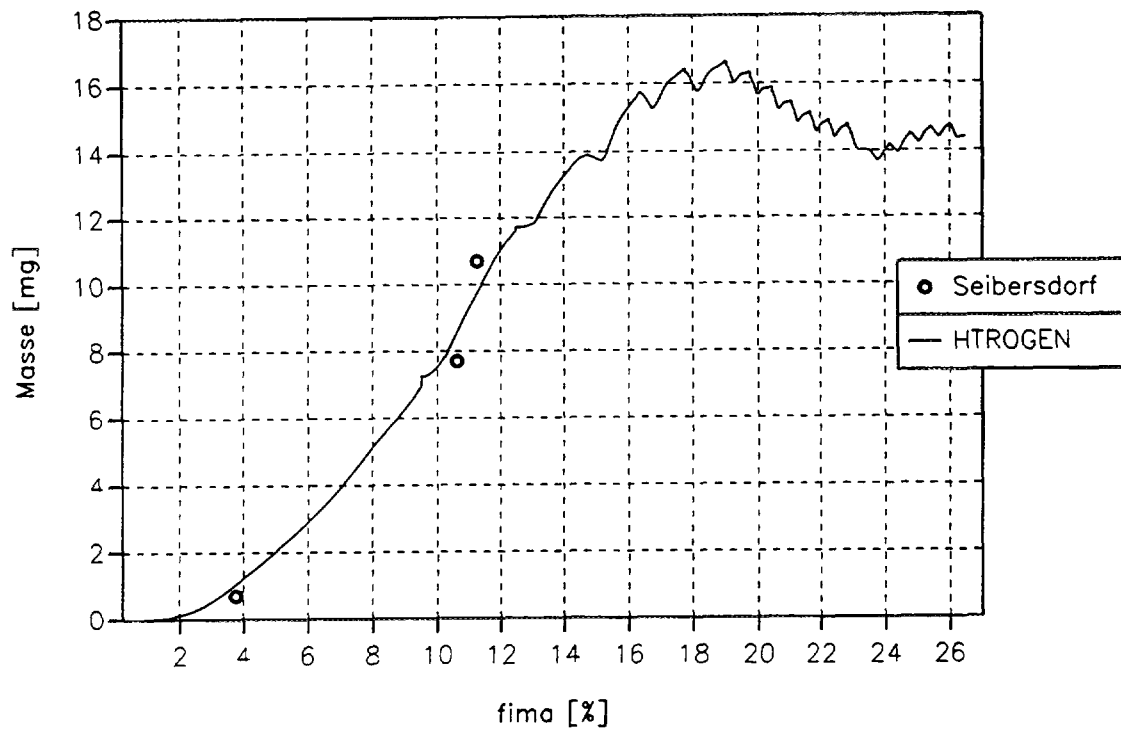
Fig 20

The curve of U234 (Fig 15) looks completly different as that gained from the HEU fuel. This is caused by the composition of the initial material. The fuel enrichment of about 16% has not been produced by the enrichment of natural uranium but by merging with reprocessed irradiated material.

Over all the comparison for uranium isotopes give a quite good agreement (Fig 15-17).

The development of the plutonium isotopes shows a somewhat under estimation at Pu238 and Pu239 at higher burn-up values (Fig 18 a. 19). That may be caused, that the test element were moving in the reactor some other than in the simulation.

However the development of the equilibrium stage fore Pu239 after moving through the core one time is confirmed as shown in Fig 19.

The development of Pu241 (Fig 20) is verified by the measurement points [10].

Finally a quite good agreement is shown by the comparison between the measured data and the results of calculations by HTR2000/HTROGEN.

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OEFZS-A-2749, November 1993

MOX FUELS FOR REDUCED ACTINIDE GENERATION

K. ANANTHARAMAN, P.D. KRISHNANI,
H.D. PURANDARE, D.S.C. PURUSHOTHAM
Bhabha Atomic Research Centre,
Trombay,
India

Abstract

The Indian Nuclear Power Programme at present is mainly based on Pressurised Heavy Water Reactors (PHWRs) with the exception of two Boiling Water Reactors (BWRs) at the Tarapur Atomic Power Station (TAPS), which was the first to start generating commercial nuclear power in India. Reprocessing and recycling of spent fuel and recycling of plutonium (Pu) has been adopted to improve utilisation of fissile materials and to shrink the spent fuel inventory. Plutonium utilisation has been contemplated mainly in our fast reactor programme. In addition, studies have been in progress to use Pu as MOX fuel and in combination with thorium (Th) in thermal reactors. This is in line with global trends to reduce actinide generation in reactors and for developing Pu fuels in combination with Th.

Development of UO_2-PuO_2 fuel was initiated in the `70s with the objective of establishing a fuel fabrication process flow-sheet, followed by verification of the process through experimental irradiations. Simultaneously, studies were undertaken on the physics end engineering aspects of utilisation of Pu in both PHWRs and BWRs. The successful irradiation testing of UO_2-PuO_2 rod clusters in a research reactor gave confidence in the design and fabrication procedures which in turn led to the setting up a fabrication plant for the manufacture of MOX fuel assemblies for our power reactors. Introduction of MOX fuel assemblies has been made in a BWR, with plans for progressively increasing the fraction of Pu in the core.

A study was also made for using MOX fuel in the PHWRs of 235 MWe size, using 19 rod fuel bundles. An option which has 7 central rods with 0.4% PuO_2 in MOX with the outer 12 rods being of nat.UO_2 has been selected for implementation. As the burnup is large in the MOX design, the production of Pu will be considerably less throughout the reactor life.

As India has abundant thorium reserves, fuel cycles utilising Th have been under consideration. Depleted uranium (dep.U) fuel bundles were used for initial flux flattening in our earlier PHWRs. In order to initiate thorium fuel cycles and to gain experience in its various aspects, ThO_2 fuel bundles were loaded in the recently built power reactors for initial flux flattening. It is planned to reprocess these bundles and recover uranium-233. Studies are under way on various fuel cycles involving Th.

This paper highlights the studies on the schemes mentioned above which facilitate reduction of actinide inventories and reduce their generation.

1. INTRODUCTION

The Indian Nuclear Power Programme at present is mainly based on Pressurised Heavy Water Reactors (PHWRs), except for two BWRs of the Tarapur Atomic Power Station (TAPS), the first nuclear power station in India to commence operation in 1969. The PHWRs use nat.UO_2 fuel and the latter LEU. India has adopted the philosophy of closed fuel cycle and recycling of plutonium (Pu) essentially for the fast reactor progaramme. In addition, work has been in progress in the area of utilisation of Pu as MOX in thermal reactors as an interim measure due to delay in the fast reactor programme. Also, as India has abundant reserves of thorium, work has been initiated on studies of fuel cycles using Th. These schemes are expected to reduce actinide generation in line with the global trends in this direction.

2. UTILISATION OF MOX IN BWRs

2.1 Physics Studies

The LWRs (Light Water Reactors) have low conversion ratio and are therefore ideally suited for 'burning'Pu. The two BWRs at TAPS use LEU as fuel. A study has been carried out to use MOX in these reactors, using a fuel assembly design containing an array of 6 x 6 rods, with one water rod in the spacer capture rod position. Figs. 1 (a) and (b) show the design of the LEU and MOX fuel assemblies. The MOX assembly incorporates fuel rods of 3 compositions of PUO_2-UO_2. The reactors employ 3-batch cycling of 18 months and the average burnup obtained for LEU is 20 GWd/ST. While designing the MOX fuel assembly, one of the major constraints was that there should be no change in hardware and that gadolinium (Gd) may not be used, unlike the LEU design. In order to limit the reactivity due to the absence of Gd, the discharge burn-up of MOX fuel assembly was reduced to 15 GWd/ST. In this study, the isotopic composition of Pu was assumed to be as follows:

$$Pu-239:Pu-240:Pu-241:Pu-242::0.7322:0.2182:0.0380:0.0104$$

The physics analysis has been carried out with the computer code LWRBOX using a 28 group WIMS cross section library developed to calculate lattice parameters as a function of burnup for rectangular assemblies of LWRs.

Table-1 gives some of the important physics characteristics for the two types of fuel assemblies, LEU (R-2) and MOX at hot operating conditions with 40% void. This table has 2 values

$L = 1.6 \% \, U^{235}$ $L = 0.9 \% \, Pu$

$M = 2.1 \% \, U^{235}$ $M = 1.55 \% \, Pu$

$H = 2.66 \% \, U^{235}$ $H = 3.25 \% \, Pu$

$\bigcirc = WATER\ ROD$
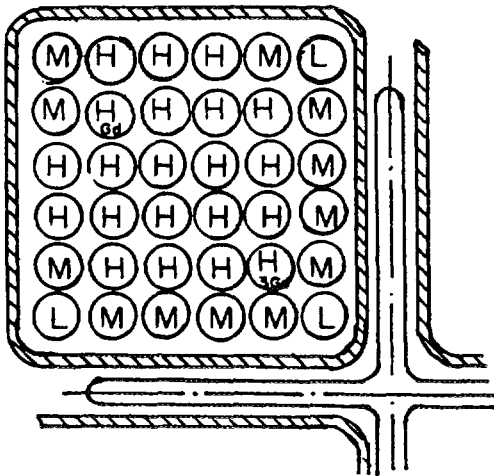
FIG. Ia - LEU ASSEMBLY
(R-2)
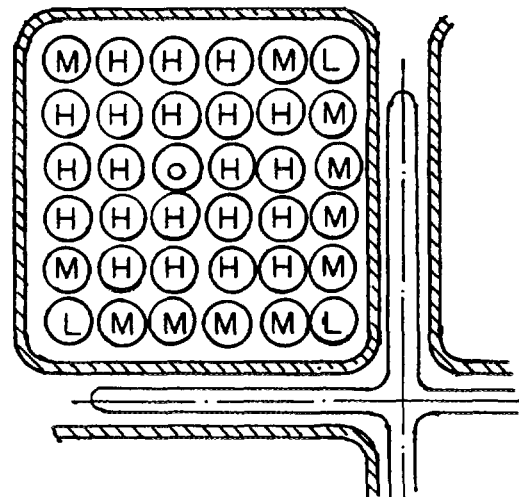
FIG. Ib - MOX ASSEMBLY

FIG. I - FUEL DESIGN FOR BWR^S

TABLE-1

VARIATION OF K_{inf}, LOCAL PEAK FACTOR, M^2, B_{eff}

AND LIFE TIME (L) WITH BURNUP
(HOT VOID - 40%)

Burnup	K_{inf}		LPF		$M^2 (cm)^2$		$B_{eff} \times 10^{-3}$		L(Sec) $\times E^{-4}$	
(Gwd/ST)	LEU	MOX	LEU	MOX	LEU	MOX	LEU	MOX	LEU	MOX
0.0	1.1259	1.2039	1.187	1.165	84.79	81.65	7.78	4.22	0.3466	0.2170
0.0*	1.0985	1.1795	1.189	1.172	84.68	81.60	7.79	4.24	0.3450	0.2157
1.0	1.1136	1.1589	1.166	1.171	84.61	81.64	7.46	4.26	0.3381	0.2204
3.0	1.1492	1.1314	1.157	1.174	84.57	81.74	6.96	4.30	0.3337	0.2294
5.0	1.1460	1.1096	1.141	1.177	84.50	81.84	6.62	4.33	0.3366	0.2380
7.5	1.1133	1.0842	1.124	1.171	84.42	81.99	6.30	4.37	0.3445	0.2498
10.0	1.0811	1.0605	1.107	1.161	84.39	82.13	6.04	4.40	0.3538	0.2618
12.5	1.0512	1.0386	1.090	1.155	84.41	82.27	5.83	4.43	0.3645	0.2724
15.0	1.0232	1.0171	1.074	1.138	84.46	82.42	5.65	4.45	0.3762	0.2847
17.5	0.9971	0.9964	1.062	1.120	84.53	82.57	5.50	4.48	0.3884	0.2971
20.0	0.9731	0.9767	1.060	1.110	84.60	82.73	5.34	4.49	0.4006	0.3097

* With Xenon

TABLE-2

WEIGHT FRACTION OF NUCLIDES

LEU FUEL ASSEMBLY

Burnup (GWd/ST)	U^{235}	U^{236}	U^{238}	Np-239	$\dfrac{U^{235}}{\text{Total U}}$	Total U
0.0	2.4021-2	0.0000-0	9.7598-1	0.0000-0	2.4021-2	1.0000-0
10.0	1.4233-2	1.7337-3	9.6777-1	4.0633-5	1.4468-2	9.8374-1
15.0	1.0829-2	2.3042-3	9.6335-1	4.3194-5	1.1090-2	9.7648-1
20.0	8.1196-3	2.7335-3	9.5869-1	4.5820-5	8.3747-3	9.6955-1

ALL MOX FUEL ASSEMBLY

Burnup	U^{235}	U^{236}	U^{238}	Np-239		Total U
0.0	6.9352-3	0.0000+0	9.6793-1	0.0000+0	7.1140-3	9.7487-1
0.0	4.8949-3	3.9498-4	9.6030-1	9.8215-5	5.0694-3	9.6558-1
15.0	4.0346-3	5.5053-4	9.5623-1	3.9969-5	4.1991-3	9.6062-1
20.0	3.2626-3	6.7899-4	9.5193-1	4.1779-5	3.4343-3	9.5589-1

TABLE-2 (Contd.)

WEIGHT FRACTION OF NUCLIDES

Burnup (Gwd/ST)	Pu^{239}	Pu^{240}	Pu^{241}	Pu^{242}	Am^{241}	$\frac{Pu^f}{Pu^t}$	Pu^t
			LEU FUEL ASSEMBLY				
0.0	0.0000-0	0.0000+0	0.0000+0	0.0000+0	0.0000+0	0.0000+0	0.0000+0
10.0	3.6067-3	8.1572-4	3.0193-4	4.6824-5	2.2756-7	8.1922-1	4.7711-3
15.0	4.2256-3	1.3340-3	5.5819-4	1.4079-4	6.9139-7	7.6436-1	6.2586-3
20.0	4.5422-3	1.8098-3	7.9053-4	2.8556-4	1.2915-6	7.1792-1	7.4290-3
			ALL MOX FUEL ASSEMBLY				
0.0	1.8424-2	5.4915-3	9.5674-4	2.6246-4	0.0000+0	7.7107-1	2.5135-2
10.1	1.3886-2	6.7236-3	2.2184-3	5.6476-4	4.2711-6	6.8163-1	2.2893-2
15.0	1.1531-2	6.9763-3	2.5616-3	8.1190-4	7.2913-6	6.4407-1	2.1881-2
20.0	1.0019-2	7.0530-3	2.7539-3	1.0951-3	9.7280-6	6.1053-1	2.0920-2

TABLE-3

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

FUEL COMPOSITION : UO_2 - 4% PuO_2

S.No.	Designation	Max.Linear Rating W/cm	Burnup MWd/Te	Remaraks
1.	AC-2	414	16,265	Standard
2.	AC-3	490	16,000	Standard
3.	AC-4	490	2,000	**

**Limited burnup planned.Design variables studied include fuel-clad gap,annular pellets,grain-size & cluster-size.

corresponding to zero burnup, the first without taking xenon into account and the second with xenon-135 and Rhodium-105 at equilibrium. The local peaking factor (LPF) for the MOX fuel assembly compares very well with the LEU fuel assembly. Thus, thermal margins are not affected in this design.

Table-2 gives weight fraction (WF) for various isotopes of U and Pu in the two types of fuel assemblies at selected burnups for 40% void case. These have been calculated by normalizing the weight fraction of metallic part to unity at zero burnup. Also given are the ratio of U^f to total U, total U wf, ratio of Pu^f to total Pu and total Pu w.f. The discharge burnups of MOX and LEU fuel assemblies are expected to be about 15 and 20 GWd/ST respectively. In the case of LEU fuel, 7.4 Kg of Pu is produced per initial tonne of U with 61.1 wt% Pu-239, 24.4 wt% Pu-240, 10.6 wt% Pu-241 and 3.8 wt% Pu-242. In the case of All-MOX fuel, Pu is consumed to the extent of 3.2 Kg per tonne of heavy elements present initially. The spent fuel will have low quality Pu with 52.7 wt % Pu-239, 31.9 wt% Pu-240, 11.7 wt% Pu-241 and 3.7 wt% Pu-242.

2.2 Irradiation Studies and Manufacture of MOX Fuel:

In the early '80s, a set of experimental irradiations of MOX fuel rods were taken up in the research reactor CIRUS, as part of the programme on the development of MOX fuel for power reactors. A summary of the irradiation experiments is given in Table-3. Based on these successful tests, a flow-sheet was developed for the MOX fuel fabrication. Subsequently, a plant has been built at Tarapur where currently MOX fuel manufacturing operations are in progress. The plant has two parallel fabrication lines. Initially, one of the lines has been used to fabricate two MOX fuel assemblies of the All Pu Design which are now undergoing irradiation in a BWR of TAPS. These assemblies have seen exposure of about 5 GWd/T so far. Further work is on hand with the objective of progressively increasing the number of MOX fuel assemblies in the BWRs within the permissible loading limits.

3.   UTILISATION OF MOX IN PHWRs

The PHWRs are the mainstay of the Indian nuclear power programme. Pu produced in them is meant to be used to fuel the fast breeder reactors in the second phase. However, due to slower progress of fast reactor programme, utilisation of Pu as MOX fuel in a recycle mode is contemplated for the interim. A study has been made for the use of MOX fuel in the 220 MWe reactors with minimum deviation in the basic design of the fuel bundle, the reactor hardware, the control system etc.

The PHWR has a horizontal calandria, penetrated by a large number of fuel channels arranged in a square lattice pitch of 22.86 cm. The fuel channels consist of a calandria tube and an inner pressure tube through which the fuel bundles are moved by the on-power fuelling machine. The pressurized hot heavy water coolant flows through the presssure tube. Each pressure tube contains 12 fuel clusters of 19 rod design, of which only 10 lie in the core portion. The cool moderator is contained within the low pressure calandria. The control and shutoff devices are vertical and contained within the low pressure moderator. The earlier PHWRs employed moderator dumping as the shutdown system compared to the two-shutdown systems in the standard 220 MWe PHWR.

The fuelling scheme is on-power bi-directional multibundle shift scheme. For the nat.U fuelled reactor, 8 bundle shift has been chosen. The core is divided into 2 to 3 radial zones to achieve radial power flattening in equilibrium condition, and in each zone a discharge burn-up is so chosen that the design total power is obtained and the limits on the maximum channel power and bundle power are satisfied.

3.2 Design of MOX Fuel Bundle.

For the introduction of MOX in the PHWRs, the limits on the power for a bundle and for each of the coolant channels are already fixed by the present nat.U fuel design. Likewise, the locations and the number of control and safety devices and the capability of the fuel handling system are fixed. This would mean that the new fuel configuration has to be so optimised that the ensuing local and global power distributions are very close to the existing one.

It was decided that integral KdO max. of 40 W/cm, as in the case of nat.UO_2 fuel, would be the limit maintained for the MOX fuel as well. After analysing different cases, a MOX-7 design with
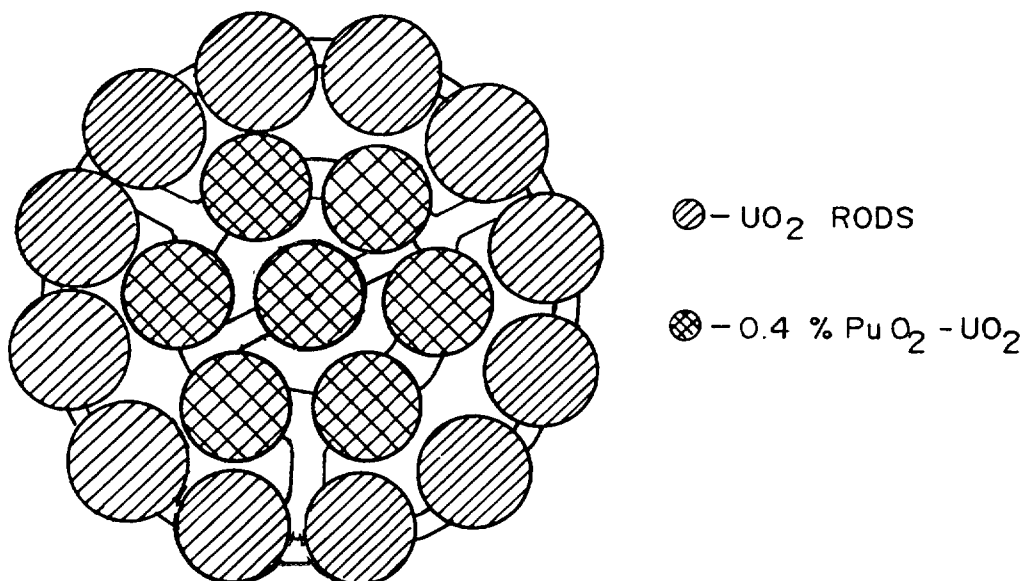

$\oslash-$ UO_2 RODS

$\otimes$ -0.4 % Pu O_2 $-UO_2$

FIG. 2 - MOX FUEL BUNDLE FOR PHWR

TABLE-4

DESIGN DATA FOR INDIAN PHWR OF 220 MWe

ELECTRIC POWER (MWe)	:	220
THERMAL POWER TO COOLANT (MWth)	:	756
CALANDRIA VESSEL DIAMETER (cm)	:	600
No. OF FUEL CHANNELS	:	306
LATTICE PITCH (cm)	:	22.86
No. OF BUNDLES PER CHANNEL	:	12
MAXIMUM CHANNEL POWER (MW)	:	3.2
No. OF ADJUSTER RODS	:	6
No. OF SHIM RODS	:	2
No. OF SHUT OFF RODS IN THE PRIMARY SHUT DOWN SYSTEM	:	14
No. OF VERTICAL LIQUID POISON TUBES IN THE SECONDARY SHUT-DOWN SYSTEM	:	12

central 7 rods having 0.4 wt% PuO_2 in nat.UO_2 and the outer 12 rods of nat. UO_2 as shown in Fig.2 was selected for further studies and implementation. Based on power distribution within the fuel bundle, the bundle power limit of MOX-7 turned out to be 384 KW, compared to 420 KW for nat.U fuel bundle. A fuelling scheme utilising nat.U for the central 44 channels of the reactor, and MOX for the remaining 262 channels, was developed to avoid reduction of reactor power. With this kind of zoning, maximum bundle power is seen only in the nat.U bundles, and is within acceptable limits. Besides, this results in an increase of average burnup from 7 to 10.7 Wd/Te.

3.3   Isotopic Composition

Table-4 gives the isotopic composition in terms of g/Kg of initial heavy isotopes at zero burnup and at exit burnup for the two types of fuel bundles. The net production of Pu in the MOX-7 design is 3.6 Kg per tonne of initial heavy isotopes compared to 3.7 Kg per tonne of initial U in the nat.U case. As the exit burnup is large in the case of the MOX design, the production of Pu throughout the reactor life will be considerably less. The quality of Pu in the MOX fuel is slightly poorer. The fissile Pu content for MOX-7 is 64.3 wt% compared to 75.5 wt% for nat.U fuel.

4.   THORIUM UTILISATION SCHEMES

A judicious use of our resources involves not only production of power but also conversion of good part of the fertile material - Th into fissile material. The Th utilisation is a 2 stage process - 1) conversion of Th into U^{233} and 2) using this in-situ or after reprocessing and fabrication. In this section, the work carried out on Th is discussed.

4.1   ThO₂ in PHWRs

The PHWRs use nat.UO_2 in all the channels. During reactor start-up at initial fuel loading, such a scheme does not have the necessary power flattening and the reactor can achieve only 70% full power during the initial stages. In order to reduce the power peaking in the centre of the core, the reactor is initially loaded with dep.UO_2 bundles in the centre of the core. As an alternative, ThO_2 bundles can also be used for power flattening. This scheme represents a unique way for production of U^{233} without any penalty in the fuel burnup of nat.U fuel.

The Th fuel bundle design was carried out in such a way that it is similar to the U fuel design to a large extent, so that fuel handling and hydraulic matching with nat.U bundles are taken care of. 4 such bundles were loaded in Madras Atomic Power Station Reactor to study their reactor worthiness. Based on this experience, Th bundles have been used in Kakrapar Atomic Power Station for initial flux flattening and a similar scheme is planned for our future PHWRs. The Th bundles have seen a maximum power of 315 KW and burnup of 4 GWd/Te.

4.2   Programme on ThO₂-PuO₂ Fuel:

In the PHWR, an R&D programme has been planned in the following sequence:

   i) A small programme of mixed Th-Pu oxide fabrication
   ii) Irradiation of this fuel in experimental
   reactor - CIRUS, and carrying out PIE
   iii) About 40 channels in MAPS to be loaded with this
   to gain experience with Th fuel in power reactor conditions
   iv) A programme of reprocessing these bundles to
   recover U^{233} and
   v) Physics calculational methods for this system to
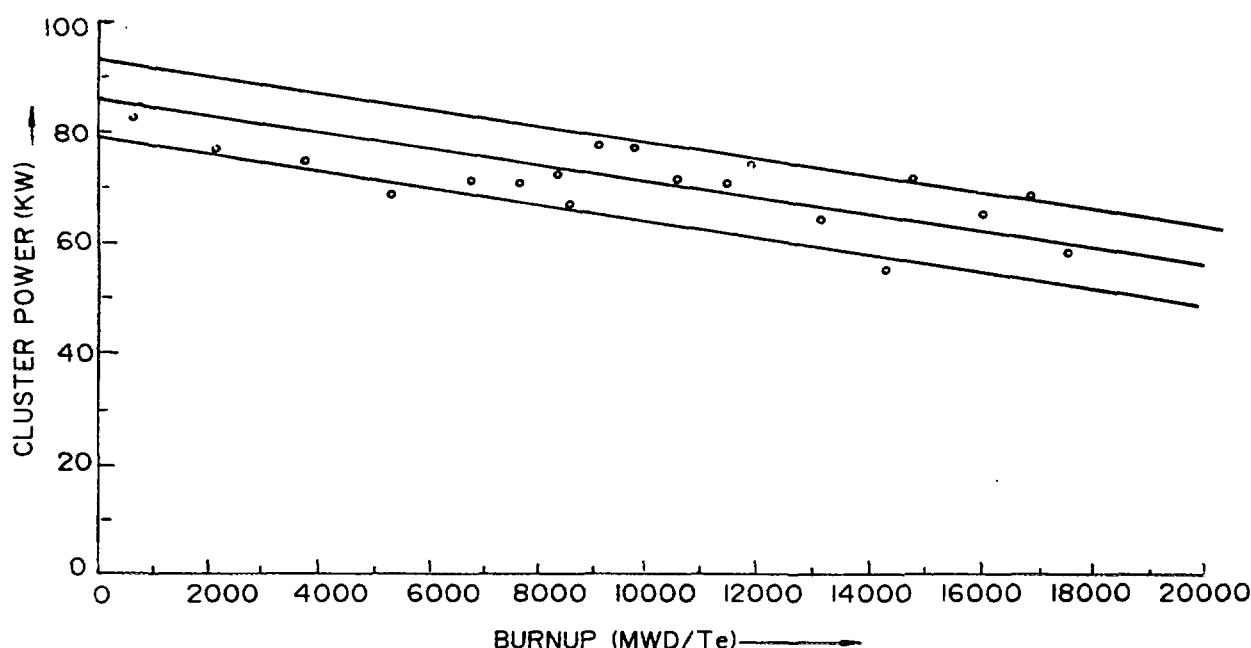   be developed into the form of computer code
   packages.

FIG.3 CLUSTER POWER WITH IRRADIATION FOR ThO₂ - PuO₂

113

TABLE-5

ISOTOPIC COMPOSITION OF PHWR FUEL WITH BURNUP(g/kg of initial heavy nuclides)

Burnup (GWd/T)	U^{235}	Pu^{239}	Pu^{240}	Pu^{241}	Pu^{242}	Total Pu
NATURAL UO_2 FUEL						
6.0	7.114	0.000	0.000	0.000	0.000	0.000
6.7	2.499	2.555	0.845	0.182	0.045	0.627
MOX-7 FUEL						
0.0	7.114	1.023	0.356	0.077	0.021	1.477
10.5	1.624	2.886	1.606	0.398	0214	5.104

An experimental cluster of ThO_2-PuO_2 fuel rods has been irradiated to a burnup of 18.5 GWd/T. The peak linear heat rating of the fuel was 385 W/cm. The variation of cluster power with irradiation is shown in Fig-3. It is planned to introduce about 40 (Th-Pu)O_2 bundles in MAPS-I as the next step of the programme.

5. PLUTONIUM-BASED FUELS WITH INERT MATRIX

Plutonium-bearing fuels, which would not breed any further Pu but only burn it, have assumed great significance in recent times. As such, metallurgical work related to the development of Pu-containing inert matrix fuels have been taken up for study. Dispersion type fuels, with matrices of ZrO_2, MgO and Al_2O_3 are under investigation. Dispersions with 10% UO_2 in MgO and Al Q were prepared and studied prior to studies on 10% PuO_2 bearing matrices.

Doping with MgO/Y_2O_3 to stabilize ZrO_2 was found necessary to prepare good quality pellets of UO_2-ZrO_2 fabricated by the standard flow-sheet with sintering at high temperature in reducing atmosphere. However, in the case of PuO_2-ZrO_2, as pellets show cracks at temperatures >1300°C, trials with sintering in air are being taken up. Optimization of starting materials characteristics is being done in the case of MgO and Al_2O_3 matrices in order to achieve densities in the desired range (94-96% theoretical density). The latter shows improved densities with small additions of MgO.

5. CONCLUSIONS

Plutonium-based fuels for light water/heavy water reactors are expected to reduce the inventories of Pu while giving substantial fuel cycle benefits, particularly in the latter case. Schemes for the utilization of thorium reduce the generation of Pu and feasibility of high burnup ThO_2-PuO_2 fuels may be demonstrated. The inert-matrix based Pu bearing fuel fabrication routes under study would lead to evolve schemes to effectively burn Pu.

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THORIUM FUEL-CYCLE DEVELOPMENT THROUGH PLUTONIUM INCINERATION BY THORIMS-NES (THORIUM MOLTEN-SALT NUCLEAR ENERGY SYNERGETICS)

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

K. MITACHI
Department of Energy Engineering,
Toyohashi University of Technology,
Toyohashi, Japan

A. LECOCQ
European Association for Molten-Salt
Reactors Development,
Orsay, France

A. FURUHASHI
Nuclear Material Control Center,
Tokyo, Japan

S.E. CHIGRINOV
Radiation Physics and Chemical Problems Institute,
Minsk, Belarus

Abstract

Thorium fuel-cycle has benefit on not only trans-U element reduction but also their incineration. The disadvantage of high gamma activity of fuel, which is useful for improving the resistance to nuclear proliferation and terrorism, can overcome by molten fluorides fuel, and practically by THORIMS-NES, symbiotically coupled with fission Molten-Salt Reactor (FUJI) and fissile-producing Accelerator Molten-Salt Breeder (AMSB). This will have wide excellent advantages in global application, and will be deployed by incinerating Pu and producing ^{233}U. Some details of this strategy including time schedule are presented.

1. INTRODUCTION

In the next century the world needs huge energy, which is indispensable to solve social and environmental problems. For such purpose, it seems that nuclear fission energy will be most promising, if the following issues would be solved: (a) safety (including elimination of severe accidents), (b) radio-waste, (c) anti-nuclear proliferation and terrorism, and (d) public/institutional acceptance relating with simplicity, flexibility and economy in the global application.

115

Therefore, the reduced generation of actinides, which is the aim of this Meeting and straightly relating with the above items (b) & (c), would be one of the most important improvements in nuclear energy technology at least.

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

In here, we should recognize that the globally necessary fission energy will be "1,000 - 2,000" TWe·Year in the next century. This is "500 - 1,000" times larger than the past fission energy production of only "2" TWe·Year, although even such huge nuclear energy will not be enough to prevent the CO_2 Greenhouse effect.

Now the most promising new fuel material: molten salts, esp. molten fluorides should be chosen depending on the long R & D efforts during 50 years after the end of Second World War, which is being accelerated after the end of the Cold World War.

In this field the most significant results were obtained by ORNL, USA during 1947 - 1976 [1], with the additive contributions of Indian, French, Russian, Japanese etc. [2, 3].

2. CRITERIA FOR MOLTEN FLUORIDE FUEL CYCLE SYSTEM.

The following criteria for molten fluoride fuel will be required as shown in Table 1. Now new fertile material should be ^{232}Th of low mass number, which is effective for reduction of actinide generation such as Np, Pu, Am, Cm etc..

Table 1. Criteria for New Nuclear Fuel Selection.

	OLD CONCEPT	NEW CONCEPT	ADVANTAGES
fertile	^{238}U	^{232}Th	few TRANS-U elements
fissile	^{239}Pu, ^{241}Pu	^{233}U	high eta value
fuel form	solid	liquid	high gamma by ^{232}U
fuel compo.	oxide, alloy, ceramics	fluoride-melt	chemical inert, lower vapor press.
moderator	H_2O, D_2O, Be, graphite	graphite	high melting temp., high radiation resis.
coolant	H_2O, D_2O, He, Na	fuel-salt	chemical inert, lower vapor press.
elec. gene.	steam turbine	steam turbine (higher temp.)	higher therm. effici.
fissile breeding	fission-breeder	near breeder + spallator	power-size flexible, flexible breeding

A high gamma activity of new fissile ^{233}U fuels due to the byproduct ^{232}U will be remotely managed by the fluid-type fuels. In practice, depending on the intense study of ORNL, the molten fluoride-salts:

$$^{7}LiF - BeF_2 - ThF_4 - ^{233}UF_4 = (72-x) - 16 - 12 - x \text{ [mol\%]}$$

should be chosen as a most typical example.

In early stage historically the other fuel salts such as $NaF- BeF_2 - ThF_4- ^{233}UF_4$ had been examined in advantages of no isotope separation of ^{7}Li and of no production of tritium. However, this salt has not enough solubility of ThF_4, which is less than 6 mol%.

Moderator Graphite can be used in bare form without any corrosion and wetting. Graphite is one of the best high-temperature resistant and radiation resistant materials.

In the past reactor design, the heat removal of fuel materials was done by water, liquid metals or gas. However, these have several troubles, and are origins of "severe accidents". Now we should choose a chemically inert and low vapor pressure medium, and this is the above fuel salt itself.

At moment, our reactor design is also using the steam generator technology for electric generation same as the previous reactors, although higher output temperature and higher thermal efficiency more than 45 % will be expected in super-super critical steam condition. Therefore, intermediate coolant salt will be necessary, but it is also chemically inert and has low vapor pressure, and contributes not only on the more reliable and safe operation, but also on the direct supply of high-temperature industrial heat in future.

For the minimization of radio-waste generation, which includes the minimization of actinide generation, a breeding thorium fuel cycle should be closed, furnishing the recirculation of neutron reaction products with their extraction as little as possible. This is possible more easily in the "symbiont" type system such as our proposal: THORIMS-NES (Thorium Molten-Salt Nuclear Energy Synergetics) [2]. It is not a "hybrid" type fission breeding power station such as "Molten-Salt Breeder Reactor [MSBR]", which is a non-flexible critical system in large size only, and still is not enough in breeding ability to allow the steep growth rate such as 10 years in the doubling time of power generation.

The THORIMS-NES depends on the symbiotic (synergetical) breeding system coupling the following two systems:

(A) Simple Fission Power Stations [utility facilities] : Molten-Salt Reactors [MSR] of fissile self-sustaining or higher conversion-ratio without continuous chemical processing except fission-gas removal.

(B) Fissile Producers [process plants] : Accelerator Molten-Salt Breeders [AMSB] of subcritical system (spallator) depending on the spallation neutron generated by 1 GeV proton.

These are backuped by the dry chemical processing plants for spent fuel management to establish the flexible and simple fuel cycle composed of molten salt only, which has high flexibility in fissile species and concentrations.

The final pure "Th-U molten salt fuel cycle" will be able to create a rational nuclear industry without Trans-U elements in practice. The transient approach for realization of "New Th Fuel Cycle" from the present U-Pu cycle will not be difficult, if the positive utilization (and elimination) of accumulated plutonium are allowed. The detailed explanation will be shown in Sec. 3.3. and Chap. 5.

3. APPLICATION TO PU-INCINERATION
FOR EARLY DEPLOYMENT OF TH-UTILIZATION.
3.1. Incineration by MSR--"FUJI-Pu".

In ORNL, 1969, it was demonstrated by their experimental reactor: MSRE that MSR is practically useful for burning Pu added as partial fuel component.

In recent years this result has been deeply examined relating with the disposition of weapon head Pu and reactor Pu from spent fuels by ORNL [4] and us [5, 6].

Our standard MSR was studied in several sizes such as 7 MWe miniFUJI [7], 100-150 MWe FUJI [8], and 1 GWe superFUJI [9]. They have merits of (a) no core graphite exchange, (b) no continuous chemical processing except removal of fission-gases and tritium, and (c) high conversion-ratio such as "near breeder".

In here we shall discuss Pu incineration behavior comparing the following three versions of FUJI-Pu:

[A] FUJI-Pu1: initial fissile is PuF_3 only, and fertile is ThF_4 (12 mol%),

[B] FUJI-Pu2: the same, but removes ^{233}U in interval of 300 days, and

[C] FUJI-Pu3: the same, but lower fertile content 8 mol%.

[A] and [B] were reported in IAEA Meeting of last year [6], on the basis of their 900 days burning behavior. Here, the version [C] will be briefly presented including 400, 900 and 1200 days burning behavior.

The reactor specification is not fully same as FUJI-Pu1 and -Pu2, and shown in Table 2. Some important characteristics of Pu burning are shown in Table 3 comparing with the other versions.

Table 2. Principal Design Parameters of FUJI-Pu3.

Thermal capacity	250 MWth	Max. neutron flux:	
Net elec. generat.	100 MWe	Graphite (>52KeV)	6. 1x10^{13}n/cm^{2}s
Thermal efficiency	40 %	Metal (>0. 8MeV)	2. 3x10^{11}n/cm^{2}s
Reactor vessel:		Fuel salt:	
Diameter/Height	5. 7/5. 2 m	PuF_3 concentration	0. 3 mol%
Core-zone:		ThF_4 concentration	8. 0 mol%
Radius/Height	1. 57/1. 27m	Volume in reactor	11. 0 m^3
Graphite frac.	80 vol%	Total volume	16. 5 m^3
Blanket-zone:		Flow rate	0. 55 m^3/s
Thick. Rad. /Axi.	0. 33/0. 4 m	Temperature In/Out	838/938 K
Graphite frac.	60 vol%	Fuel conversion ratio	0. 64-0. 60
Refrector-zone:		Inventory:	
Thick. Rad. /Axi.	0. 9/0. 9 m	Pu Initial*/Final	0. 73/1. 19 ton
Graphite frac.	99 vol%	Th	18. 9 ton
Power density	6. 6 KWth/1	Graphite	213. ton

[* Pu from PWR fuel--33GWd/t burn up and one year cooling:
1. 5w/o^{238}Pu, 55. 0w/o^{239}Pu, 25. 3w/o^{240}Pu, 13. 2w/o^{241}Pu, 5. 0w/o^{242}Pu]

Table 3. Net Production Rates of Pu, U and Minor Actinides

	BWR (U)	BWR (MOX)	FUJI-Pu1	-Pu2	-Pu3	-Pu3	-Pu3
Therm. output [MWth]	3,000	3,000	250	250	250	250	250
Elec. Genera. [MWe]	1,000	1,000	100	100	100.	100	100
Operation	45gmd/t	45GWd/t	900d	900d	900d	400d	1,200d
Net Production [Kg/GWe Year]							
Total Pu	230	-649	-919	-980	-979	-999	-969
Am + Cm	25	133	62	71	104	99	107
$^{233}U + ^{235}U + ^{233}Pa$	0	0	453	489	494	541	470

[BWR (U) is operated with UO_2 fuel, and BWR (MOX) with mixed-oxide fuel.]

From Table 3, it will be concluded that FUJI-Pu3 will be better and more practical than others due to its simple operation mode and high performance. Such improved Pu burning performance was achieved by modifying not only ThF_4 content but also some suitable core design parameters. The time interval of produced ^{233}U removal for improved performance will be able to be chosen to 5 - 10 years. This chance for processing will be used for F management [cf. Sec. 4. 4].

Fairly high Am and Cm production is the result of using LWR Pu.

3. 2. Incineration by AMSB-Pu.

Pu and TRU can also be transmuted by AMSB, producing ^{233}U in parallel, in which the production ratio of ^{233}U to transmuted Pu is much higher than the case of FUJI-Pu [10, 11].

However, the development of AMSB-Pu will be delayed about 5 -10 years than FUJI-Pu due to the large-current accelerator development and proton-injection port engineering, although AMSB has significant advantages in the issues of radiation-damage, heat-removal and reactor-chemistry.

3. 3. General Strategy by THORIMS-NES.

It was hopefully demonstrated that the complete elimination of Pu existing at present and future in the world will be really established economically not spending any dead works, if we use the following strategy [9, 10], which is briefly explained here again:

(1) D-plan: Pu (and trans-U elements [TRU]) separation straightly in the form of molten fluorides by Dry-process from the spent solid-fuels accumulating in the world. The technological basis has been examined by French, Russian, etc. [12].

(2) F-plan: Pu-burning and ^{233}U-production by Fission MSR [FUJI-Pu], as explained already in Sec. 3. 1.

(3) A-plan: Pu-burning and ^{233}U-production by AMSB-Pu, in parallel with F-plan. This was shortly mentioned in the above Sec. 3. 2..

The accumulated Pu in the world will become more than ten thousands till the middle of next century in the recent trend. However, applying the above F- and A-plans the entire elimination of Pu and the establishment of pure Th fuel-cycle Nuclear Energy Industry will be realized in some earlier stage before 2050

The radio-waste isotope soluble in fluoride fuel (or target/blanket) salts are principally retained in molten-salt fuel cycle, which will be transmute to non-radioactive isotopes depending on not only neutron reactions of several energy spectrum but also natural transmutation of short-life radio-isotopes in salts.

The chemical processing of radio-wastes and non-radioactive chemical wastes in salt will be arranged in some time interval such as several years considering the balance of several factors such as neutron economy, structural materials compatibility, nuclear materials solubility, etc.. One example for such evaluation will be shown in Sec. 4. 4..

4. SIGNIFICANT ADVANTAGES' IN SEVERAL TECHNOLOGICAL ASPECTS.

4. 1. Resources.

Thorium resources have been sufficiently discovered in the world. The most popular form is a "black (or heavy) sand" [monazite] in beach, which will not produce any environmental disturbance Only two million tons Th is enough for the electric production of 1, 000 TWe year in the next century, practically burning the one-third of Th. Uranium resources are poor (one-third of thorium), and were already extracted 1. 5 - 2 million tons till now.

4. 2. Some comments on Reactor Safety.

MSR (and AMSB in general) is a significantly safer reactor, and has essentially "no severe accidents". The most important safety performances are coming from the following factors:

(1) The system pressure is lower than 5 atm at least.

(2) The fuel salt is chemically inert, and $_{not}$ reactive with air and water.

(3) The boiling point of fuel salt is 1673K, much higher than operation temperature 973K, and it has low vapor pressure.

(4) The fuel will become just critical when it coexists with the graphite moderator. Therefore, leaked fuel salt will not introduce re-criticality.

(5) MSR has a large negative prompt temperature-coefficient. 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 in ^{233}U fission is smaller than that in ^{235}U However, it is controllable owing to the longer neutron-life, and large negative prompt temperature-coefficient

(7) As the fuel composition can be made up anytime if necessary, the excess reactivity and control-rod reactivity are sufficiently small, and the reactivity shift by control-rods is small.

(8) Gaseous fission products including tritium are removed from fuel salt always, and their leakage in accidents can be minimized.

The triple confinement principle of radio-activity are ensured in MSR as same as in the other solid-fuel reactors. Its illustration in MSR has been shown in Fig. 2 comparing with the case of PWR. The reliability of each barriers in MSR is much higher than the case of PWR depending on not only the factors mentioned the above (1), (2), (3) and (8) but also the soundness of the first barrier, which is composed of simpler reactor-vessel, piping and intermediate heat-exchanger tube-wall. This latest tube-wall is very much thicker than "fuel-clad" tube wall and has not any serious irradiation and thermal-stress

The "severe accident" will never occur in MSR even assuming the full stops of primary and secondary pumps all and the fault of reactor scram, although the detailed examination will not presented here.

Even in the case of "severe reactor destruction" by military attack or sabotage, Graphite-moderated Molten Fluoride Reactors might have mostly safer characteristics comparing with any other ordinary Solid Fuel Reactors:

(i) only a weak release of gaseous radioactivity due to fission-gas removal in daily basis,

(ii) no core melt down, and no re-criticality due to isolation of moderator graphite and fluid fuel, which might be drained automatically and/or leaked. Leaked fuel-salt is frozen to stable glassy mass confining radio-activity, and never producing any aerosol like as aqueous fuel solution.

(iii) Although high quality reactor graphite with high heat-capacity/thermal-conductivity is not easily fired in virtue of no existence of high temperature heat-sources. Even if graphite firing started, simple suffocation extinguishment will be enough, because graphite will not be wetted by salt except having a small residual fuel salt only. It means very weak radioactivity release without any terrible fear.

4. 3. A Comment on Reactor Operation/Maintenance.

Reactor systems in THORIMS-NES is generally very safe as mentioned in the above section, and their operation/maintenance work will become simpler and easier Here, some additive evidences will be given.

(1) The reactor vessel is not opened during its life, and the number of control rods is very few (or no rods might be expected) Therefore, the reactor vessel will be a simple welded tank itself without any big flange, and no fuel-handling machine at all

(2) The primary circuit system is confined inside a high-temperature containment of about 810K, which results more simple configuration without any heater, insulator, shield and instrumentation except some flowmeters and surface-level meters, etc..

Therefore, the full-remote maintenance, inspection and repairing will be easy, not introducing any worker exposures.

(3) If fuel (or target/blanket) salt is leaked from systems, the bottom of the high-temperature containment includes a spill-pan. Leaked salt will be automatically guided to drain-tank If needed, the radioactive salt can washout by clean carrier salt (^{7}LiF-BeF$_2$).

It should be recognized that these salt will be frozen as non water-soluble glassy matter stably sticked on wall in lower temperature. The radioactive contamination of reactor building will be negligible by handling such repairing facilities. This is significantly different from the troublesome contamination in "Aqueous Homogeneous Reactors" [13].

Although we should take care on off-gas system management, the general worker exposures will not be important, which was suggested the recent evaluation by National Academy of Sciences, USA[14].

As MSR(FUJI or FUJI-Pu) is self-controllable, the operation work is very simple and main items are (a) off-gas management, (b) redox electrochemical potential control of fuel salt, and (c) semi-periodical additions of Th, Pu and/or U in the frozen eutectic-salt forms

Th $\cdot$ ^{7}LiF $-$ 29 m/o ThF$_4$ (m. p. 838 K),

Pu : ^{7}LiF $-$ 19.5 m/o Pu F$_3$ (m. p. 1016 K), and

U . ^{7}LiF $-$ 27 m/o UF$_4$ (m. p. 763 K).

Pu will be added to keep criticality about once per week. Th content is kept constant adding about once per 100 days to hold the variation less than 1 %

The chemical balance by fission of ^{233}U will introduce some excess free F atoms. However, in FUJI-Pu3 the main fission is depending on ^{239}Pu, and neutrality will nearly be held in result. By the above addition of fuel components ^{7}LiF concentration will increase to 71.8 m/o, which will be corrected by addition of 0.66 m/o BeF$_2$ in total.

4.4. F.P. Behavior in FUJI-Pu3.

In FUJI-Pu3, the 1200 days operation will result F.P. production as shown in Table 4.

(1) Gaseous species: Fission gases will be isolated directly from fuel salt, and tritium from cover-gas of secondary system mostly.

(2) Undissolvable components in fuel salt: The total amount was assumed 91.1 Kg Some part will be filtrated, and residual deposit will not disturb the salt flow, because the salt volume ratio is fairly big 20 Vol% in core.

(3) Dissolvable components·

a) Stable salts except three-valency ones: These are sufficiently low concentration not producing any chemical problems.

b) Three-valency salts: Considering the solubility limit, the total amount of three-valency salts should be less than 1 mol%. In this case Pu^{3+} is 0.48 mol% and the other total is 0.078 mol%. It will promise the 10 years operation at least.

The general behavior of F.P. will tentatively have not any serious problems.

4.5. Nuclear Proliferation Resistance.

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

Table 4.   F. P. Amounts After 1200 days Operation in FUJI-Pu3.
[* Amount variable by the behavior of precursor]

(1) Gaseous-species:		(2) Undissolvable components:			
Xe	about*63 Kg	Mo	27. 7 Kg	Rh	7. 3 Kg
Kr	about 19 Kg	Tc	7. 3 Kg	Pd	21. 3 Kg
T	about 32 g	Ru	25. 3 Kg	Ag	1. 9 Kg

(3) Dissolvable components:

a) Stable salts except three-valency ones:

I	about	7. 8 Kg	0. 0061 mol%
Br	about	1. 3 Kg	0. 0016 mol%
Cs	about	58. 0 Kg	0. 043 mol%
Rb	about	21. 6 Kg	0. 035 mol%
Sr	about	35. 4 Kg	0. 040 mol%
Ba			0. 0023 mol%
Zr			0. 024 mol%

b) Three-valency salts:

Pa	0. 0022 mol% [in ope.]	[0. 00025 mol% after 90 days]
U (+++)	0. 0064 mol%	
La	0. 0063 mol%	
Ce	0. 0145 mol%	
Pr	0. 0066 mol% ·	
Nd	0. 0200 mol%	
Pm	0. 0013 mol%	
Sm	0. 0044 mol%	
Y (about 14. 75 Kg)		
	0. 016 mol%	sub-total 0. 078 mol%
Pu	0. 482 mol%	TOTAL 0. 56 mol%

(1) **Macroscopic view in global fuel cycles**: Pu in spent fuels of various thermal reactors are steadily increasing. Especially vast amount of them is expected in developing countries in near future through promotion of nuclear power generation mostly with LWRs.

Pu brings proliferation risks even when it remains in spent fuels. When the issue of energy resources urges us on the separation and use of Pu -- that will be very likely -- proliferation risks will further increase unless we have not good scheme of utilizing Pu.

Th fuel cycle development through Pu incineration by THORIMS-NES is the best scheme we have for this purpose, since it actively reduces and simultaneously suppresses new Pu production in spent fuels from LWRs.

Pu 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 advantages over FBR fuel cycle. Therefore, THORIMS-NES would be able to make great macroscopic contribution to global fuel cycles.
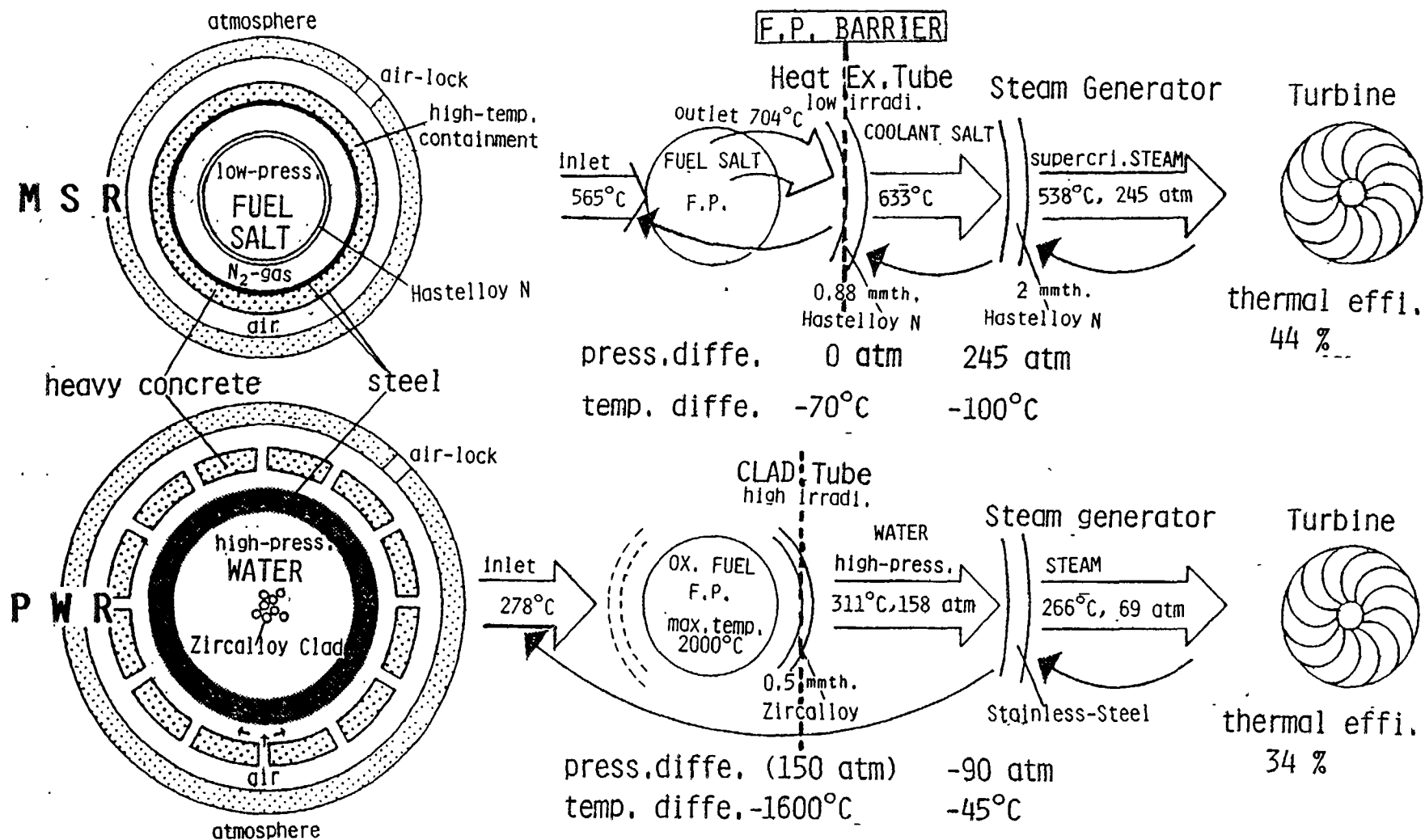
Fig. 1. Comparison of Safety-Barriers in "MSR" and "PWR".

(2) <u>Pu vs ^{233}U</u> (<u>FBR vs MSR</u>) : Significant quantity (SQ) in nuclear safeguards is not so much different between Pu and ^{233}U, but diversion resistance will be significantly larger in ^{233}U.

^{233}U usually contains more than 500 ppm ^{232}U and its daughter nuclides. Some of them emit strong high energy (above 2 MeV) gamma rays. They bring 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 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 is 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) than Th of main fertile material in virtue 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 potential to easily become weapon material. U-Pu fuel cycle never get out of this yoke.

FBR fuels must be recycled in fairly short periods to retain their breeding power in practical level. So annual throughput of Pu in FBR fuel cycle will become very large and brings significant safeguards and transportation problems. The situation in MSR/THORIMS-NES is much easier, because MSR will become fuel self-sustaining near breeder, resulting few transportation and few reserving fissiles.

(3) <u>Microscopic view in reactor site</u>: 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 Pu or ^{233}U weights 800 Kg with the volume of about 250 liters. This makes the theft effectively impossible.

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

Reprocessing and refabrication (re-preparation) of MSR liquid fuel is much easier than those of FBR solid fuel . This will reflect to the possible difference of theft and diversion between both reactor types. Transportation -- the vulnerable point in fuel cycle -- can also be much reduced in MSR, which is principally fuel self-sustaining "near breeder" and is usually adopting on-site processing.

These advantages can similarly be hold in the case of AMSB (accelerator molten salt fissile producer). AMSB and fuel processing facilities will be non-utility/process plants in essence, and will be accommodated inside of Regional Centers heavily safeguarded.

To summarize aboves, it should be strongly recommended to convert Pu 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 in global scale.

5. MAIN TECHNOLOGICAL PROBLEMS AND TIME SCHEDULE IN R&D.

As mentioned in Chap. 2, the realization of MSBR has several difficulties, which will be solved by a new concept of THORIMS-NES in the following points:

(A) MSR should eliminate the core-graphite exchange and the continuous chemical processing in situ, not aiming to establish the "breeders". Now a simplified MSR: FUJI will become easier in development approaching from the smaller size power stations such as FUJI of 100 - 200 MWe.

FUJI can serve as flexible utility power stations.

(B) In the next step delaying a little from FUJI, we should develop some fissile-producers to realize a "molten-salt breeding fuel-cycle", in which the doubling-time of power generation growth has no limitation.
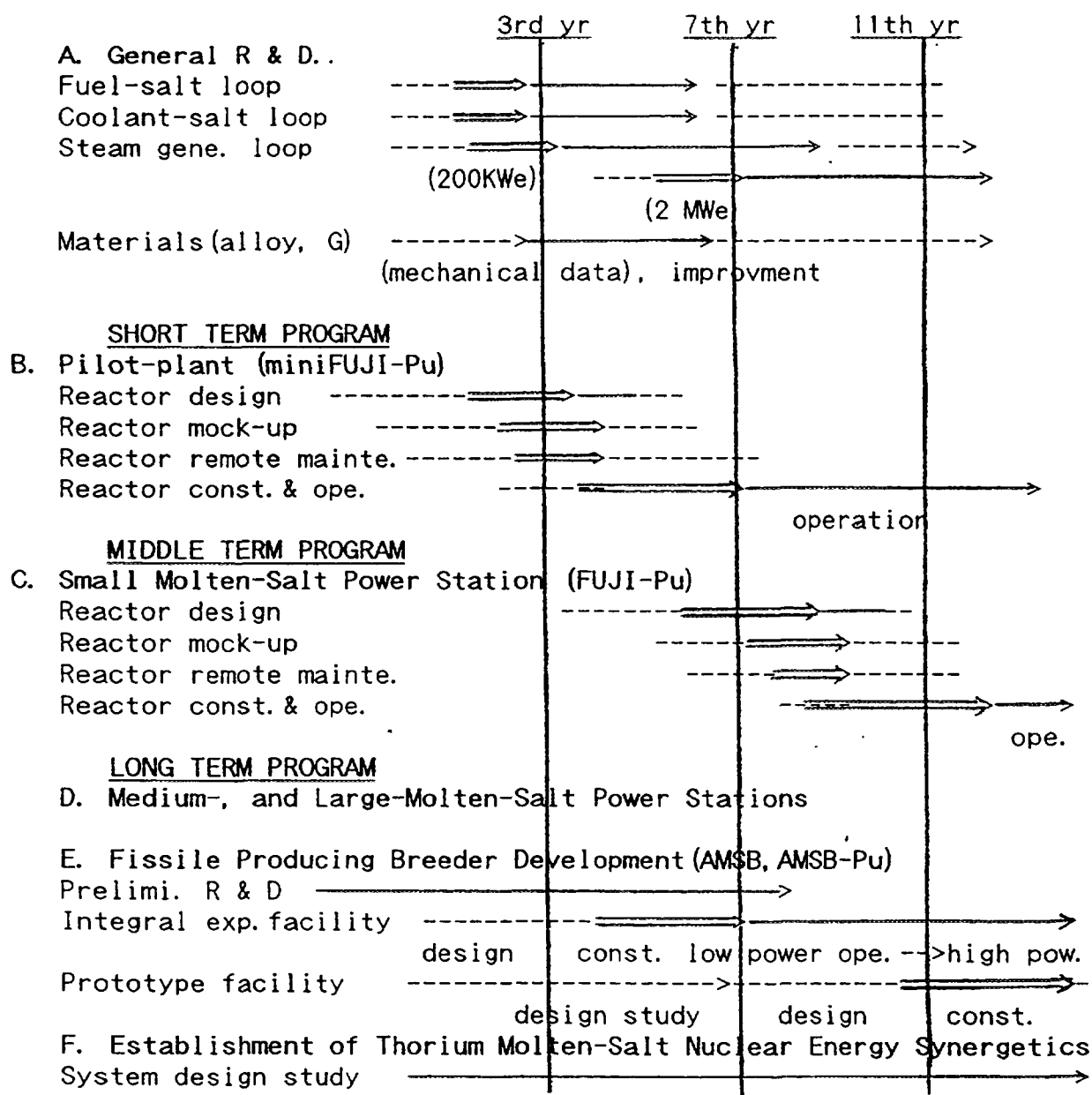

Fig. 2. Developmental Schedule in THORIMS-NES
(====> construction: ----> operation)

The most promising one will be AMSB using 1 GeV proton spallation reaction in single-fluid type target/blanket salt system. AMSB will be easier in development owing to no serious radiation damage of solid materials.

(C) To realize THORIMS-NES, we are recommending to couple with Pu-disposition work as mentioned in Sec. 3. 3, because the Pu burning in MSR will not introduce any serious difficulty [cf. Sec. 3. 1 & 4. 4].

Now how many years would be necessary to deploy the real Pu incineration by FUJI-Pu ? The answer in a study of LLNL was 23-27 years as referred in the recent NAS report [14].

However, if we follow on the strategy based on THORIMS-NES, the commercial application will not be impossible even after 15 years or some after decision.

This strategy is based on the followings:

[1] The first step -- miniFUJI-Pu project:

Fully depending on the successful result of MSRE, ORNL, a similar but three-times lower flow-rate Nuclear power Station: miniFUJI-Pu should be build and operate spending 6 - 7 years period as shown in Fig. 2.

The basic R&D works will not need so many items, due to no relation with a high performance "breeder" conditions. The detailed examination was done in the previous reports [2].

[2] The second step -- FUJI-Pu project:

Now in a few years delay, 100-200 MWe size FUJI-Pu power stations might be built and operated, and the commercial operation for Pu-disposition will be commissioned about 12-15 years after starting miniFUJI-Pu project [cf. Fig. 2].

As a long term program, larger MSR program and AMSB program should proceed in parallel as shown in Fig. 2, which was discussed in the previous papers [2].

6. CONCLUSIONS.

For the reduction of trans-U elements the Th-^{233}U breeding fuel-cycle should be established. The globally applicable system in the next century will be symbiotic THORIMS-NES composed of simple MSR power stations such as FUJI, superFUJI, and fissile-producer by spallation reaction such as AMSB.

The real industrialization of such system will be accelerated by coupling with Pu disposition. The initial deployment will rationally proceeded by FUJI-Pu3, which would be continuously operated 5 - 10 years interval restarting after the extraction of ^{233}U and some FP. The necessary time for FUJI-Pu3 development will be only 12 - 15 years.

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A STUDY FOR OPTIMAL TRANSMUTATION SYSTEM

W.S. PARK, T.Y. SONG, H.S. SHIN, C.K. PARK
Korea Atomic Energy Research Institute,
Taejon,
Republic of Korea

Abstract

Couple of transmutation systems are being under investigation to design the optimal transmutation device. Several basic studies were performed for that objectives: (1) select the radioactive nuclides to be transmuted: (2) investigate the physical characteristics of each nuclide; (3) study the most favorable neutron energy environment for the transmutation. The existing LWR and LMFBR cores were found to be not a satisfiable ones in terms of transmutation rate itself.

Introduction

Since the late of 1980s, the development of nuclear transmutation technology has been one of the main topics in nuclear research. Much more attention is expected to be paid in near future because of the increased concerning on environment and the difficulty to have a geologic repository.

As an alternatives or supportive options for geologic isolation of the nuclear waste. Kaeri embarked in the research for nuclear transmutation. Various transmutation technologies for high level radioactive waste(HLW) are currently being proposed at many organizations in the world such as thermal/fast reactor based and accelerator driven subcritical systems[1]. No detail assessments among the technologies have been performed to date. It might be very difficult to have international consensus on those systems. Each nation(or institute) has developed her own technology on the basis of the nuclear fuel cycle and the technical strong points she has.

The technologies currently introduced are supposed to be everything the transmutation device can be in terms of technical and economical feasibility. Kaeri decided to develop her own model throughout two stage plans. In the first stage. Kaeri will set up basic model the most preferable to Korea by evaluating the proposed technologies. In the second stage. the conceptual design will be performed for the optimum transmutation model. On the way for the goal of the first stage, a couple of basic studies were done. The results of basic studies and the temporary direction Kaeri has were discussed in this paper.

Methodology for Optimal Basic Model Determination

Basic model is expected to be determined based on the procedure shown in Figure 1.

Selection of Nuclides to be transmuted
- Evaluation of Enviromental Risk
- Nuclear Characteristics

Neutronic Performance of System
- Transmutation Rate
- Fission Energy Produced

General Performance of System
- Technical Feasibility
- Cost Analysis
- Public Acceptance

Optimal Basic Model Determination
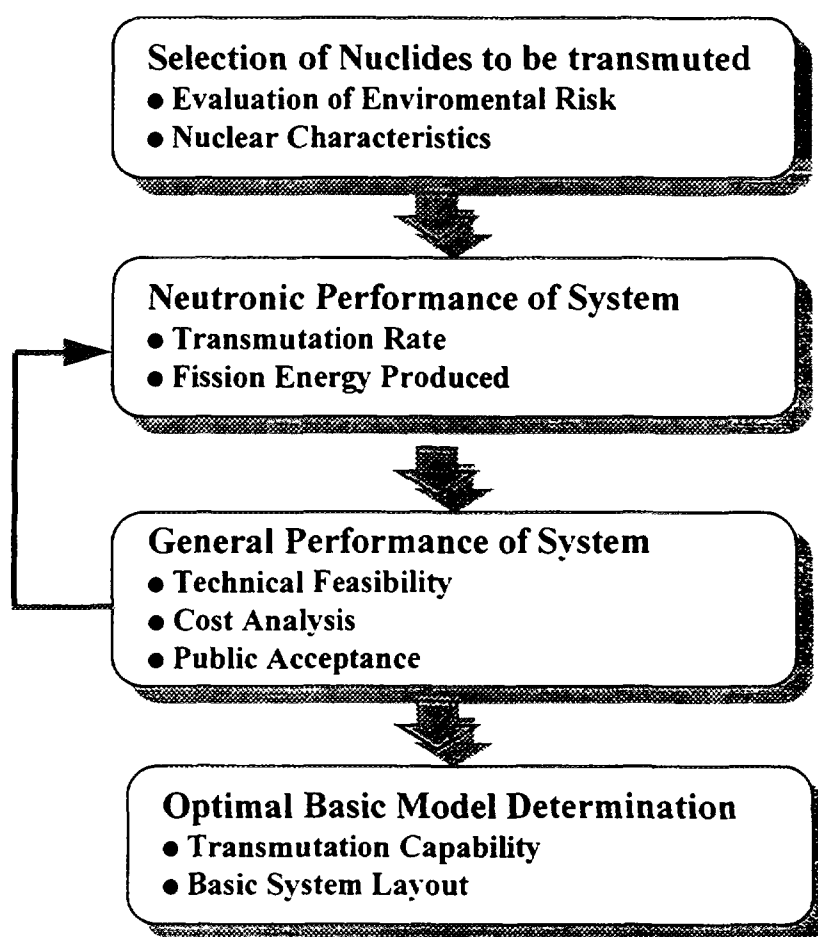- Transmutation Capability
- Basic System Layout

Figure 1 Optimal System Design Procedure

As it is known, one ton of LWR spent fuel includes 10 kg of actinides and 30 kg of fission products[2]. Plutonium occupying 92% of actinide is considered as an energy resource rather than waste to be eliminated. Two major factors are considered for the selection of the nuclides to be transmuted: (1) nuclear characteristics; (2) environmental risk due to the nuclide. In the evaluation of the nuclear characteristics of each radioactive nuclide, its half life and toxicity in terms of water dilution volume are included. On the other hand, solubility and retardation factor are major factors to be evaluated in the environmental risk point of view.

The objectives of transmutation is not just to convert the nuclide from one to another but is to reduce the toxicity to the acceptable level within the time period human can handle. The transmutation rate of each nuclide varies from system to system because a neutron cross section does from nuclide to nuclide. In order to estimate the overall transmutation

130

efficiency of the proposed system, following performance index of neutron energy spectrum was set up,

$$PI = W_t \sum_i \omega_i \varepsilon_i + W_f \sum_j \omega_j f_j \quad\text{-----------------------------------(1)}$$

where,

$W_{t,f}$: weighting factor for toxicity reduction and the amount of fission energy

ω_i : weighting factor accounting for the toxicity and the amount of nuclide i

ε_i : the ratio of the toxicity without transmutation to the toxicity with transmutation for nuclide i

f_j : amount of energy generated by fission of nuclide j

In equation (1), first term account for the toxicity reduction and second term is for the energy production by fission. By including the fission term in PI, the system which has a neutronic performance enhancing the system economy more can be preferred. The optimal system is the one maximizing PI. Table 1 shows the transmutation systems currently proposed and being investigated.

Table 1 Typical Transmutation Systems

Neutron Energy Spectrum	Transmutation System
Thermal	• LWR Based • CANDU Based • Accelerator Based(ADDT)
Fast	• LMFBR Based • Accelerator Based

General performance of a system is evaluated in terms of cost, technical feasibility, public acceptance, etc. The optimal basic model is finalized by considering the general performance as well as the neutronic performance.

Basic Studies and Results

Selection of Nuclides

As it was mentioned before, plutonium was not considered as a nuclide to be transmuted. The assessment on the nuclear characteristics and environmental risk of nuclides produces Table 2. The nuclides in Table 2 are the major radioactive materials to be eliminated.

Table 2. Long-lived Radioactive Nuclides to be Transmuted

Nuclide	Half Life(year)	Amount(gram)[#]	Water Volume(m^3)[*]
Np-237	2.1E6	440	2.4E8
Am-241	432	500	8.6E11
Am-243	7.4E3	90	5.0E10
Tc-99	2.1E5	760	8.5E7
I-129	1.6E7	180	2.9E9

\# : Amount per one ton of spent fuel
* : Water Volume required to dilute the toxicity of nuclide to the level of MPC(m^3/MG of Nuclide).

Neutronic Performance of the System

The transmutation capabilities of LWR and LMFBR based systems were investigated by using the neutron energy spectra of a typical LWR and LMFBR. The calculations were done by using ORIGEN2[3] and WIMS[4].

The meaningful transmutation device should be able to reduce the amount of toxicity considerably within a manageable time period. In this study, 500 year was assumed as a period man can predict. In order to see the changes in the toxicity reduction rate due to the employment of transmutation system, the toxicity was calculated for two cases: 1) no irradiation and natural decay for 500 years; 2) irradiation for 3 years and natural decay for remaining 497 years. Also the amount of the original nuclides remained after 500 years was evaluated to see the relationship between the toxicity reduction and the reduction of the original nuclide. Four different flux levels were employed to check the sensitivity of the transmutation efficiency to the flux level. Table 3 summarizes the results of the evaluation. In the table, the numerical value represents the ratio of the value obtained from the irradiated case to the one from no irradiated. Thus lower value means higher efficiency.

For the toxicity due to the fission products(I-129, Tc-99), LWR based system has higher performance than LMFBR based system. In addition, the destruction rate of original nuclide is directly reflected in the toxicity change in fission product transmutation. Very similar trend can be found in Am-241/243 transmutation. However, the case of Np-237 is totally different. Any reduction in the toxicity can be found in both of the systems although considerable amount of Np-237 itself can be destroyed. It means that most of Np-237s are destroyed by neutron capture under existing LWR and LMFBR energy spectra and those capture reactions generate lots of higher toxic actinides. It should be noted that the flux levels are ~ 10^{14} n/cm^2-sec and ~10^{15} n/cm^2-sec in existing LWR and LMFBR core,

132

respectively. In those flux range the transmutation rate is very small except the case of Am-241/243 and the small transmutation rate loads too much burden onto the separation system. Thus present LWR and LMFBR cores are not suitable for the purpose of the transmutation.

Table 3. Comparison of transmutation capability between LWR and LMFBR based system

		Np-237		Am-241/243		I-129		Tc-99	
		MR	TR	MR	TR	MR	TR	MR	TR
LWR System	1.0E13**	0.97	9.15	0.90	0.89	0.995	0.995	0.991	0.991
	1.0E14	0.73	65.5	0.42	0.39	0.95	0.95	0.917	0.917
	1.0E15	0.05	61.7	0.05	0.05	0.61	0.61	0.421	0.421
	1.0E16	0.001	1.1	0.001	0.001	0.007	0.007	0.001	0.001
LMFBR System	1.0E13	0.998	1.4	0.99	0.976	0.999	0.999	0.999	0.999
	1.0E14	0.98	4.7	0.97	0.965	0.996	0.996	0.996	0.996
	1.0E15	0.84	32	0.88	0.96	0.964	0.964	0.958	0.958
	1.0E16	0.18	72	0.25	0.23	0.69	0.69	0.654	0.654

MR : Mass Ratio [M(Irrad)/M(No Irrad)]
TR : Toxicity Ratio[T(Irrad)/T(No Irrad)]
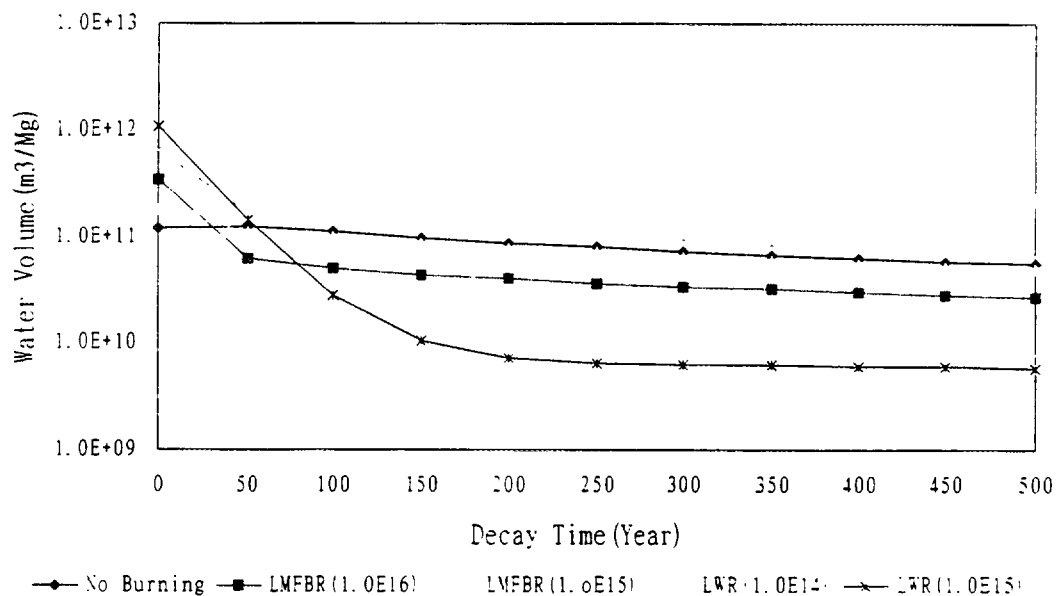** : neutrons/cm^2-sec

Figure 2. Plutonium Toxicity Variation after 3 year burning at Various Flux Conditions

Although plutonium is not considered as the nuclide transmuted in Kaeri, some calculations were done to examine how much toxicity can be reduced when plutonium is burnt up in existing LWR or LMFBR reactors. It was found that a considerable amount of toxicity reduction can not be expected under present flux level of LWR or LMFBR. This fact means that plutonium burning in LWR or LMFBR is not a burning of toxic actinide but is just a generation of energy. Figure 2 shows the results of the calculations.

Summary

Kaeri is currently investigating the transmutation capability of each proposed system and doing some study on its general performance such as a technical feasibility, economical gain/loss, and public acceptability

It is possible to transmute a radioactive nuclide by using various types of particle such as proton, electron, photon. Using neutron is found to be the most effective way. It is noted that each neutron has its own production cost although the cost varies from system to system. From the economical point of view, the effectiveness of a neutron should be maximized

It is found that the capture cross section of Np-237 is 10.000 and 100 times higher than the fission cross section in LWR and LMFBR energy range respectively[5]. Thus more than 99% of Np-237s are destroyed by neutron capture which might produce more toxic higher actinide. Table 3 reflects this kind of phenomena. The neutron cross section study tells that Np-237 and Am-241 have a reversing point around neutron energy of 1 MeV where fission cross section starts being higher than capture cross section. Fissioning destroys the nuclide in the most efficient way in terms of toxicity reduction rate and system economy. Therefore neutron higher than 1.0 MeV is recommendable for the transmutation of the toxic actinide. On the other hand, low energy neutrons are much more effective for the transmutation of toxic fission products

From the preliminary study, following tentative directions were obtained,

- Existing LWR and LMFBR core conditions are not appropriate for the transmutation, even for the fission products

- The preferred neutron energy for actinide transmutation is totally in oppositesste from that of fission product transmutation

- Although very high thermal flux level(> 10^{16} n/cm^2-sec) can transmute both of actinides and fission products altogether, such a system is not practical because of the irradiation problem of structural material and its extremely low neutron economy(high system cost)

134

- It is almost impossible to transmute the toxic nuclides to be selected using one single neutron energy spectrum. It might be suggested to develop a hybrid system where two extreme neutron energy spectra can coexist.

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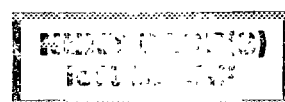
A PARTITIONING-FREE TRANSMUTATION CONCEPT OF NUCLEAR WASTE REDUCTION

S. TACZANOWSKI
Faculty of Physics and Nuclear Techniques,
University of Mining and Metallurgy,
Cracow, Poland

Abstract

The idea of a symbiotic nuclear energy system, consisted of an Accelerator-driven Fuel Regenerator (AFR) and a number of LWRs serviced by it, is the subject of this study, in view of supposed safety and partitioning avoidance advantages. The design premisses leading to this concept are widely discussed. The regeneration of spent fuel halves the amount of high level waste from the symbiotic reactors while neither mechanical nor chemical intervention into the fuel is needed. Thus, the concept itself does not draw directly on any partitioning, The need of hard neutron spectrum favouring neutron captures over fissions, that conditions the regeneration of the fuel and the requirements of safety /e.g. fire resistance/, substantiate the choice of a lead coolant. Also the latter reasons have given rise to a need of constancy /over the whole fuel regeneration campaign/ of the neutron mutiplication factor k_{eff}. This requirement, important for systems safety, can be achieved due to the compensation of fissile breeding in the spent fuel either by burning of separate fissile material (e.g. c-Pu or w-Pu) or by adequate shuffling of the fuel. Such a stabilization of k_{eff} safely allows, in turn, for its higher values and thus higher energy gains i.e. for reducing the accelerator current - a crucial techno-economical parameter of the system. In addition to this, some other ways of its reduction have been suggested, namely, by the enhancement of source neutrons /e.g. by the use of fissionable targets/. The doubling of the burnup and thus extension of the fuel cycle is obtained here without unavoidable otherwise, high initial enrichments and troublesome burnable poisons. In the case of U-Th cycle the existence of the relatively long lived intermediate nuclide (^{233}Pa) can be utilized also for stabilization of the k_{eff} during the time of transmutation.

Summarizing, the advantage of the present symbiotic accelerator-driven transmutation concept managing without partitioning lies in the heightening of systems' safety due to the constancy of k_{eff} with simultaneously fuel self-regeneration and fissile material /e.g. Pu/ abatement. The character of AFR, symbiotic with LWRs and not pretending to be a turning-point in the development of nuclear energy by revolutionary replacement of all the present power plants would rather facilitate smooth launching and deployment of the accelerator-driven nuclear technology.

1. INTRODUCTION

The negative attitudes vs. nuclear energy have not remained without influence also on the directions of its development. Among positive consequences of this well ambivalent social phenomenon one can count the search for new solutions in the very sensitive item coupled with

nuclear energy - the nuclear waste At current level of world deployment of nuclear energy /ca 340 GW$_{el}$ installed civilian capacity/, the yearly amount of actinides in the spent fuel is about 10 000 metric tonnes If one completes this value with the military spent fuel /i e from submarines and reactors supplying weapons-grade Pu for warheads/ the above number becomes rather conservative The overall projected cumulative quantity for the year 2000 is 250 000 t The danger that poses such amount of high level waste containing 5 000 tonnes of fissile material has been recognized already pretty long ago As a result, just this question of safety has been motivating decision-makers and scientists to deal with the pertinent problems and has nearly totally over-shadowed another important premiss that is also substantiating the search for more far-sighted solutions of the nuclear waste problems than simply a geological storage This overlooked aspect is the enormous energy content of actinides, equal to a striking number $8 \cdot 10^{20}$ J in the spent fuel produced annually in the world i e more than a twofold yearly total world energy consumption One should consider as a duty of present generations not only to protect the future ones from the thinkable harmful influence of the waste just being left to them, but still more to assure for them its future use as a precious energy source In this view, *any irreversible disposal of actinides becomes* at least *deeply unjustified*, if not simply mischievous to our descendants

In addition to this the approaching exhaustion of organic fuels and its foregoer - the price shock must not be ignored It is true that we have been accustomed for decades to hear that the world resources of oil would be exhausted within about 30 /or some more/ years and we stopped to believe in such warnings long ago since these decades were passing by and the perspective of exhaustion has remained that remote as before Nevertheless, independently of our wish /or rather against it/ one day this gloomy forecast *has to* prove true The world seems to forget that observed extension of static lifetime of reserves resulted from the continuous rate of discoveries of new deposits, what was conditioned by the lack of knowledge of the earth crust and by the imperfection of the exploration methods Both these determinants are much less valid now, while the growth in consumption still can be well approximated by an exponential leaving only the value of its index (ranging from ca 2 to 4%/yr, the last value regarding natural gas) for discussion The impact of exponential growth on the lifetime is presented in the Fig 1
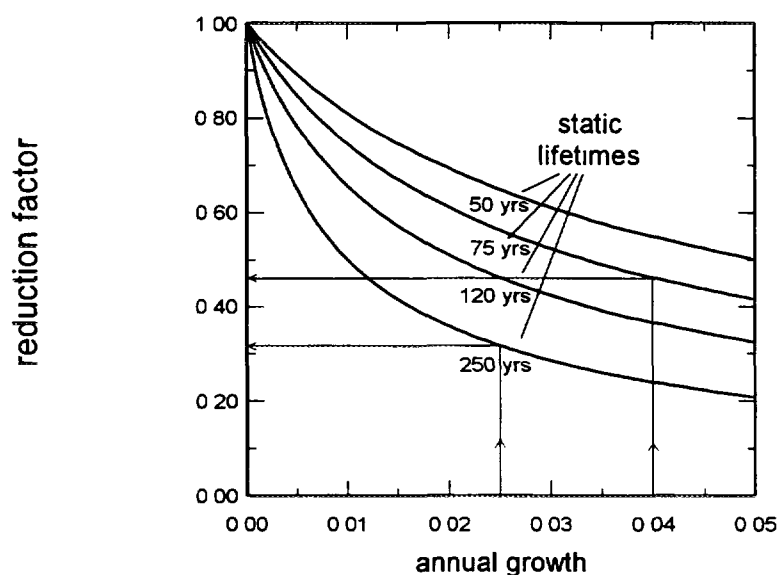

Fig 1 Reduction of static reserves lifetime for exponential growth of consumption

The drastic shortening of the reserves lifetime (e.g. from 75 to ca. 35 years, for 0.04 growth rate) shown above signifies that the static lifetime of reserves is hardly longer meaningful. Besides, it should not be understood as a strict prediction. The lifetime to be really expected is yet longer, since the approaching difficulties in satisfying the increasing demand will result first in curb of the fuels production, whereas simultaneously some additional reserves will be proven. However, the inseparable associate of endangered supply of any commodity is the price increase. It is very difficult to foresee already now when such a drop in production of hydrocarbons and its follower - the price jump occur. Nevertheless, at present trends in the world energy market indicating a constant increase /ca. 2.5 % yearly/ due, among others, to the unquenchable and inevitable tendencies in the developing countries, the time to elapse before the foreseeable rapid increase in energy prices is to be expressed at most in a few decades. In turn, the maturing of a new, advanced technology is a long process that cannot be accelerated whatever expenses would be born. Similarly, its deployment is not rapid too, in view of, among others, the time-consuming process of gaining and sharing experience needed for creation of a community of specialists in this new field.

Therefore, to prepare in time - for our descendants - a solution of this difficult situation caused by human insatiable energy hunger seems to be our moral obligation. The development of Accelerator-driven Transmutation Systems (ATS) [1-3] is a right step in this direction.

2. DESIGN PREMISES OF A TRANSMUTATION SYSTEM

The impact of a 1 GeV proton upon a heavy (e.g. Pb) target results in about 30 /or more in fissionable targets/ fast neutrons that can induce desired nuclear transmutations of various nuclear materials. These are: actinides - fertile and/or fissile ones, fission products or spent nuclear fuel [4,5], which is the main source and itself a part of nuclear waste. The recent rebirth of interest in the transmutations [1-3,6] is mainly due to a significant progress in the accelerator technology permitting us to expect sufficient target currents and thus neutron yields only now assuring reasonable reaction rates.

In general the two primary objectives of transmutations can be seen:
1) The incineration of radwaste of various kinds
2) The supply of fuel for energy production

These objectives are aimed at the development of a new, safer nuclear energy, though the degree of breaking out changes associated with, can be diverse. Besides, it should be noted that both aims are joined with very deep physical links in spite of certain discrepancies resulting from different priorities assigned to the above mentioned targets of transmutations. Nevertheless, apart from the primary objectives, the premisses of design of the transmutation systems unfortunately are in part inherently antagonistic. This difficulty is on the one hand due to the above different fundamental motivations and to the different hierarchy of factors on the other. It seems lie deep in the multidimensional nature of the problem. Thus, certain conflicting tendencies are rather obvious, for instance, in the relation between the economy and safety of any technology. The increased requirements of safety draw behind additional investments and heighten the maintenance cost to a level that may prove prohibitive. Also the fundamental physical properties of nuclear waste (content of fissile materials, high radiotoxicity) occupy an over-exposed place in the social and political rank of risks. All the above signifies that the final shape of the concept must be a compromise between contradictory demands and thus a unique undisputable solution can hardly be expected.

The connection between selected design premisses and certain physical properties of the transmutation system will be discussed below. But first, one should recognize the need of efficiency of any undertakings, in awareness that just the economical incentives are motivating most human actions at social level, not mentioning the business and political ones.

Thus, in order to cope with the tasks that have arisen, the studies should first of all, though - by necessity - rather roughly, deal with economical aspects of ATSs.

2.1 Economical Aspects

Unfortunately, in fact till now, there are no unquestionable and complete economical evaluations of existing energy options, not to mention the emerging ones, thus, a simplified analysis seems justified, like the approach presented below.

As a general minimum condition of economy of any enterprise the compensation of expenses by its income can be taken. Besides, one should not forget that the contribution of uranium raw material to the cost of electricity from nuclear power does not exceed recently several (3-4) per cent. In the following reasoning the 1 GW_{el} yearly sale of electricity has been considered as a unit of the income/expense. Next, the following division of LWR costs: 50% - capital, 35% - fuel cycle and 15% - operation and maintenance, has been assumed [7]. As for the ATS, its capital and O&M expenses respectively, equal to a triplicate and a fourfold of the ones of LWR, have been postulated. Such a picture indicates that the ATS electricity sale can only hardly cover a half of its expenses, thus the other half should be compensated from other income source. According to the present proposal (see Section 3.) it can be brought by taking upon the troubles /and means/ spent for the fuel cycle at several LWRs. It signifies supplying them in return with fuel and freeing of the nuclear waste.

In any case, however, alone the production of energy in accelerator-driven systems cannot bring an income, which might compensate immense investment cost of the accelerator, well exceeding the one of LWR. Thus one can conclude, *that Accelerator-driven Systems must be confined neither to solving the fuel cycle questions alone nor to mere energy production.*

2.2. Socio-Political Acceptability

The mentioned above negative attitudes vs. nuclear energy as well as the politics in general exert influence also on the research in the respective field. At least for pure pragmatic reasons they must not be neglected. The concept of accelerator-based nuclear energy is a trial to soften these anti-nuclear phobias. Therefore, among others, to present it in the convincing way is an important task. Unfortunately, the nuclear community does not seem to be sufficiently familiar with such a deeply interdisciplinary questions. In addition to it, by its proper nature the accelerator-based nuclear energy cannot appear in the eyes of the public as an option having nothing in common with radioactivity, fissile materials, etc. in spite of the fact that it is genuine step forward in that direction. According to the saying: "one cannot have eaten the cake and still to have it" - a rebours - *there is no possibility to destroy nuclear waste and not to manage with it at all.* On the other hand within the accelerator driven technologies there are solutions more or less protected against a diversion of nuclear materials. Certainly the one proposed herewith, as a partitioning-free one belongs to the non-proliferation oriented.

2.3. Selected Physical and Technical Determinants

While designing an accelerator-driven nuclear transmutation system aimed, for instance, at efficient transmutations of actinides, one must never forget that since only fissioning is recognized as a definitive way of getting rid of these, *the rate of actinide transmutation per power unit is absolutely limited.* The process, thus inseparable from energy release, has a well fixed intensity (per energy unit) and thus its rate is directly determined by the size i.e. the power of the device. Having then left little to improve in this aspect, the optimization efforts should be concentrated on other factors i.e. first of all on the safety requirements and the optimum use of the accelerator. Leaving the safety aspects to be discussed in the section 2.4, let us start from the optimization aimed at the efficient reduction in the accelerator current i.e. in the main factor determining its immense costs.

A radical drop, at unchanged power of the system, down to the level achievable with the use of a cyclotron, thus permitting us to replace the incomparably more expensive linac with the former, would be a true success. /The maximum feasible cyclotron currents seem to be definitely limited to values only slightly exceeding some ten mAmps, because of the space charge effects/.

2.3.1 Energy Gain of the ATS

The dependences between the energy gain G of the ATS and the accelerator current I, based upon the formulas discussed in the Appendix are illustrated in Fig.2.
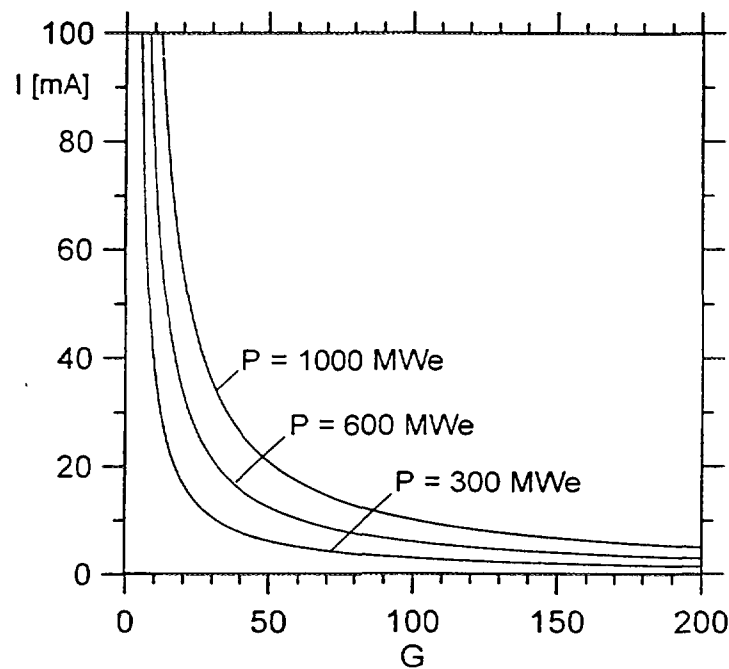

Fig. 2. Accelerator current necessary at given net power of the ATS vs. its energy gain
(accelerat. voltage U = 1 GV; accelerat. effic. = 0.5; Carnot effic. = 0.4)

As it can be seen in the Fig.2, the necessary accelerator currents ranging attractively below the ca. 10-20 mA, demand very high energy gains of the system. Unfortunately, the low currents require high energy gains of the transmutation system drawing behind high values of neutron multiplication factor, that, in turn, threat with undermining the safety of the system (see 2.4.). Therefore, a clear particular objective appears: loosing the strict links joining the energy gain of the system with its k_{eff} or, more precisely - a maximization of the number of neutrons independent of k_{eff} i.e. preceding the k_{eff}-based multiplication chain. To a reasonable extent this aim seems attainable in view of feasible enhancement of the source neutrons for the further fission chain reaction. These are the accelerator beam generated, spallation and primary fission neutrons. An increased number of these neutrons relaxes our demand for high neutron multiplication, thus for less safe k_{eff}. /One should always consider inevitability of step-wise changes in this value, following, for instance, a necessary fuel shuffling or simply a system reloading, thus somewhat lower values of k_{eff} seem by far more desirable/. The increased number of fissions /caused by more abundant source neutrons/ in the system, assuring a higher energy gain can prove sufficient for a reduction in the accelerator current. The number of k_{eff}-independent neutrons can be enhanced provided that:

1) the number of source neutrons /per proton/ is maximized - e.g. by the use of fissionable materials in the target;

2) the number of fissions /per neutron/ induced directly by source neutrons is higher than the one per average system neutron [1].

The distribution of both fissile and fertile materials in the system with respect to source neutrons will determine the importance of the latter effect. However, one is limited by the admissible power density /and its peaking/ in selecting the system arrangement and composition. For instance, a superfluous accumulation of fissile material directly in the target may easily lead to an excessive energy release in there. Summarizing, the question is what values of k_{eff} would be needed that the necessary energy gains of the system be achieved.

The approximate shape of the connection between the ATS energy gain and the - k_{eff} is shown in the Fig.3. The fuzziness of the curve results from the dispersion of the values of several significant parameters corresponding to different variants of the ATS. These are: lead vs. fissionable targets and thus the number of source neutrons n = 30 or 70 respectively and the projectile direct energy release $w_p{}' = 800, 3300$ MeV. /Obviously, intermediate values of these parameters are even more realistic/.
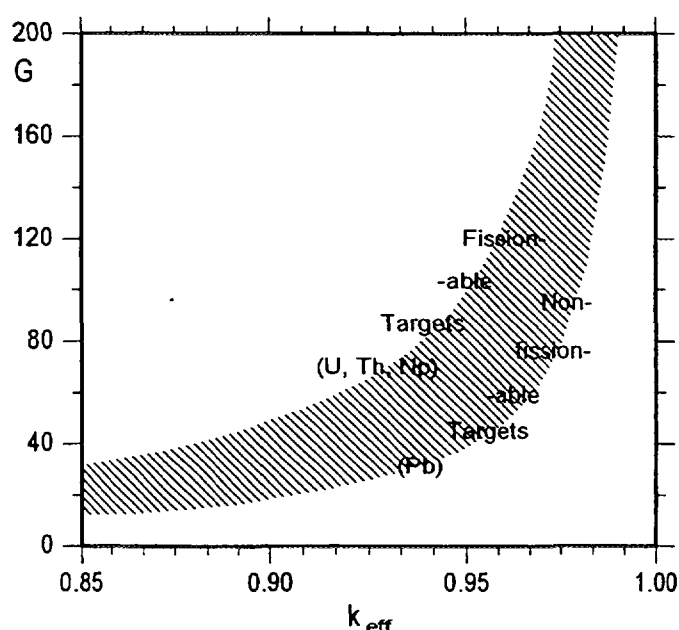

Fig. 3. Energy Gain of the ATS vs. its neutron multiplication factor

On the basis of the Fig.3 one can conclude that the sufficient energy gains seem achievable. The higher gain values require fissionable targets that simultaneously assure the two objectives: significant additional energy release independent of the fission chain (i.e. of the k_{eff}) and thus the number of k_{eff}-independent neutrons. Whether such target brings also a higher multiplication of the first generation of neutrons (that need not be necessarily identical with the mean k_{eff} of the assembly) is not certain. To find out, whether it is higher than the average requires further, more discerning evaluations. Nevertheless, such positive effect cannot be excluded although the important constraint should not be forgotten - the admissible nuclear heating. The distribution of energy release in subcritical systems is obviously much more peaked than that in the exactly critical systems, thus just the maximum power density in the central zone of the system can restrict its power (i.e. size) as a whole. Anyway, in general, a number of such calculations has to be carried out in order to determine the accessible areas in the parameters phase space. The links between them should be investigated for identifying possible correlations or anticorrelations. It has to be ascertained whether the desirable values of some important parameters do not draw behind inadmissible values of another ones. Particularly prohibitive demands may issue from the requirements of technical feasibility and

142

safety In view of the former the variant of transmutation aimed at the fuel self-sustention of the AT is worth consideration, as conditioning the independent operation of any nuclear energy unit. The respective constraints are subject of the item below

2 3 2 Breeding vs burning

As it has been mentioned earlier, the fundamental objective of the system can be confined to the destruction of nuclear waste or can include also transmutation of fertile nuclides into fissile ones According to the selected purpose the number of bred fissile nuclides per one destroyed fissile nuclide i e the conversion ratio C_r of the system should be different In the case of actinide destruction oriented systems their C_r need not be high since the regeneration of the fissile actinides may not be necessarily desirable Yet, there are following arguments against C_r be too low

a) the need of abatement of non-fissile actinides
/the transmuting them into fissile ones is usually the way of most efficient incineration/
b) the need of stabilizing the energy gain of the system,
/otherwise it might fast reduce, due to burnup not compensated by the breeding/

Anyway, independently of the option, the neutron balance in the system imposes certain constraints over the pair of very important parameters of the ATS, namely, its k_{eff} and C_r (Fig 4)
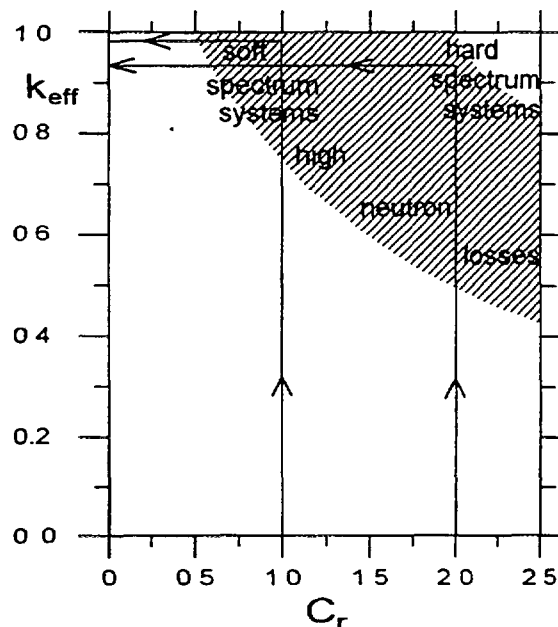

Fig 4 Approximate relationship between neutron multiplication factor and fuel conversion ratio in subcritical systems

As one can expect and see in the Fig 4 there is no strict trade-off between the neutron multiplication and conversion ratio of externally driven subcritical systems Both k_{eff} and C_r are subject to a number of other factors characterizing the system that can differ significantly in each particular solution Nevertheless, in accordance with intuition the most important constraint is the inaccessibility of the region of high values of both parameters k_{eff} and C_r simultaneously The number of excess neutrons that might serve to various transmutation purposes is well limited for higher k_{eff}s In other words one can have either high neutron multiplication (and energy gain) or high conversion ratios Obviously also neutron losses (or other uses) significantly reduce achievable values of k_{eff} and C_r (Among other uses one can

143

see the necessity of devoting more than one neutron for transmuting those nuclides that require more captures to become fissile, e.g. ^{237}Np). In conclusion, a more scrupulous search of the parameter phase space with pertinent neutron transport calculations for identifying the accessible areas seems useful.

2.4 Some Safety Aspects

One of the most important factors that might favorize the ATS vs. other options of emerging nuclear energy systems is its supposed advantage in the field of safety. This optimism is based upon subcriticality of ATS, since the other sources of hazard such as the presence of radiotoxic materials in general cannot be avoided. It signifies thus, that if the ATS superiority is to be genuine the elimination of the danger of supercriticality should be guaranteed with the sufficiently high certainty. In this view one of the most important postulates is to keep the neutron multiplication factor k_{eff} below a certain value recognized as a safe one, all the time during transmutation. How to achieve it - is suggested below.

2.4.1 Balanced transmuting

The state k_{eff} = const can be satisfactorily assured, provided that the amount of the effective fissile inventory in the system is kept approximately unchanged. This apparently severe requirement can be relatively easily achieved while:

- having the fissile material in the liquid form [2] (that allows for a continuous control of the fissile content in the system;
- diversifying the media filling the assembly.

In low enriched media /as e.g. LWRs' fuel/ for hard neutron spectra, a higher rate of fissile breeding than burning can be expected. This signifies, however, that there, like in fast breeders, a net breeding should be expected, thus making the k_{eff} to grow continuously over the transmutation time. But, if an additional amount of fissile material is introduced into the system, its increased burnup will compensate the breeding in the fuel to be regenerated. This may be done with any fissile material at disposal: e.g. a weapons grade Pu or a civilian one. If neutronic abatement of the Pu is to be considered at all, at least the separation of uranium must be carried out, in order to prevent its transmutation in Pu in the neutron flux.

The other solution suggests the diversification of the neutron spectrum within the system, i.e. in the two different zones: one of harder spectrum and another of a softer one. In this case the burning in the softer spectrum would compensate the breeding in the harder one, at comparable enrichments in both zones. On the other hand one should be aware of the limitations of the flux shaping esp. within moderate volumes. Softening of the flux cannot be restrained to a strictly defined areas, thus the differences in the neutron spectra cannot be arbitrarily great.

In any case one must not forget that both the processes - fissile incineration and breeding are not uniform in space and time, therefore, they occur at different rates within the system volume over the whole transmutation time. This effect bringing some more difficulties (i.e. indicating that a simple point model of the ATS is a very rough approximation) provides us with an additional field of manoeuvre - the shuffling of the fissile material within the system volume. Thus, the remedies for stabilization of the neutron multiplication factor are:

- the fissile displacement towards the areas of weaker fissioning flux - in the phases of increasing k_{eff} i.e. during the dominance of breeding processes;
- in the opposite direction while the incineration of fissile materials is prevailing.

In addition to the above one should expect that the shuffling of actinides in the system anyway may prove unavoidable as a result of nonuniformity of transmutation processes. The

local transmutation rates vary in a very significant way. An earlier attainment of the desired transmutation objectives in some areas than in another ones directly draws behind the necessity of removal of the transmuted material and its replacement with a fresh one. Since one can hardly expect that the parameters of the latter will not deviate from those of the stuff just removed, some shuffling for compensation of this difference seems always necessary.

The additional complication is due to some time-dependent processes mentioned below.

2.4.2 Fissile fuel trajectories

The changes in composition of the media filling the system, occurring within the scale of days/months, that may be called in short fuel trajectories, are the source of important effects. They can result from the existence of an intermediate nuclide, of similar decay time, between the primary fertile one and the final fissile one, in the chain induced by neutron capture in the fertile nuclide. As a result, the fissile nuclide appears in the system with a certain delay, what may prove beneficial, though not free from some risk. For the usual U-Pu fuel cycle (the intermediate nuclide - ^{239}Np, $T_{1/2}$ = 2.4d) the effect seems not a very realistic because of the extremely high fluxes needed. Approximate evaluations indicate the values of the order of 10^{16} n cm^{-2}·s^{-1} by rather hard spectrum. In contrast with it, this phenomenon can be observed for the Th-U cycle distinct by the $T_{1/2}$ = 27d of the intermediate nuclide ^{233}Pa

This effect is not to be neglected, since one of the key safety questions to be decided while designing the system, is the choice of maximum admissible k_{eff}, depending on its behaviour during the transmutation processes. The changes in fissile content in the system (the ones after its shutdown including) directly affect the level recognized as the safe one. The case of Th-U cycle, is to be discussed in more detail and illustrated in the Fig.6., section 3., the subject of which, among others is the transmutation of thorium based fuel.

3. SPENT FUEL REJUVENATION CONCEPT

The indications issuing from the above analyses have lead us to the variant of transmutations presented below The fertile nuclides contained in the *spent nuclear fuel* can be transmuted into fissile ones, then these being fissioned are producing energy while some radioactive fission products are transmuted into stable ones. The feasible in this way re-use of spent fuel reduces the environmental impact /per produced energy/ of the nuclear power by simple doubling the amount of energy extracted from the fuel If simultaneously can be carried out a destruction of actinides accompanied inseparably with the release of energy contained in these, the idea appears unquestionably environmentally benign

Therefore, this proposal is an attempt to demonstrate the concept of Accelerator-driven Fuel Regenerator (AFR) in belief that it may help in the deployment of nuclear energy In the present concept, the AFR being used for rejuvenating of the burned-up fuel is perfoming a triple function

- the "augean" one of a symbiont co-operating with several fission reactors for transmuting (fissioning) actinides and (partly) fission products generated in there, i e for depoisoning and disactivating the nuclear reactor waste,
- of a device producing net energy from fissions,
- of a breeder of fissile material in the spent fuel from symbiotic LWRs

In the concept of rejuvenation of spent nuclear fuel in the AFR significant additional (to electricity production) gains can be expected thanks to the two benefits worth emphasizing, in view of their positive environmental effect [5]

1 Direct reutilization of unburned fissile nuclides contained in the fuel,
2 Avoidance of one fuel cycle

Therefore, it should noted that because of the radiation damage in the cladding and fuel material no more than single regeneration of spent fuel is assumed feasible. It signifies that the further treatment of the fuel after the second campaign remains an open question. It seems that no option can be excluded at this moment, even the final disposal, though, as it has already been stated, *the irretrievable getting rid of actinides is dissuaded* here. Therefore, some form of interim storage of spent fuel seems more advisable, as preserving the possibility of its later use. Besides, any other solution of nuclear waste problem can be applied too.

On the basis of the considerations in the paragraph 2.1 one can see that solving the problems of fuel cycle of a certain number (say, 3) of LWRs with the AFR is essential for the latter to be economic. It should be noticed that - conservatively - no gains from the abatement of additional fissile material (eg. Pu) have been assumed here. In effect, the economy of the device is solely based upon the production of energy and saved cost of fuel cycle of symbiotic LWRs. Being more exact, the number of cooperating units S can be different, e.g. much higher, in the case of better utilization of the fuel in these reactors (Fig.5).
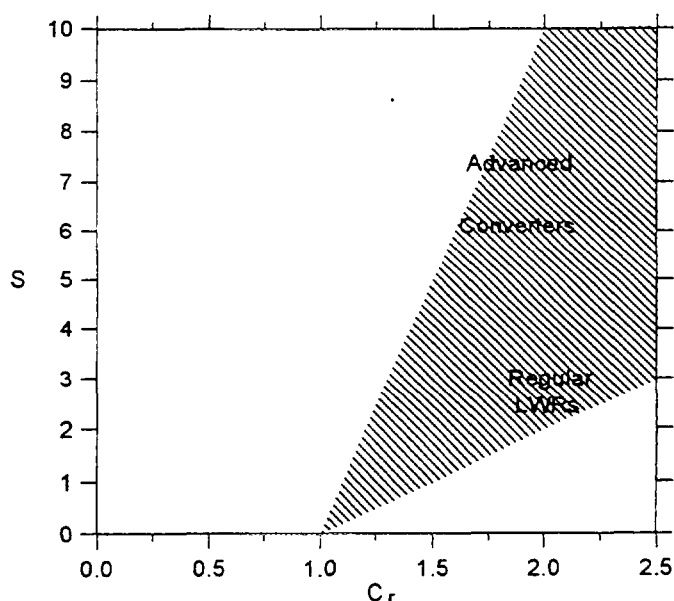

Fig. 5. Number S of LWRs supported by the AFR vs. its fissile fuel conversion ratio C_r

It is worth to notice that alone the number of LWRs supported by the AFR may not influence the economy of the AFR functioning as a part of symbiotic system. This conclusion stems from the fact that the reactors of higher conversion ratios require less fuel thus, approximately (i.e. neglecting the MA), there is little difference whether the given rejuvenation yield regards larger or lesser number of client units. However, one obvious limit may appear, namely the material damage of the rejuvenated fuel. For advanced converters the burnup may prove so high that no reuse of this fuel be possible. Thus, in this case the application of the present transmutation option would be confined to the servicing of regular LWRs.

Obviously, the most basic factor conditioning the independent operation of a nuclear energy device is its fuel self-sustaining. This, in turn, would easier occur in hard neutron spectra, since there a higher fissile breeding rate than a burning one can be expected in low enriched media /as e.g. spent fuel/. In this conditions, however, an additional burning of fissile material is needed for compensation of the breeding in the fuel to be regenerated. Such a need appears, since in addition to the constancy of k_{eff} over the transmutation time to keep the energy gain G of the system constant is desirable, in order to have always the optimum load of accelerator. As it has been already mentioned, either addition of available fissile

146

material, for instance, Pu, e.g. the one recovered from the burned-up LWR fuel or adequate shuffling of the fissile material in the system (see 2.3.2) may stabilize the energy gain. However, according to the considerations regarding time-dependent effects (2.4.2) in the case of thorium based fuel, they should not be neglected.

3.1 The Th-U Cycle Spent Fuel

The half-life time of the Th-U cycle intermediate nuclide ^{233}Pa (27 d) proves sufficiently long that the desirable transmutation level be achieved before its decay. Whether such effect is significant or not is decided by the maximum admissible density of nuclear heating in the given matter. It is just this value that puts the limit on the admissible flux which, in turn, determines the transmutation time needed for fuel rejuvenation (e.g. achieving of the desired enrichment level). Therefore, the Th-U fuel demonstrates some particular properties that provide us with additional possibilities of control of the fissile material content in the system during transmutation (Fig.6).
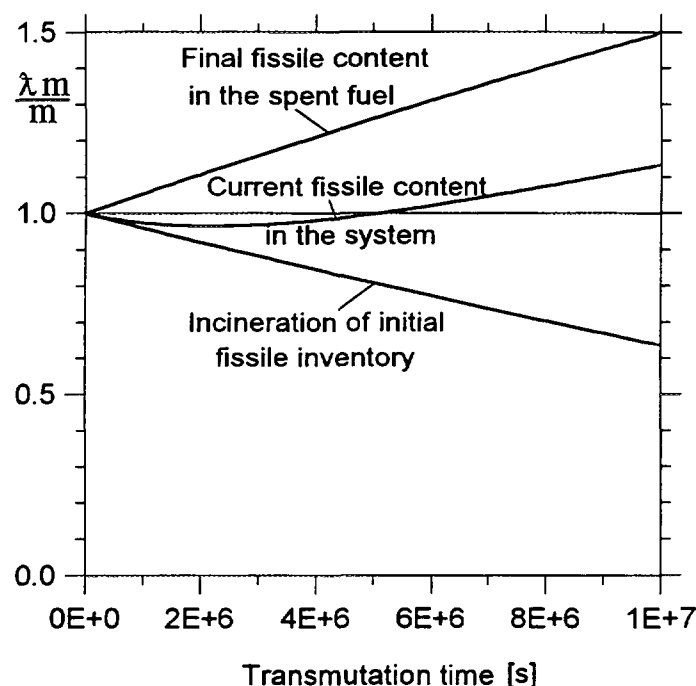

Fig. 6. Example of the fissile content trajectory for the Th-U cycle in the AR .

The fuel trajectories in the Fig.6 indicate that within the ca. 3 months of transmutation the result of delay in the buildup of fissile material just being bred can significantly compensate the prevalence of breeding over burning process. The effect of reducing the changes in the current content of fissile material in the system at simultaneous spent fuel regeneration and incineration of added fissile material is shown. This effect can well soften the need of shuffling the fissile material for stabilizing the system k_{eff} and gain. At the same time, however, the increase in fissile content resulting from decay of ^{233}Pa after the shut-down of the AFR gives rise to adequate measures against the hazard of criticality. The picture as a whole demonstrates the possibility of achieving at the same time the conflicting goals of transmutation in the AFR i.e. incineration and breeding of fissile materials. In addition to the above it can be mentioned also the flexibility of the use the above effect. For instance, if the extra breeding is considered undesirable, then not only the amount of fissile material can be used for its balancing with incineration but also the intensity of transmutation. A lower neutron flux would slow it down to last, say, 1 year, after which all the ^{233}Pa is decayed into ^{233}U.

Another problem determining the performance of the AFR is the neutron flux distribution thus also the power density. Their non-uniformity already seen in reactors, is still more accentuated in subcritical systems driven with centrally placed neutron source. The solution of this question seems lie in dispersion of the source over larger volume (Fig.7.).
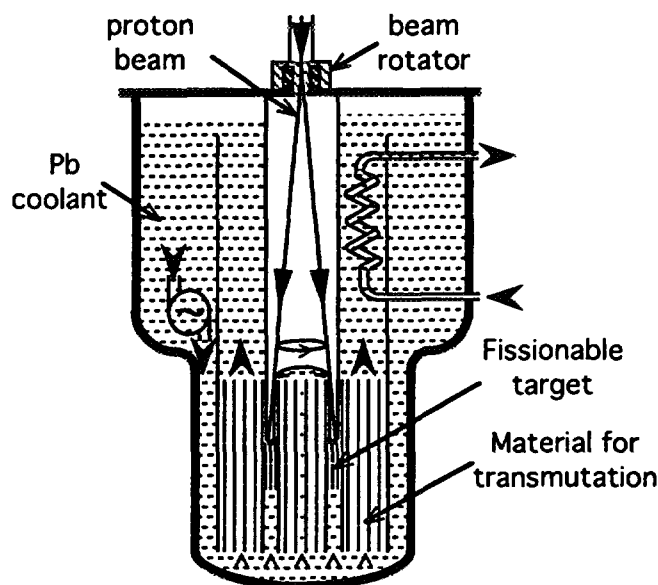

Fig.7 A sketch of the Accelerator-driven Fuel Regenerator with shell actinide target

As one can see in the Fig.7. the proton beam is spread over a cylindrical shell target. Such concept has the advantages of flattening the peak of power density in the target itself and its immediate neighbourhood by simple increasing of the volumes in question. It can have also a positive effect in view of the radiation damage occuring in this area. The reduction of the damage may result in admiting a pebble bed type solution of continuous feeding the target (optionally containing actinide spheres cooled) with liquid lead. The respective evaluations as well as the preliminary transport calculations are just being carried out. The exact calcuations require, however, adequate geometric description in a 3-D code coupled to the burnup one and a consideration of high energy neutrons component (>15MeV), also in view of their contribution to the radiation damage, as well as completing the yields of fission products.

4. CONCLUSIONS

It seems that first, the *symbiotic character of AFR*, as *based upon existing LWRs* - thus, reducing the overall investment effort, is worth underlining. It does not urge to a revolutionary turning-point in development of nuclear energy by pretending either to replace all the present power plants or by solving definitively problems of their waste. Rather to the contrary, its *strict cooperation with LWRs*, that do not seem yet threatened with extinction within a foreseeable future, *would rather facilitate smooth launching and deployment of accelerator-driven technology*.

Another advantage of the proposed AFR concept is the fulfilment of fundamental condition of systems' safety i.e. the quasi steady k_{eff} over the whole fuel rejuvenation campaign at simultaneous: fuel self-sustention of the system and fissile abatement. Such a stabilization of k_{eff} in subcritical systems allows, in turn, for its higher values and thus higher energy gains i.e. for safe reduction of the accelerator current. The choice of *fire resistant, low-pressure, high boiling-point and tritiumless lead coolant contributes also to the safety of the system*. The rejuvenation of spent nuclear fuel by means of neutron induced transmutations according to *the*

148

present concept, needs neither mechanical nor chemical intervention into the fuel. Thus, the concept itself does not draw behind directly any partitioning, and therefore its nonproliferation-oriented character also deserves emphasizing.

The other suggested way of decreasing the accelerator current by *enhancement of source neutrons in shell actinide target* can positively focus the efforts devoted to the optimization of design. A certain advantage of the Th-U cycle due to ^{233}Pa has been indicated.

Nevertheless, since the structure and composition of the assembly are far from being optimized, still a wide margin for improvements in its performance remains, whereas many technological questions (radiation damage, corrosion, cooling etc.) are far from being solved.

Summarizing, on the basis of presented discussion and evaluations, certain simplifying assumptions notwithstanding, the concept of fissile fuel rejuvenation, is revitalized. Therefore, the concept of Accelerator-driven Fuel Regenerator should help in deployment of the nuclear energy as being just a nuclear technology that reduces the nuclear environmental impact, what can have a positive psychological and thus social significance.

APPENDIX

The interdependence of parameters of accelerator-driven transmutation system is presented. These are: the accelerator current I and neutron multiplication factor k_{eff} - both necessary for a given net electric power production P_{el}, fissile fuel conversion ratio C_r, and the number of symbiotic LWRs (support ratio) S. Let us start from the energy gain G of the system:

$$G = [w_p{}' + n(Q_f/v)k_o/(1-k_{eff})]/w_p \qquad (1)$$

where

$w_p{}'$ = energy released in the target per proton
Q_f = fission energy
n = number of source neutrons per proton
k_o = number of first generation neutrons per source neutron
v = mean number of neutrons per fission
w_p = proton energy

while the accelerator current is:

$$I = P_{el} / [U(G\eta_{th} - 1/\varepsilon_a)] \qquad (2)$$

where

U = proton accelerating voltage
η_{th} = efficiency of thermal-to-electric energy conversion
ε_a = accelerator efficiency

In a system where one can assume that the contribution of neutrons from non-fissile materials (i.e. ~ fast fissions) compensates the losses (i.e. parasitic absorptions and leakage) the conversion ratio approximately is:

$$C_r = \eta\varepsilon (1-\ell)/k_{eff} - 1 \qquad (3)$$

or

$$k_{eff} = \eta\varepsilon (1-\ell)/(C_r + 1) \qquad (4)$$

where

η = mean number of fission neutrons per absorption in fissile nuclide
ε = increase in the number of neutrons due to fast fissions
ℓ = fraction of lossed neutrons

And finally the number of reactors provided with fuel bred in the above system is:

$$S = (C_r - 1)/(1 - C_{rL}) \qquad (5)$$

where

C_{rL} = fissile fuel conversion ratio of symbiotic reactors

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SOME RESULTS ON DEVELOPMENT, IRRADIATION AND POST-IRRADIATION EXAMINATIONS OF FUELS FOR FAST REACTOR-ACTINIDE BURNER (MOX AND INERT MATRIX FUEL)

V. POPLAVSKY, L. ZABUDKO,
L. MOSEEV, B. ROGOZKIN, I. KURINA
Institute of Physics and Power Engineering,
Obninsk, Russian Federation

Abstract

Studies performed have shown principal feasibility of the BN-600 and BN-800 cores to achieve high efficiency of Pu burning when MOX fuel with Pu content up to 45 % is used. Valuable experience on irradiation behaviour of oxide fuel with high Pu content (100%) was gained as a result of operation of two BR-10 core loadings where the maximum burnup 14 at. % was reached. Post-irradiation examination (PIE) allowed to reveal some specific features of the fuel with high plutonium content. Principal irradiation and PIE results are presented in the paper. Use of new fuel without U-238 provides the maximum burning capability as in this case the conversion ratio is reduced to zero. Technological investigations of inert matrix fuels have been continued now. Zirconium carbide, zirconium nitride, magnesium oxide and other matrix materials are under consideration. Inert matrices selection criteria are discussed in the paper. Results of technological study, of irradiation in the BOR-60 reactor and PIE results of some inert matrix fuels are summarised in this report.

1. Introduction.

Utilization of fast reactors to burn actinides is an effective way of resolving the long-lived nuclear power waste issue. In Russia extensive study of fast reactor cores which can efficiently burn plutonium and minor actinides (MA) is under way in order to demonstrate the possibility of actinide utilization in BN-600 and BN-800 reactors.

To achieve the purpose of effective actinide burning the design of the breeder reactors BN-600 and BN-800 need to be modified. Modification of BN-600 and BN-800 breeding cores with traditional oxide fuel proceeds in two stages. The first one is connected with the removal of radial and axial breeding zones from the core and their replacement by non -breeding blankets. In this case the breeding ratio 0.73., and fast reactor becomes a plutonium burner. The second direction is connected with the Pu content increase.

Studies performed have shown principal feasibility of the BN-600 and BN-800 cores to achieve high efficiency of the Pu burning, up to 360 kg/year, when MOX fuel with 45% Pu is used/1/. Technological study on development and fabrication of MOX fuel with high Pu content is under way now aiming the fuel manufacturing for the BOR-60 irradiation. Important experience on irradiation behavior of high Pu oxide fuel was gained as a result of the irradiation of two BR-10 core loadings with PuO_2 and post irradiation examination (PIE) of spent fuel pins in the hot cell. Irradiation and PIE results are summarized in this report.

Use of the inert matrix fuel without U-238 will provide the maximum burning capability as in this case the conversion ratio is reduced to zero. For the core with inert matrix fuel the fuel materials are considered on the base of zirconium carbide, magnesium oxide, aluminium nitride and some others. Calculative investigations of the BN-800 type core with different types of matrix fuel are carried out. The fundamental technological investigations are necessary in this direction on the selection and characterization of the new fuel materials.

2. MOX fuel.

2.1. Development and fabrication of MOX fuel with high Pu content.

During the 1994 the laboratory study of MOX fuel with 45% Pu fabrication was carried out in the VNIINM, Moscow. The fabrication techniques of chemical coprecipitation and mechanical mixing

of oxide powders were investigated, the physical, chemical and technological properties were studied. It was shown that as for mechanical mixed so for coprecipitated powder it is possible to obtain the homogeneous fuel from the powder and the solid solution after the sintering. Adjusting some fabrication process parameters it is possible to fabricate pellets with rather wide range of density values (9.5 - 10.7 g/cm^2). The pellet solubility with high Pu content (45%) was studied also. It was found that it is necessary to increase the solution period significantly comparing with the 30% Pu MOX fuel. Presently these investigations are continued. Several fuel pins are manufactured to be irradiated in the BOR-60 reactor.

2.2. Irradiation experience.

The set of experimental subassemblies (SAs) irradiations with MOX pellet fuel were organized in BOR-60, BN-350, BN-600 reactors with an aim to study fuel performance. Besides, big experience was gained with MOX vibropacked fuel in BOR-60 (Table 1).

TABLE 1. MOX FUEL IN BR-10, BOR-60, BN-350, BN-600 REACTORS

Reactor	BR-10-PuO$_2$	BOR-60	BN-350	BN-600
Pellet fuel				
Pin number	3300	400	1800	1524
Max. burnup % at.	14.	24.*	10.8	10.5
Max. linear rating, kW/m	17.	50.	48.	48.
Vibropack fuel				
Pin number	-	12800	254	762
Max. burnup % at.	-	28.*	7.2	9.8
Max. linear Rating, kW/m	-	52.	48.	48.

*) The irradiation was arranged in special dismountable assembly.
The Pu content in MOX fuel irradiated was less than 30%. All fuel pins were intact.

First results on irradiation behavior of oxide fuel with high Pu content (100 %) was obtained in BR-10 where two core loadings with PuO$_2$ were irradiated. Principal design and operational parameters of BR-10 fuel pins are shown in Table 2.

It should be noted that in the pin design of the first core loading the fission product gas plenum was not foreseen that time because of both a small design experience and an inadequate knowledge about the fuel behavior under irradiation. As a result, the fission product gas pressure reached 30 MPa and that was the reason of fuel leakage.

Ten SAs at the peak burnup values from 2.2 to 6.7% at. were selected for PIE. Failed fuel pins were found in six SAs. Two of these 6 SAs at burnup 4.9% at. contained pin claddings with cracks. Each of the other four SAs, at burnups of 5.6, 6.1, 6.6, 6.75% at., contained two or three pins, failed at its withdrawal into two pieces with fuel bits on them.

From the beginning of the second core operation, because of manufactured microcracks in one or several fuel pin claddings, the repeated fission gas releases were detected, however, with no changes in delayed neutron activity in coolant. At burnup of 10.8% at. the failed cladding detection system (FCDS) has registered the first failed pin by delayed neutron signals. With the further core operation using the delayed neutron measurements FCDS has registered the other four signals. After appearance

of delayed neutrons in coolant the radioactivity of fission products in it increased by one order but was not too high to create obstacles to repairs of boxes in primary coolant circuit.

An out-of-reactor FCDS was used and 13 SAs with failed fuel pins were found. Pin cladding defects of the "fuel-coolant" type were found in three of them.

TABLE 2. PRINCIPAL DESIGN AND OPERATIONAL PARAMETERS OF BR-1 FUEL PINS WITH PuO_2

PARAMETER	I LOADING	II LOADING
SAs number	83	72
Fuel pins number	1577	1368
Wrapper flat-to-flat size, mm	26.	26.
Wrapper wall thickness, mm	0.5	0.5
Cladding diameter, mm	5.	5.
Clad wall thickness, mm	0.4	0.4
Core height, mm	280.	320.
Gas plenum height, mm	-	105.
Wrapper- clad material	18Cr-9Ni-Ti	18Cr-10Ni-Ti 16Cr-15Ni-3Mo
Maximum burn-up, % at.	6.6	14.1
Maximum clad temperature, C	580.	585

2.3. Results of post-irradiation examination of fuel pins.

For PIE seven SAs at maximum burnup of 1.42, 3.18, 4.62, 5.81, 7.69, 9.21, 12.06% at. with PuO_2 were selected. The following investigations were carried out:

-measurement of cladding diameter changes,
-pin cladding tightness examination by means of gas composition analysis,
-gamma-scanning of fuel pins,
-fuel and cladding microstructure investigation,
-study of mechanical properties of cladding materials,
-analysis of fission gas release rate.

As a result 8 fuel pins with gas leakage were revealed from SAs with maximum burnup of 7.69, 9.21, 12.06% at.

The following conclusions were derived from these PIEs:

-Noticeable gas release was observed at burnups of 4% at. and more.

-Solid fission products were distributed in fuel column according to the neutron fluence and had not evidently migrated along fuel column. No fuel mass transfer in the axial direction was observed either.

-Radial cracks were observed in all cross-sections of fuel column investigated. In many cross-sections the fuel had crumbled out. The central hole diameter was 1.15-1.25 and was similar for all cross-

sections investigated. For the midplane three zones with the distinct types of fuel microstructure were observed: the columnar grains zone, the equiaxial grain zone and unrestructured zone. This microstructure is typical of oxide fuel irradiated in fast reactors.

-Cladding of all fuel pins at maximum burnup levels of more than 4% at. suffered internal corrosion as a result of FCCI during irradiation. FCCI varied qualitatively and quantitatively for different parts of cladding depending on irradiation conditions.
In the lower part of the fuel column no FCCI was observed with the exception of the 5 mm thick layer in which the grain boundaries were sensitive to etching. Near the core midplane the intergranular cladding penetration was 30-50 mm . In the upper part of the fuel column (at burnup of more than 8% at.) the intergranular penetration was 120 mm and followed by a partial dissolving or loss of single grains.
The perforating intergranular cracks in claddings from failed fuel pins were found near the core midplane and above. Besides the perforating cracks the numerous microcracks on the inner surface of the cladding with various penetrations were observed on these cross-sections. For these fuel pins the FCCI penetration was 90-120 mm. On the outer surface of PuO_2 pellets which were taken from the core midplane with burnup 12% at. a complex phase with Pu, Cs, Cr, Pd, Fe, Ni in its content have been found.

At present 32 fuel pins with MOX fuel from 4 SAs of BN-350 and 40 fuel pins from 5 SAs of BOR-60 have been investigated in the IPPE hot laboratory. The fuel pins were examined visually and no failures or defects were found.

The accumulated PIE results are the followings.

-Solid fission products were distributed in fuel column according to the neutron fluence (similar to the UO_2 fuel) and had not evidently migrated along fuel column.

-Gas release increased noticeably with burnup increase after 3% at. Gas release from MOX was almost the same as from uranium dioxide and reached the value of 95% at burnups more than 10% at. Besides the burnup gas release depended on fuel fabrication method , fuel composition, O/M ratio. With the equal burnup values gas release from the coprecipitated fuel was less. Gas release from the fuel with O/M=2. was less than from the fuel with O/M<2. The fuel with lower density retained more fission products , that probably could be connected with the dependence between open and closed porosities. With linear rating increase (37.- 50.W/cm) gas release increased by 15 - 20%.

-MOX fuel had typical microstructure consisting of three zones whose size was basically dependent on linear rating value in cross-sections investigated. The cladding - fuel gap contained fission products. In the outermost part of mechanical mixing fuel pellets precipitates of UO_2 and PuO_2 were found in the columnar grain zone." Sol-gel" fuel was more homogeneous after irradiation , though the phases consisting of fission products were observed in fuel microstructure.
-In the mechanical mixing fuel with O/M =2. the Pu enrichment of columnar grain zone near the central hole was observed (15 - 20% more than the initial structure). In a more homogeneous fuel such type of Pu enrichment was not more than 2 - 7%. It depended on O/M ratio of solid solution. The less O/M ratio was, the less Pu enrichment of central pellet part was. No Pu redistribution along fuel column height was found.

-FCCI depends on oxygen chemical potential as a function of O/M ratio in fuel. Depending on the initial O/M ratio the radial redistribution of oxygen took place during irradiation. With the O/M initial ratio increase and burnup increase the oxygen potential increase was observed especially in the outermost part of pellets that led to FCCI increase. No oxygen redistribution along the fuel column height was observed. Radial oxygen redistribution was seen primarily in midplanes and above (cross-sections with higher level of linear rating).

-FCCI depth increased with burnup, temperature, linear rating, initial O/M ratio increase. Intergranular and matrix type of cladding penetration with microcracks on an inner cladding surface was observed. The chemical compatibility of MOX pellet fuel with cladding materials used was just the same as that of oxide pellet fuel for equal irradiation parameters. No influence of fabrication method and plutonium composition was found.

3. INERT MATRIX FUEL.

3.1. Criteria on inert matrix selection.

The following criteria should be taken into account when inert matrix material is selected:
-workability,
-compatibility with fuel in the total range of operational temperatures,
-high melting point,
-compatibility with clad and coolant,
-irradiation stability (no big size increase, structural integrity and so on),
-thermal conductivity,
-mechanical properties (ductility, strength, high linear thermal coefficient),
-low neutron cross section,
-low activation,
-solubility when reprocessing.

3.2. Development and fabrication.

On the base of above mentioned criteria the list of inert matrix materials was selected to carry out further technological investigations : MgO, MgO+Me (IPPE, Obninsk), ZrC, ZrN, AlN (VNIINM, Moscow).

-PuO$_2$+MgO

On the first stage the work was carried out with Th and U as simulators instead of Pu. The coprecipitatipon process was used to obtain UO$_2$-MgO and ThO$_2$-MgO. The technological route is shown on Fig.1.
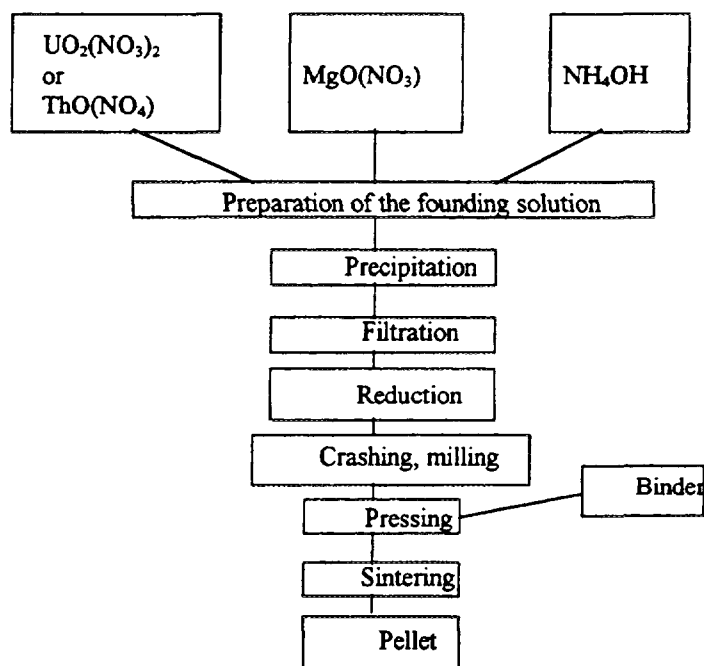

Fig.1. Cercer fuel fabrication route.

The most optimum parameters of fabrication process were chosen. Pellets fabricated have the structure with homogeneously distributed oxides UO_2 (ThO_2) and MgO. Pellet density, thermal conductivity were measured. Several pellets were reprocessed . The optimum solution parameters have been found (without special dopes). Presently the work extends aiming the fabrication of 2 -3 fuel pins with PuO_2 - MgO for BOR-60 irradiation.

-PuN+ZrN, PuC+ZrC.

One of the principal criterion of fuel with inert matrix its reprocessing ability. From this point of view solid solutions of plutonium carbides, plutonium nitrides and inert matrix ZrC, ZrN are seems to be the best candidates.

Two synthesis processes of UC-ZrC were developed /2/:
-from initial metals,
-from the initial oxides.

The problem was to define experimentally the optimum carbon quantity to exclude the sesqui-phase. Using these techniques the following fuel was fabricated:
-56% UC +44% ZrC, 55% PuC+45% ZrC for the core region,
-15% UC +85% ZrC for the blanket region
The solid solution obtained consisted of two phases. The nonuniformity of Pu distribution was less than 5%.

Technological research on synthesis of solid solution of UN+ZrN from initial oxides and fabrication of fuel columns with different density , shape and size was performed also. As it was mentioned above the synthesis method from oxides of PuC+ZrC solid solution was developed. It's properties are close to the properties of PuN+ZrN solid solution. This fact and existing experience of UN+ZrN fabrication proved the feasibility of carbothermal synthesis of PuN +ZrN from the initial oxides. The feasibility of fabrication of UN+ZrN, PuN+ZrN solid solution from the initial metals was demonstrated also.

3.3. Irradiation.

The subassembly with 19 fuel pins was irradiated in the BOR-60 , 7 fuel pins contained 55% PuC+45% ZrC fuel , 12 fuel pins contained 56% UC+44% ZrC. Irradiation parameters are shown in Table 3.

TABLE 3. IRRADIATION PARAMETERS OF INERT
MATRIX FUEL IN BOR-60

Parameter	Value
Max.burn-up,% at.	8.
Max.fluence,cm^{-2},E>0.1Mev	$4.43*10^{22}$
Max. linear ratiNG, kW/m	40.2
Max clad temperature, C	635_+25

All fuel pins were intact.
The principal PIE results of inert matrix fuel are the following:
-no gas release from the fuel,
-fuel swelling is equal to 1% per 1% of fuel burnup,
-the fine-grain structure and round-form voids uniformly distributed through the fuel (as for the unirradiated fuel),

-instead of initial two phases only one phase was observed , which seems to be rather favourable factor for fuel performance,
-homogeneous distribution of Pu,
-local carburization of cladding only in the upper part of pins.

4. CONCLUSION.

1. To achieve the purpose of effective actinide burning, the design of the breeder reactors BN-600 and BN-800 have to be modified. The modification being considered include :
-elimination of blanket region,
-increase in the Pu content in MOX fuel,
-use of Pu fuel with an inert matrix.

2. Laboratory study on fabrication and reprocessing of MOX fuel with 45% Pu was carried out . Several fuel pins are manufactured for the BOR-60 reactor. Important experience on irradiation behavior of oxide fuel with high Pu content (100%) was gained as a result of the irradiation of two BR-10 core loadings. The results of PIE on PuO_2 fuel rods performed which were irradiated in the BR-10 reactor up to the burn-up 12 % at. and on traditional MOX fuel rods which were irradiated in BN-350 and BN-600 reactors allowed to make the following conclusion on irradiation behavior of high Pu oxide fuel:
- There is no difference between PuO_2 or UO_2 or MOX fuels for swelling, gas release, fission products behavior, microstructural changes. These properties changes depend on fuel burn-up and temperature.
-Fuel-cladding interaction increases with the Pu content increase. The lowering of the initial O/M ratio in the PuO_2 fuel and the utilization of improved cladding steel could probably decrease cladding corrosion damage to the level of UO_2 fuel pins.

3. Use of the fuel without U-238 provides the maximum burning capability. Inert matrix materials under consideration are magnesium oxide, zirconium carbide, zirconium nitride and some others. The laboratory study on oxide fuel with MgO inert matrix was carried out. The fuel was fabricated, some physical. chemical and technological properties were studied. The first experience on irradiation behavior of PuC+ZrC, UC+ZrC fuels was obtained in the BOR-60 reactor. The experiment demonstrated the real possibility to use the inert matrix fuel as a fast reactor fuel.

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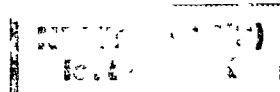
A STRATEGY FOR Pu DESTRUCTION IN PWRs

J. MAGILL, P. PEERANI, H. MATZKE, J. VAN GEEL
European Commission,
Institute for Transuranium Elements,
Karlsruhe, Germany

Abstract

We consider fuelling the core of a LWR with assemblies of standard and inert matrix based Pu fuels such that there is no net rate of Pu production. The Pu in the inert matrix fuel is obtained by recycling that from spent UO_2 fuel. The Pu produced in the standard UO_2 assemblies during reactor operation is then balanced by the Pu burnt in the inert matrix assemblies. This can be achieved by replacing approximately one fifth of the standard assemblies with inert matrix based Pu assemblies. Extended irradiation of the inert matrix assemblies is also considered.

Calculations are presented for a homogeneous core containing standard and inert matrix assemblies at various stages of irradiation. On this basis, it is suggested that the large variations in k_∞ associated with individual assemblies of inert matrix based Pu in a LWR neutron spectrum can be considerably reduced by a suitable choice of the cycle loading pattern for the assemblies.

Introduction

In conventional PWR, fissile U^{235} is present to a level of about 3.2% in a U^{238} matrix. Although not directly fissionable, this fertile U^{238} is transmuted to Pu isotopes in the reactor neutron spectrum. During the lifetime of the fuel in the reactor some of this Pu is burnt. At the end of the fuel lifetime, however, a considerable amount of Pu remains and makes a major contribution to the radioactivity of the spent fuel.

Ideally, one would like to use a non-fertile, inert matrix instead of U^{238} and thereby avoid the main source of radiotoxicity[1]. In practice, however, problems arise when one attempts to do this. One such problem is associated with the Doppler effect in U^{238}. If for some reason the neutron flux increases in the reactor it results in an increase in temperature. This in turn leads to a thermal broadening of the absorption resonances at 5eV in U^{238}, which results in neutrons being lost from the system and thereby acts as a self regulating mechanism to stabilise the reactor.

Various options are available by which one can reduce the net amount of Pu produced while at the same time having a significant amount of U^{238} present. In one such scheme the spent fuel could be reprocessed to remove the Pu isotopes and then mixed with fresh UO_2 fuel. This is the basis of Mixed Oxide (MOX) fuel fabrication.

In this paper, we consider an alternative to this approach, in which the recycled Pu is used in an inert matrix (Pu/IM). Assemblies of such Pu/IM pins would then be mixed with standard UO_2 assemblies.

In a LWR, plutonium is produced by neutron irradiation of the fertile matrix of U^{238}. In the case of the pressurised water reactor considered in the next section, plutonium is produced at a rate of approximately 9kg Pu per tonne of fuel during the lifetime (879 EFPD) of the fuel in the reactor (corresponding to approximately 300kg Pu per GW_e, see ref. 2). An inert matrix having the same volume as one tonne of standard fuel and containing initially 50kg R-Pu (i.e. 10 w/o assuming the density of the inert matrix is half that of the standard fuel) will burn approximately 40kg Pu in the same neutron spectrum and over the same time, as shown in table 1. Hence, in a steady situation where no net Pu is produced, approximately 18% (i.e. 9/49) of the core should contain inert matrix assemblies.

In a second scenario, the inert matrix assemblies could be kept in the reactor for another 879 days i.e. twice as long as the standard assemblies. Over an irradiation period of 2x879 days approximately 46kg Pu are destroyed. On average, over 879 days 23kg Pu is destroyed. Using the same arguments as above, in a steady situation where no net Pu is produced, approximately 28% (i.e. 9/32) of the core contains Pu/IM assemblies.

Table 1. Composition (%) of different types of plutonium.

	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
MOX Grade Plutonium (R-Pu)*	1.5	55.7	21.1	16.2	5.5
Weapons Grade Plutonium (W-Pu)	0	94	5.3	0.7	0

*Plutonium recovered from pressurised water reactor fuel (3.2% U^{235}) with burnup 33GWd/t

Notice in this latter scenario, keeping the inert matrix in the reactor for a further three cycles increases the amount of Pu burnt only from 40kg to 46kg. Clearly more Pu would be burnt if after three cycles irradiation, fresh Pu/IM assemblies were substituted since over the next three years irradiation another 40kg of Pu will be burnt instead of only 46-40 = 6kg! This does mean, however, that larger quantities of Pu are present in the spent inert matrices i.e. 20 rather than 8kg Pu per ton of inert matrix.

Calculational Details

The calculations were made using the point depletion code ORIGEN2[3] and checked independently with RADONN[4]. The one group cross section library used, PWRUS.lib[3] as that for a standard pressurised water reactor with 3.2% ^{235}U initial fuel enrichment, power level of 37.5 MW/tonne, and burnup of 33 GWd/tonne (achieved after 879 EFPD-effective full power days). Details of the calculation have been given elsewhere[5].
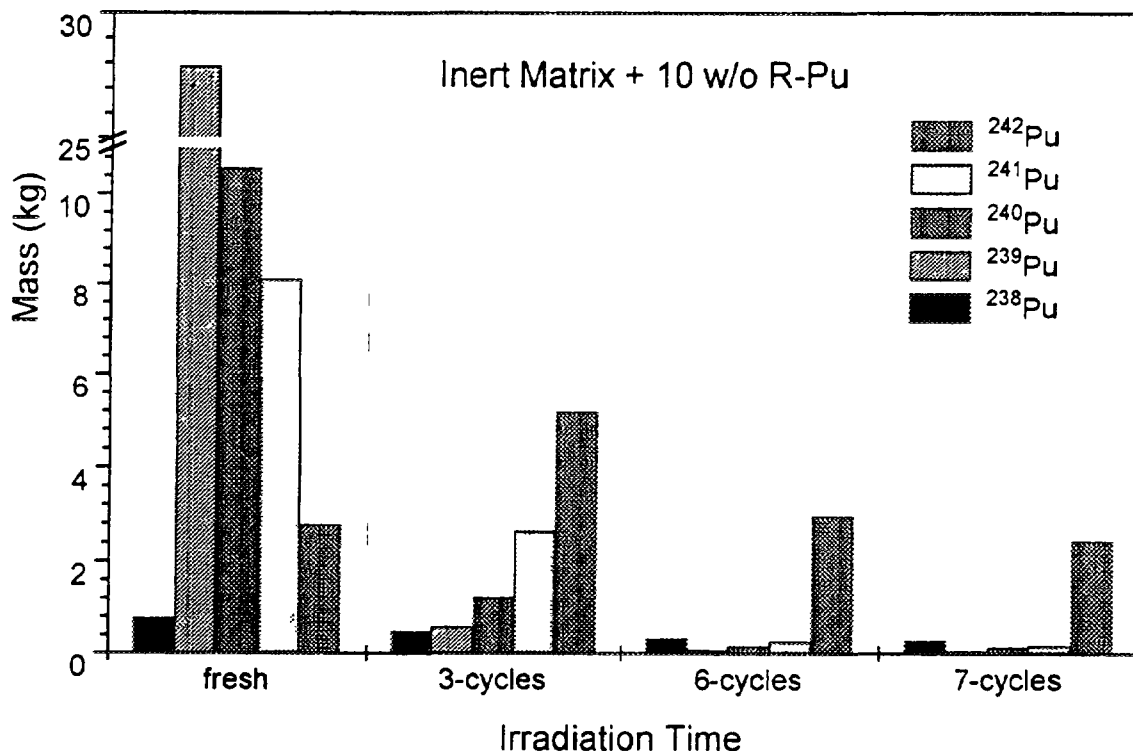

Fig. 1 Pu isotopic masses in 0.5 tonne of matrix in fresh fuel, and after 3, 6, and 7 cycles in a PWR (3 cycles = 878 EFPD) Fresh fuel contains 10 w/o R-Pu

Pu/Inert Matrix irradiation in PWR neutron Spectrum

We consider the irradiation of an inert matrix based fuel containing 10 w/o reactor grade Pu (R-Pu) in a PWR The relevant details of the PWR have been described[5] It is also assumed that the presence of the inert matrix plus fissile material does not disturb the neutron spectrum of the reactor (in the next section the variation of k_∞ for this fuel is considered) The results of an irradiation over three cycles together with extended irradiation over six and seven cycles are shown in fig 1 and summarised in table 2 for the plutonium isotopes Here the masses of isotopes present per tonne of inert matrix are given at various irradiation times In table 2, results of an irradiation using weapons grade Pu (W-Pu), with composition given in table 1, rather than R-Pu, are also given Although the amount of Pu burnt increases with irradiation time (after 7 cycles 94% of the Pu has been destroyed), the rate at which it is being burnt is decreasing Hence, over the first 3 cycles 40kg Pu is burnt Over the next three cycles only 6kg is burnt In an additional cycle only 0 7kg is burnt

Table 2 Evolution of an inert matrix containing 10 w/o Pu in the neutron spectrum of a PWR (3 cycles = 879 EFPD) Densities of the inert matrix is assumed to be half of that of the standard fuel

	fresh	3 cycles	6 cycles	7 cycles
total W-Pu	50kg	7 9kg	2 4kg	1 9kg
total R-Pu	50kg	10 0kg	3 8kg	3 1kg

Criticality Aspects of Pu/Inert Matrix Fuel

In the above calculations it was assumed that the presence of inert matrix based fuel pins did not disturb the neutron flux in the reactor A necessary condition for this to be the case is that the infinite neutron multiplication k_∞ for the standard and inert matrix based fuels are not too different In this section we consider k_∞ variations from the irradiation of standard and inert matrix based fuels
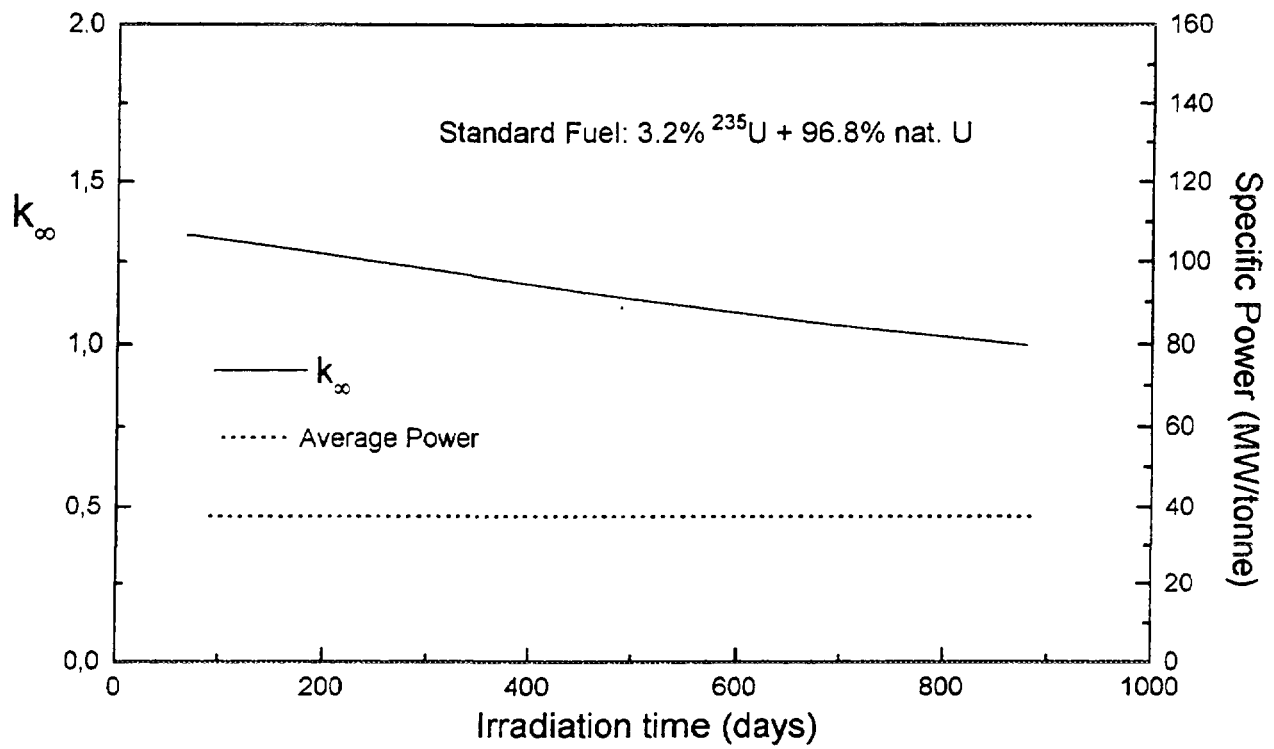
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Fig. 2. Variation of k_∞ and average power with time for a standard PWR fuel.
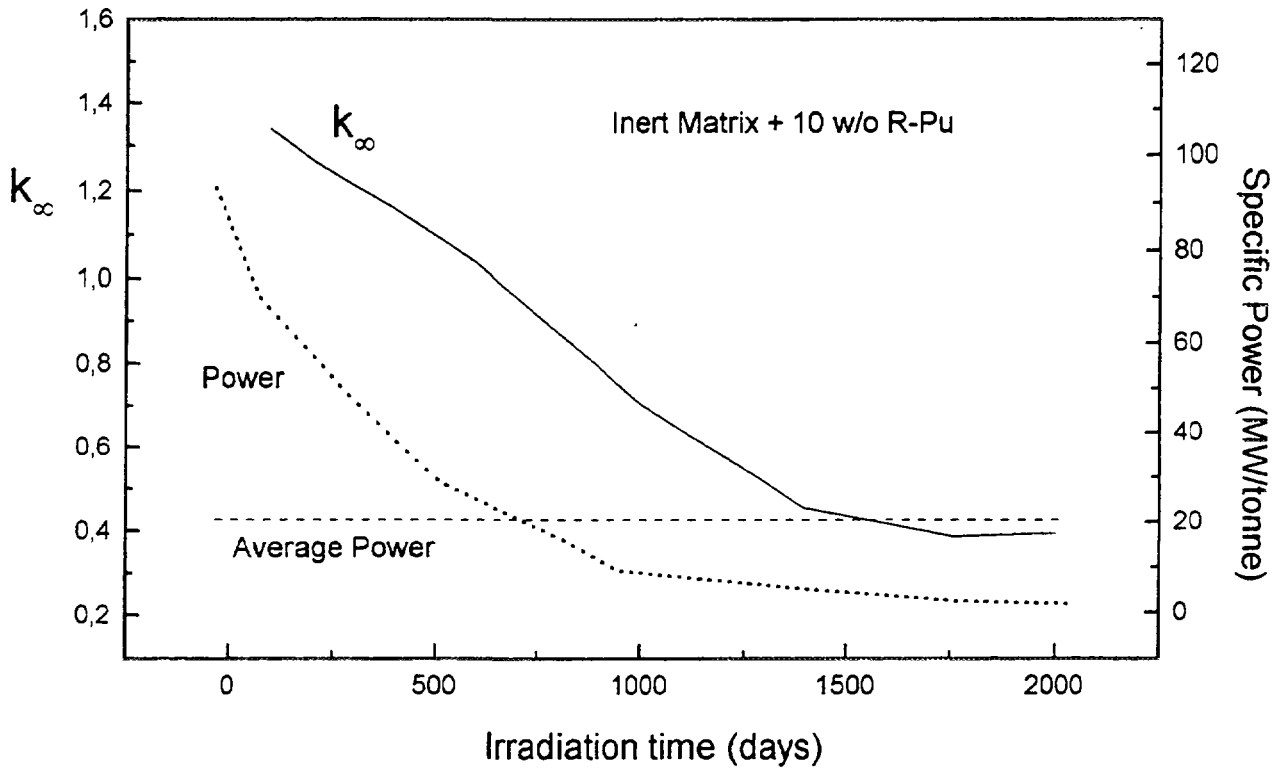

Fig. 3. Variation of k_∞ and power with time for inert matrix based R-Pu fuel during extended irradiation in a PWR neutron spectrum.

163

Regions:

	1	2	3
UO$_2$	0-1	1-2	2-3
Pu/IM	0-2	2-4	4-7

```
3
1    3*
3*   1    3*
1    3    1    3*
3    1*   3    1    3*
1*   3    1*   3    1    2*
2    2*   2    2*   2
2    2    2
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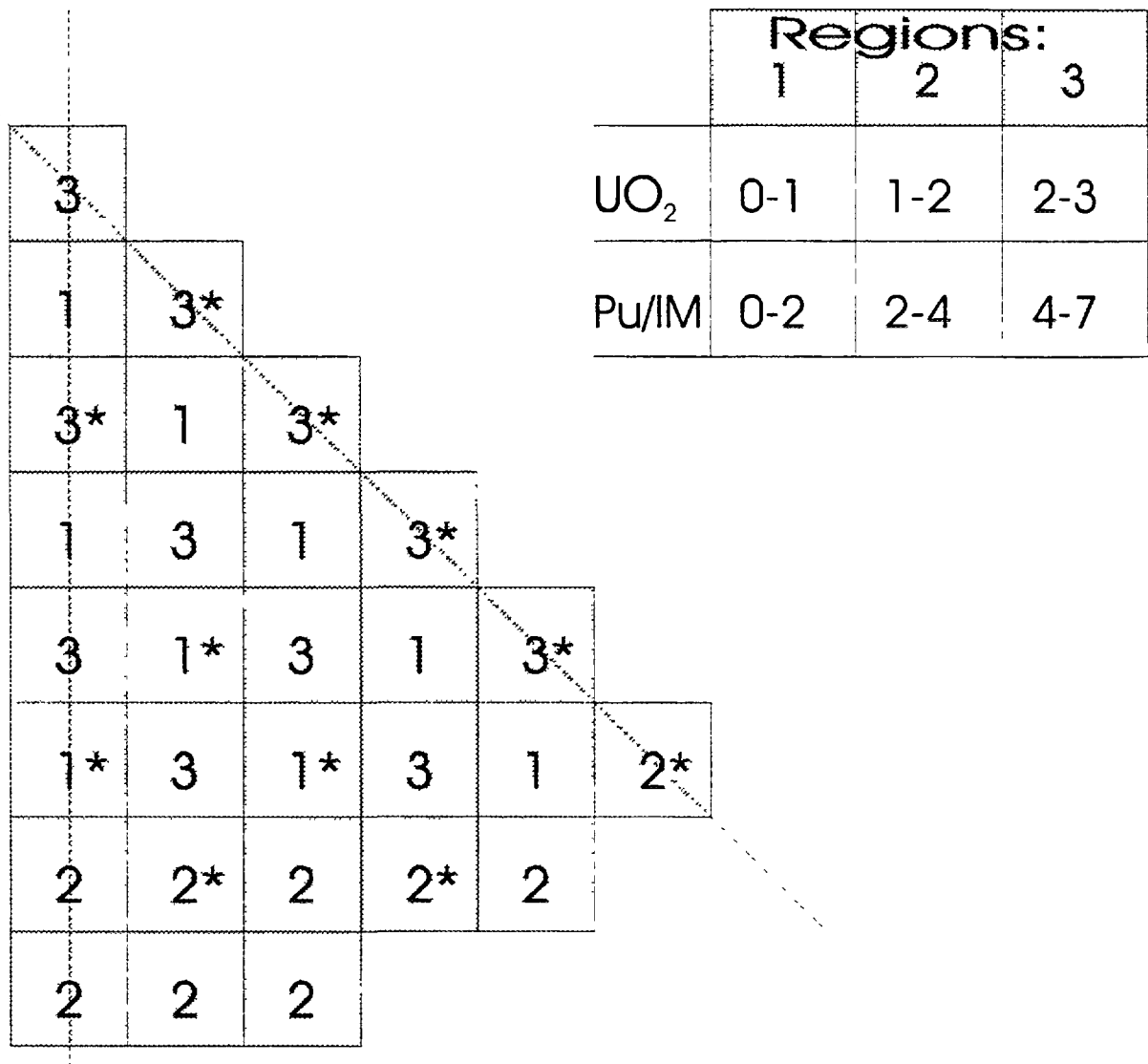
Fig. 4. Equilibrium cycle loading pattern for standard UO$_2$ and Pu/IM fuel assemblies in a PWR. Regions of the core are denoted 1, 2, and 3 and contain fresh, once burnt, and twice burnt standard assemblies. Regions denoted 1*, 2*, and 3* contain fresh, once burnt, and twice burnt Pu/IM assemblies.

In fig. 2. the variation of k$_\infty$ of the standard fuel configuration (described in the previous section) of 3.2% ^{235}U in a natural U matrix is shown over three cycle (i.e. 879 days). Also shown is the average thermal power generated. The variation of k$_\infty$ of an inert matrix containing 10 w/o W-Pu and in a neutron flux of the LWR is shown in fig 3. The irradiation time extends to 2000 days corresponding to 7 cycles of the LWR

164

During this time k_∞ from about 1 4 to 0 4 can be seen After about 900 days irradiation the inert matrix plus fissile material becomes subcritical Although $k_\infty < 1$, the Pu/IM assemblies still produce power since the multiplication factor $M \propto 1/(1-k_\infty)$

How can such an irradiation be accomplished within a LWR with a standard fuel life of 879 days? One possible scheme is to consider the core composed of three regions as shown in fig 4

In steady operation of the reactor, after each cycle (293 days), one third of the UO_2 fuel assemblies (i e those which have been in the reactor for 879 days) from positions 3 in fig 4 are removed from the reactor core and replaced by fresh fuel in positions 1 Fuel assemblies in regions 1 and 2 are moved to regions 2 and 3 respectively Similarly with the Pu/IM assemblies for a three cycle irradiation For a six cycle irradiation, the Pu/IM assemblies stay in regions 1, 2, and 3 for two cycles each For a seven cycle irradiation, the Pu/IM assemblies stay in regions 1 and 2 each for two cycle and in region 3 for three cycle
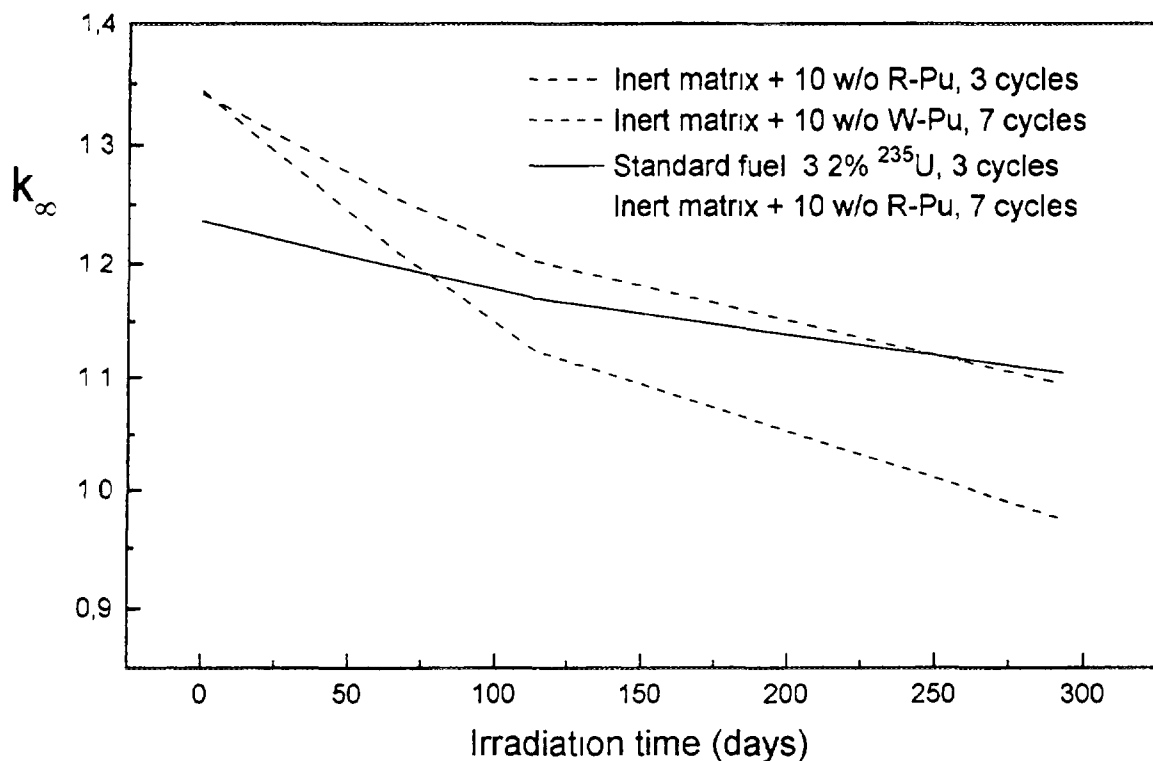

Fig 5 Comparison of k_∞ for standard and inert matrix based fuels in a PWR neutron spectrum accounting for periodic refuelling of fuel bundles

165

The above heterogeneous core layout cannot be investigated with a zero dimensional code such as ORIGEN2 However, an approximate treatment of such a heterogeneous core can be made by considering a homogeneous core containing fuel at different stages of irradiation Hence a standard core (or a Pu/IM core, three cycle irradiation) can be considered as containing fuel - one third of which is fresh, one third has been irradiated for 293 days and one third irradiated for 586 days The values of k_∞ for the UO_2 and Pu/IM fuels over one cycle (293 days) given in fig 5 show generally good agreement Initially, the value of k_∞ for the Pu/IM fuel is about 10% higher than that for the standard fuel This could be compensated for, however, by the use of a burnable poison

For the extended irradiations, the Pu/IM fuel is considered to be in the reactor for seven cycles ($\cong$ 2000 days) As described above, the core can be considered as containing a homogeneous mixture - one seventh of which is fresh, one seventh irradiated to 293 days, one seventh irradiated to 586 days, etc The results of seven cycle irradiations using R-Pu and W-Pu are shown in fig 5 It can be seen that the variations in k_∞ are still relatively large indicating that the use of a PWR neutron spectrum is not a good approximation here

Conclusions

A strategy is described whereby standard UO_2 fuel assemblies are placed together with Pu/IM fuel assemblies in a PWR such that no net Pu is generated The amount of Pu generated from the fertile U^{238} in the standard fuel is equal to the amount burnt in the inert matrix Following discharge, spent UO_2 fuel is reprocessed to obtain the Pu which is then used to fabricate fresh inert matrix fuel Spent inert matrix fuel is not reprocessed

Two options are available for burning Pu in the inert matrix either one can maximise the absolute amounts of Pu which are burnt or one can maximise the burning rates

Maximising the amounts of Pu burnt requires an extended irradiation and results in minimum levels of the Pu in the spent inert matrix fuel A zero net production of Pu requires that approximately 28% of the fuel assemblies to be inert matrix based The

166

spent inert matrix fuel contains approximately 3kg Pu in 0.5 tonne of inert matrix (which has the same volume as 1 tonne of standard fuel).

Maximising the burning rates occurs in a single 3-cycle irradiation, and results in higher levels of Pu in the spent matrix. A zero net production of Pu requires that only 18% of the fuel assemblies are inert matrix based. The spent inert matrix fuel in this case contains approximately 10kg Pu in 0.5 tonne of inert matrix.

An example of the equilibrium cycle loading pattern for a reactor core containing 180 fuel assemblies in which standard fuel is present in the reactor for 3 cycles whereas the Pu/IM fuel is present for up to 7 cycles is described. An analysis of the infinite neutron multiplication factor k_∞ for homogenous cores containing Pu/IM material at various stages of irradiation in the range of 1-7 cycles shows that extended irradiation results in large variations of reactivity. Over three cycles, however, the reactivity of both the standard and inert matrix based fuels are fairly similar. The initially higher values of for the Pu/IM fuel can be compensated by the use of a burnable poison. In addition, one may be able to accommodate such fuel within the core without significantly disturbing the Doppler effect since 82% of the core still consists of standard assemblies.

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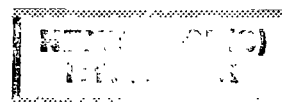
STUDIES ON HTGR Th-U FUEL CYCLE AT THE INSTITUTE OF NUCLEAR ENERGY TECHNOLOGY, TSINGHUA UNIVERSITY

ZHU YONGJIN
Institute of Nuclear Energy Technology,
Tsinghua University,
Beijing, China

Abstract

In the 1980s, thorium-uranium fuel cycle with high temperature gas cooled reactors (HTGR) was studied and the results are summarized in the present paper.

1. Physics study and design of pebble bed type HTGR with Th-U fuel (Th and high or low enriched ^{235}U)

2. Preparation of $Th(U)O_2$ microspherical fuel kernels by sol-gel process

3. Reprocessing of pebble bed HTGR Th-U spent fuel

Thorium is not a primary nuclear fuel. However, Th232 turns into fissionable U233 after the absorption of one neutron followed by two β-decays. The number of secondary neutrons in the fission of U233 by thermal and epithermal neutrons is higher than other fissionable nuclides. The conversion ratio of Th-U233 fuel cycle is higher than U-Pu fuel cycle in thermal reactors. U233 is γ radioactive due to the coexisting U232 and U233 can be diluted by U238, so it is more proliferation resistant than Pu239. The formation of Pu and transplutonium nuclides in Th-U233 fuel is far less than that in U-Pu fuel cycle. So Th fuel cycle is more environmentally acceptable. All these facts recall the attention of nuclear scientists towards Th fuel cycle.

In 1980s, the Institute of Nuclear Energy Technology(INET),Tsing-hua University,Beijing China,studied the Th-U fuel cycle with a high temperature gas cooled reactor(HTGR),mainly concerned to the reasonable utilization of natural nuclear resources.

1, HTGR is a good reactor type for the utilization of thorium[1,2]

In HTGRs, graphite is used as moderator, fuel matrix material, fuel cladding and core structural material. Helium is used as coolant, both of them have good neutron characteristics. When U233 is used as fission material and Th is used as fertile material, high conversion ratio can be obtained in HTGRs. For example, in a 80 MWe modular HTGR using Th-U fuel cycle(Th + 93% U235), conversion ratio is 0.71 instead of 0.47 for a 80 MWe modular HTGR using low enriched(~5%) U235. For a 1000MWe Th-U fueled HTGR, the conversion ratio can reach 0.82. The natural

uranium consumption of Th fueled large HTGR is only 30% of low enriched uranium fueled HTGR in the whole reactor life period. In comparison with the utilization of thorium in LWRs,HTGR using highly enriched uranium fuel can save about 50% of both natural uranium and separation power.

2, Possibility of establishing a HTGR Th-U fuel cycle in China

The Th-U fuel cycle is shown conceptually in Figure 1. The possibility of establishing a HTGR Th-U fuel cycle was studied at INET and the results are summarized briefly as follows:

2.1 Thorium resources and supply

The exploration of thorium in China has not been performed systemmetically. China has large rare earth resources and thorium mainly exists as an accompanying element in rare earth ores. Bastnasite and monazite are among these most important rare earth ores. The workable and perspective reserves of thorium in these ores are estimated as , $*10^4$ and   . $*10^5$ tons(as oxide) respectively.

At present, thorium is the by-product of rare earth industry in China. Thorium nitrate is an important by-product of rare earth factories using monazite as raw material. About 100 tons of thorium nitrate was produced in 1990. The price of thorium nitrate is lower than that of uranyl nitrate. In the extraction of rare earths from bastnasite, thorium leaves in the slag after
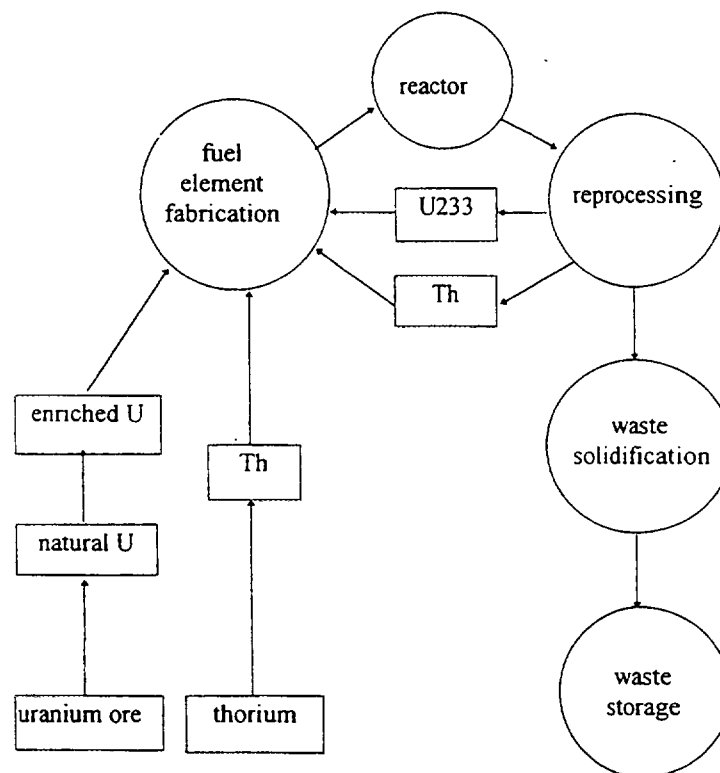

FIG. 1. Thorium fuel cycle.

the leaching of rare earths. The slag is stored under control due to its radioactivity. A process for the recovery of thorium from the slag has been developed. In China, thorium can be obtained from rare earth industry, no new exploitation and mining efforts is needed at least at the beginning stage.

2.2 Fabrication of HTGR Th fuel element[3]

The fuel element of pebble bed HTGR is TRISO coated microspherical ThO_2 and $Th(U)O_2$ kernels dispersed in spgerical matrix. The fabrication of fuel elements consists of three sub-processes, namely oxide kernel preparation,pyrocarbon and SiC coating on the oxide kernels and spherical fuel manufacturing. ThO_2 and $Th(U)O_2$ kernels are prepared by sol-gel technology followed by sintering. PyC and SiC coating on oxide kernels are carried out by vapor phase chemical deposition in a fluidized bed reactor. Spherical fuel element consisting of the coated fuel particles and graphite matrix is fabricated by quasi-isostatic press technology. Key techniques of the fabrication of pebble bed HTGR fuel elements have been mastered at INET after the research and development works of many years.

2.3 Reprocessing of spent HTGR Th-U fuel

The reprocessing of spent HTGR Th-U fuel is relatively complex owing to the structure and composition of the fuel elements. Graphite matrix and coating have to be removed to make the fuel kernels naked in the head-end process. ThO_2 and $Th(U)O_2$ have to be dissolved by HNO_3-HF due to their resistance towards HNO_3. After dissolution, Th and U are purified and separated by solvent extraction process.

In recovered U233, U232 coexists in the quantity of a few hundred ppm. γ radioactivity of uranium product increases due to the growth of daughter nuclides of U232 during standing. In recovered Th, the content of Th228 is much higher than the equilibrium content of Th228 in natural thorium. Consequently, the refabrication of recycled U and Th has to be done behind heavy shielding. The reprocessing and refrabrication cost of HTGR Th-U fuel cycle were estimated to be much higher than that of LWR U fuel on the basis of per kilogram of heavy metal treated. The cost of head-end operation is about 40-50% of the total reprocessing cost. Cost of off gas treatment is also high. Cost in fuel fabrication mainly belongs to coating process and graphite consuming. However, due to the high burn-up and high conversion ratio, the total cost of HTGR Th fuel cycle is still comparable with LWR U-Pu fuel cycle.

The reprocessing process for pebble bed HTGR Th-U spent fuel studied at INET is shown in Figure 2.

2.4 Removal of graphite matrix by burning in a moving bed burner[4]

A moving bed burner with a throughput of 100 spherical fuel elements per day was designed, constructed and tested(Figure 3) with fuel elements containing TRISO coated ThO_2 kernels. The burner combined the preheater, the first burner, and the second burner in one body. The burning temperature was kept between 800-
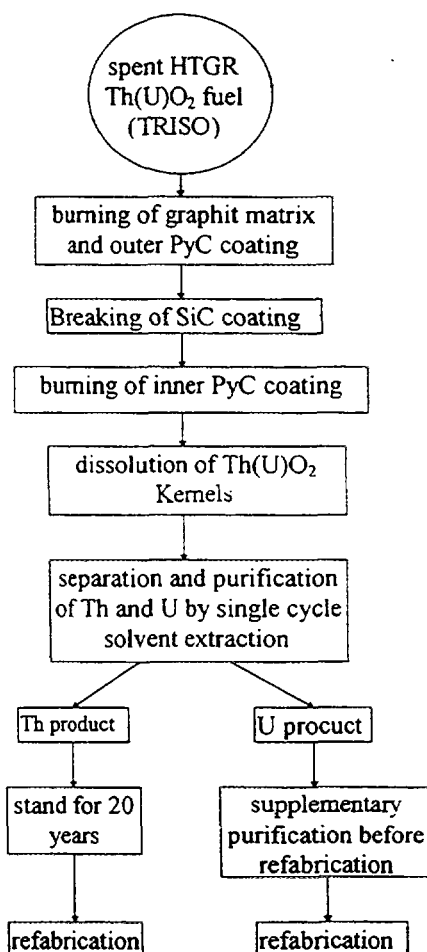
spent HTGR Th(U)O₂ fuel (TRISO)

burning of graphit matrix and outer PyC coating

Breaking of SiC coating

burning of inner PyC coating

dissolution of Th(U)O₂ Kernels

separation and purification of Th and U by single cycle solvent extraction

Th product

U procuct

stand for 20 years

supplementary purification before refabrication

refabrication

refabrication

FIG. 2. Reprocessing of HTGR Th(U)O₂ spent fuel.

850 °C. Air/O$_2$ ratio was 3/1. The specific burning rate of graphite was 800g/hr. The oxygen utilization reached 98.9%. Experimental results showed that equipment operation, product specification, and throughput met criteria required. This type of equipment is applicable to HTGR graphite sphere burning on a rather small scale.

2.5 Dissolution of ThO$_2$ kernels

Unlike UO$_2$, ThO$_2$ is not readily dissolved in concentrated HNO$_3$. Conventionally, THOREX reagent (13M HNO$_3$ + 0.05M HF +0.1M Al(NO$_3$)$_3$) is used to dissolve ThO$_2$ at boiling temperature. The time for dissolution is long and the corrosion of dissolver material is serious. A new technique was developed at INET[5] in which ThO$_2$ was dissolved in a closed pressure vessel lined with PTFT at a temperature of 180°C by a mixture of 13M HNO$_3$ + 2.5mM HF + 5mM Al(NO$_3$)$_3$. Tests with 0.5kg ThO$_2$ kernels showed that dissolution was completed within 2 hours. The pressure in the dissolver was about 1.2 MPa. Compared with conventional dissolution process, dissolving time was shortened from 70 hrs to 2 hrs and HF concentration was reduced from 0.05M to 0.0025M. The corrosion of dissolver material was minimized.
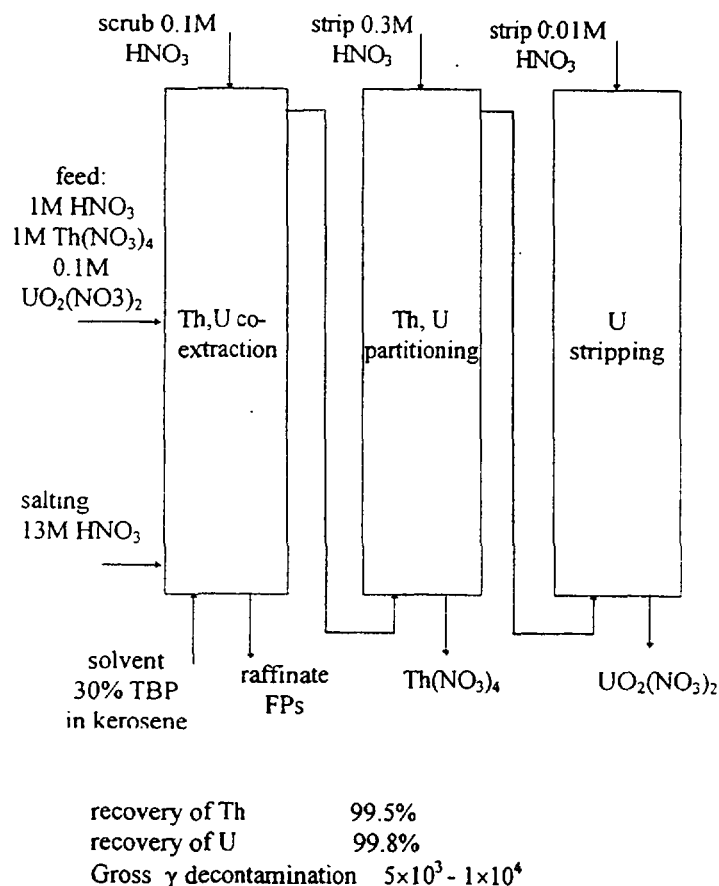
FIG. 3. Single cycle THOREX process.

2.6 Separation and purification of Th and U by a single cycle THOREX process[6]

The THOREX process using tributylphosphate(TBP) as extractant is an effective and well-known process for the purification of Th and U from fission products and the separation of Th and U. Generally, the dual cycle THOREX process was applied to get high decontamination factors and high separation factors. However, the existence of U232(T 1/2=70a) in purified U and the existence of Th228(T 1/2=1.9a) in purified Th make high decontamination from fission products meaningless. A reasonable flowsheet arrangement is shown in Figure 2.

A single cycle THOREX process was studied and was characterized by the following points:

1, using nitric acid as salting agent for Th;
2, comparatively high O/A flow ratio in Th-U co-extraction;
3, acidic feed;
4, hydrazine pretreatment of feed solution to improve decontamination;
5, 0.3M HNO$_3$ for Th stripping to avoid crud formation;

Experimental results showed that Th and U recoveries were 99.5% and 99.8% respectively. Gross γ decontamination factor was $5*10^3$- $1*10^4$. The results were verified in a semi-hot mixer-settler experiment.

2.7 Pulsed sieve-plate columns for the single cycle THOREX process[7]

The hydrodynamic and mass transfer behaviors and mathematical simulation of the steady state concentration profiles were investigated in 50mm diameter pulsed sieve-plate columns for 30% TBP-kerosene/Th$(NO_3)_4$-UO$_2(NO_3)_2$-HNO$_3$-H$_2$O system under the conditions of single cycle THOREX process. The main results are as follows:

1, optimized operation parameters are: pulse amplitute 25mm; pulse frequency 70-80 min^{-1}; void column flow rate 1.5 cm/s(two phases); throughput 54 $m^3/m^2 \cdot h$.
2, HETS(height of equivalent theoretical stage) values of H$^+$, Th^{4+}, and UO$_2^{2+}$ are all less than or near to 1 m under the following six operation modes: Th-U coextraction, Th-U coextraction scrubbing, Th-U coextraction salting, Th stripping, Th scrubbing, and U stripping.

These results proved the feasibility of using pulsed sieve-plate columns in single cycle THOREX process.

3. Conclusion

There is no existing Th fuel cycle in the world. However, the long term efforts to the research and development of Th fuel cycle given by scientists in many countries have gained large success. Th fuel cycle has its inherent characteristics: higher conversion ratio, less plutonium and transplutonium actinides formation, and higher nuclear proliferation resistibility. We believe that Th fuel cycle will attract more attention in the near future.

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NEUTRONIC ANALYSIS OF A U-Th FUEL CYCLE, LEAD COOLED ACCELERATOR-DRIVEN SYSTEM

P.A. LANDEYRO, M. GUIDOTTI
ENEA,
Rome, Italy

Abstract

This report analyses several events that involve the introduction of reactivity in an accelerator driven system, having a fast neutron spectrum, using Th solid fuel and cooled by Pb. The analysis comprises two phases: the first studies the burning of ^{232}Th in order to determine the equilibrium condition of this type of system; the second, starting from this condition (taken as reference) simulates various possible deviations, some leading to accidents. It can be shown easily that the burning capability of an accelerator-driven system can be evaluated by means of the same calculation tools and cross section sets used for a traditional reactor; the same applies also to the safety conditions.

Two irradiation cycles of 1 ton of ^{232}Th have been analyzed. At the end of the first cycle the fuel was reprocessed recovering the isotopes of Th, Pa and U and subsequently recharged after adding ^{232}Th.

The K_{inf} values calculated by ORIGEN2 remain substantially constant during the second irradiation cycle. From these data the average K_{inf} has been evaluated and with this factor the irradiation time necessary to obtain the closest K_{inf} to the average has been determined, the Th, Pa and U isotope concentrations corresponding to this time are considered those pertaining to the state of equilibrium, therefore these compositions are regarded as reference configuration.

From the calculations of the reactivity values introduced during the transients from the reference to the studied configurations result that one of the most interesting characteristics of this system is the strong void negative coefficient which is surely due to the great quantity of Pb in the reflector.

1. INTRODUCTION

This report analyses several events that involve the introduction of reactivity in an accelerator driven system, having a fast neutron spectrum, using Th solid fuel and cooled by Pb; the most important data relevant to this system are taken from ref. /1/.

An accelerator-driven system is essentially composed of a particle accelerator, of a target for neutron production and of a subcritical reactor (called "blanket"). The analysis comprises two phases: the first studies the burning of ^{232}Th in order to determine the equilibrium condition of this system; the second, starting from this condition (taken as reference) simulates various possible deviations, some leading to accidents.

2. CALCULATION METHOD

It can be shown easily that the burning capability of an accelerator-driven system can be evaluated by means of the same calculation tools and cross section sets used for a traditional

reactor; the same applies also to the safety conditions (evaluation of the neutron multiplication factor K_{eff}).

Therefore in evaluating the evolution of the concentrations during irradiation, the ORIGEN2 code /2/ has been employed, while for the evaluation of the different values of K_{eff}, the Monte Carlo code MCNP /3/ has been used. The MCNP calculations were performed using the ENDL85 cross section library (included in the MCNP code package) and another cross section library prepared at ENEA from the JEF-1 file.

Two irradiation cycles of 1 ton of ^{232}Th have been analyzed. At the end of the first cycle the fuel was reprocessed recovering the isotopes of Th, Pa and U and subsequently recharged after adding ^{232}Th. In both cycles a constant specific power of 120 MW/Ton was assumed, that causes a burnup of about 154000 MWD/Ton in reasonable agreement with ref. /1/.

The evolution of the K-infinite (K_{inf}) values calculated by ORIGEN2 is shown in fig. 1; it remain substantially constant during the second irradiation cycle. From these data the
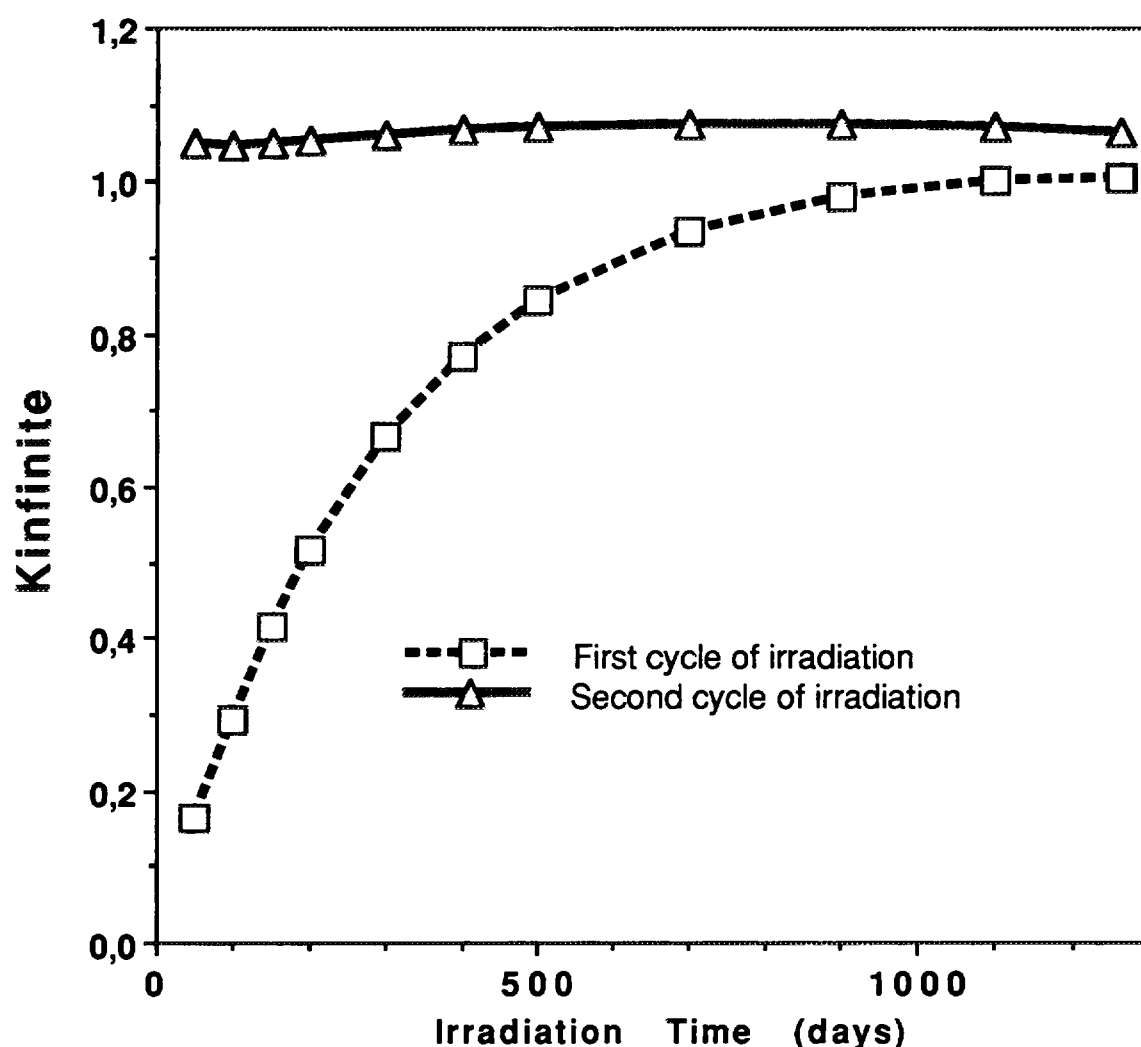

Fig. 1. Kinfinite as a function of irradiation time, ORIGEN2 data

180

average K_{inf} has been evaluated and with this factor the irradiation time necessary to obtain the K_{inf} closest to the average has been determined; the Th, Pa and U isotope concentrations corresponding to this time are considered those pertaining to the state of equilibrium, and these compositions therefore are regarded as the reference configuration.

3. CRITICALITY ANALYSIS

3.1 Validation test of the calculation method

Two critical masses experimentally determined /4/ have been considered; they correspond to two U spheres having a density of 18.62 g/cm^3 and the following composition in percent: 98.13% ^{233}U, 1.24% ^{234}U, 0.03% ^{235}U and 0.60% ^{238}U; the first sphere which is not reflected, has a critical mass of 16.2 kg, while the second, reflected by means of a shell of natural U 19.9 cm thick with a density of 10 g/cm^3, has a critical mass of 5.74 kg. Table I shows the results of the calculations for both spheres. The values are reasonable.
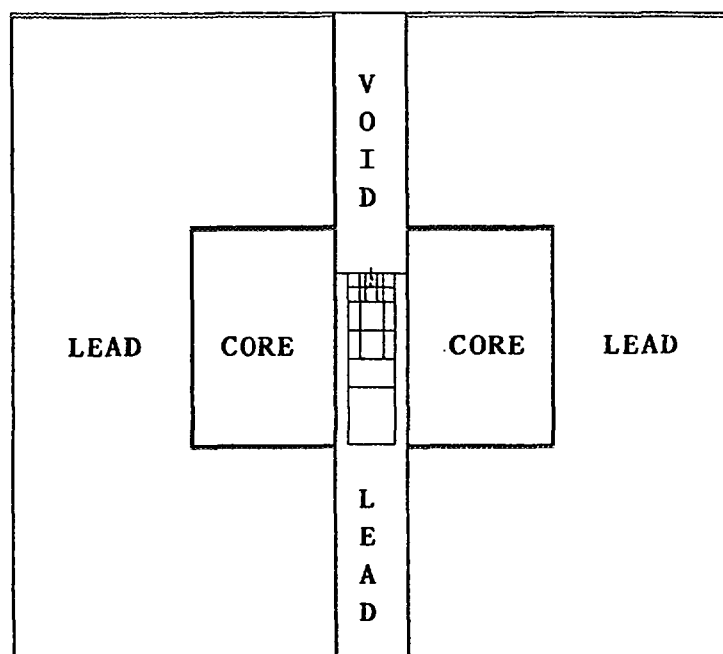

Fig. 2. Geometrical configuration used in MCNP calculations.

Table I: Neutron Multiplication factors relevant
to experimental critical masses. ^{233}U Core.

Reflector	Critical mass (kg)	k_{eff}
No	16.2	0.99314 + 0.0006
U (natural)	5.74	0.99275 + 0.0022

3.2 Geometry

To describe the geometry of the analyzed cases a MCNP model has been developed, see fig. 2. The core is made of carbon steel cladded ThO_2 fuel pins, 1 cm in diameter. The core has a height of 150 cm and a diameter of 300 cm and it contains 33 Tons of fuel pins. A cylindrical space of 50 cm is filled with Pb coolant in order to constitute the target for the high energy beam. The core is enclosed, except in the accelerator-target region, by a 150 cm thick lead reflector.

The K_{eff} corresponding to this reference condition is 1.00383 + 0.0037, the difference existing to the reference condition of ref. /1/ (K_{eff}=0.98) can be explained because in the present case are not considered the fission products.

3.3 Analyzed configurations

The studied configurations are the following:

- voiding of the neutron source, that could happen in the case of rupture of the accelerator-target interface, causing Pb at high temperature to come in contact with the accelerator (void);
- uniform decrease of the temperature to the Pb fusion point (the system will surely have to undergo maintenance, therefore will be shut down and cooled);
- spent fuel decay, that causes K_{eff} to reach its maximum after 160 days of decay when ^{233}Pa is about 1/100 of its initial value;
- emptying of the core Pb;
- emptying of the Pb from the neutron source, the core and the reflector;
- complete emptying of the Pb from the system and simultaneous spent fuel decay up to 160 days;
- "hour-glass" deformation of the core; while the top and bottom of the fuel elements remain fixed, their center is squashed against the neutron source (central zone occupied by Pb).

As the system will have an intermediate Pb - Bi circuit between the Pb and H_2O circuits /5/, accidental scenarios considering the introduction of H_2O in the core have not been studied.

4. ANALYSIS OF THE RESULTS

Tables II and III show the results of the calculations. From Table II it can be seen that the first and the second configurations have a Dk/k contained within the subcriticality margin established for this system, 0.02, see ref. /1/.

From the calculations of the reactivity values obtained passing from the reference to the studied configurations it results that one of the most interesting characteristics of this system is the large negative void coefficient; this is surely due to the great quantity of Pb in the reflector, because emptying Pb from the neutron source and/or the core causes an introduction of positive reactivity.

This fact may be taken advantage of also to overcome the problem of the positive reactivity consequent to ^{233}Pa decay. The reactor immediately after the shut down could be

Table II: Reactivity Introduction in the CERN project

Reference value for the neutron multiplication

factor k_{eff} : 1.00383 ± 0.0037

CAUSE	k_{eff}	$\Delta k/k$
Voiding of the neutron source	1.01603 ± 0.0023	0.01208
Uniform lowering of temperature to 327 °C	1.02632 ± 0.0032	0.02215
Decay of the fuel up to 160 days	1.12179 ± 0.0023	0.11110
Drainage of Pb in the core	1.04532 ± 0.0027	0.04050
Total drainage of Pb (in neutron source, core and reflector)	0.87990 ± 0.0019	-0.13177
Simultaneous total drainage of Pb and decay up to 160 days	0.97054 ± 0.0017	-0.03373

Table III: Reactivity introduction in the CERN project of the "hour - glass" deformation incident

Height of the compressed central zone (cm)	k_{eff}	$\Delta k/k$
30	1.02556 ± 0.0031	0.02142
50	1.03007 ± 0.0029	0.02580
80	1.04060 ± 0.0031	0.03597
110	1.04290 ± 0.0023	0.03828

drained of Pb, if the heat exchange situation allows it and the positive reactivity due to ^{233}Pa decay would not give rise to accidental conditions.

Table III presents the results of Dk/k calculations for the core "hour-glass" deformation accidental scenario corresponding to different heights of the compressed zone; the reactivity rises with the height and asintotically approaches that which is introduced by draining Pb from the core: 0.04050. This Dk/k value may be one of the reasons wherefor the operating K_{eff} has been reduced in ref. /5/ from 0.98 to 0.96.

5. CONCLUSIONS

The most interesting characteristics of this system is the strong void negative coefficient which is surely due to the great quantity of Pb in the reflector and to the low threshold for the n,2n reaction of this material.

The new subcriticality margin, 0.04 fixed in ref./5/, would be sufficient to overcome all the reactivity analyzed deviations of the reference condition.

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LIST OF PARTICIPANTS

Arkhhipov, V.   Nuclear Power Technology Development
                                Section
                                Division of Nuclear Power
                                IAEA
                                P.O. Box 100
                                A-1400 Vienna

Bernander, Ö   ABB Atom
                                Fuel Division, Dept. B
                                S-721 63 VÄSTERÅS
                                Sweden

Borisov, V.   Minatom
                                Moscow
                                Russian Federation

Brennan, J.   BNFL
                                MOX Technical Department
                                B582
                                BNFL
                                Sellafield, Cumbria
                                United Kingdom

Cordfunke, E.H.P.   ECN
                                P.O. Box 1
                                1755 ZG Petten
                                The Netherlands

Franken, W.M.P.   ECN
                                P.O. Box 1
                                1755 ZG Petten
                                Netherlands

Furukawa, K.   Institute of Research and Development
                                Tokai University
                                Hiratsuka, Kanagawa 259-12
                                Japan

Gibson, I.   3 Hardy Close
                                Martinstown
                                Dorchester
                                Dorset
                                DT2 9JS
                                United Kingdom

Hron, M.J.

Nuclear Research Institute
Řež plc
CZ-250 68 Řež
Czech Republic

Juhn, P.-E.

IAEA
Division of Nuclear Power and the Fuel Cycle
P.O. Box 100
A-1400 Vienna

Kadi, Y.

CERN
European Orginization for Nuclear
Research
1211 Geneva 23
Switzerland

Kim, Y.

KAERI
150 Duckjin-dong, Yuseong-ku
Taejon, 303-353
Republic of Korea

Lecocq, A.

Pdt of EURIWA
27 av des Fauvettes
91400 Orsay
France

Lee, B. W.

KAERI
Nuclear Policy Department
P.O. Box 105
Yuseong, Taejon
305-600
Republic of Korea

Liljenzin, J.-O.

Department of Nuclear Chemistry
Chalmers University of Technology
S-412 96 Göteborg
Sweden

Magill, J.

European Commission
Institute for Transuranium Elements
Postfach 2340
76125 Karlsruhe
Germany

Park, W. S.

Korea Atomic Energy Research Institute
P.O. Box 105
Yusong
Taejeon
Republic of Korea

Phlippen, P.-W.

Research Center Jülich (KFA)
Institute for Safety Research
and Reactor Technology (ISR)
P.O. Box 1913
D-52425 Jülich
Germany

Purushotham, D.S.C.

Bhabha Atomic Research Centre
Trombay
Bombay-400 085
India

Rief, H.

ISPRA
V. Matteotti, 18
I-21020 BODIO (VA)

Rütten, H.J.

Research Center Jülich (KFA)
Institute for Safety Research
and Reactor Technology (ISR)
P.O. Box 1913
D-52425 Jülich
Germany

Schapira, J.-P.

CNRS / 1N2P3
Institut de Physique Nucléaire
BP n° 1
91406 - ORSAY - Cédex
France

Taczanowski, S.

Faculty of Physics & Nuclear
Techniques
Academy of Mining & Metallurgy
Al. Mickiewicza 39
30-059 Cracow
Poland

Turenne, de

COGEMA
Nuclear Fuel Branch
Technical Section
2 rue Paul Dautier - B.P. 4
78141 Vélizy Cedex
France

Vertsymaha, O. Y.

Institute for Nuclear Research
Kiev
Ukraine

Vettraino, F.

ENEA
Nuclear Energy Division
Via Martiri di Monte Sole, 4
40129 Bologna
Italy

Werner, H.

Research Center Jülich (KFA)
Institute for Safety Research
  and Reactor Technology (ISR)
P.O. Box 1913
D-52425 Jülich
Germany

Zabudko, L. M.

Institute of Physics and Power
Engineering
249020, Bondarenko Square 1
Obninsk
Russian Federation

Zhang, Y.

China Institute of Atomic Energy
P.O. Box 275(95)
Beijing, 102413
China

96-04233