

SESSION 1: INTRODUCTORY SESSION

CHAIRMAN: B. BARRÉ (FRANCE)

OPENING REMARKS BY THE DIRECTOR-GENERAL OF THE NEA

Ladies and Gentlemen,

It is with great pleasure that I open the 3rd International Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation which is organised by the OECD Nuclear Energy Agency and hosted by the Commissariat à l' Energie Atomique.

Since the NEA was invited to take up this topic in 1988, the interest in it has grown in several of our Member countries. From the outset it was stressed that successful application of Partitioning and Transmutation would not replace the need for geological disposal for high level waste. The task is one of long-term scientific research, but it is increasingly recognised that certain short- or medium-term benefits could also be derived. While a number of bilateral agreements on P&T between OECD countries do exist, clearly, substantial benefit could be achieved from wider international activities and co-operation.

As you know, the NEA's P&T activities are conducted under the guidance of its Nuclear Development and Nuclear Science Committees. Resulting information is always shared with the Radioactive Waste Management Committee.

Under the "umbrella" of the NDC, two successful International Information Exchange meetings were held at Mito City and Argonne National Laboratory in 1990 and 1992, respectively.

During the Mito City meeting an interesting set of papers was presented both on policy orientations and on scientific aspects. The discussion, which concentrated on wide ranging ideas regarding future technologies, led to the conclusion that several disparate approaches had already been taken. It was agreed to organize small specialist meetings on suitable topics such as fundamental physics and chemical data requirements.

Therefore, the NEA helped organise two specialist P&T meetings. The first was arranged by JAERI on partitioning technology and was held in Mito City in November 1991. A wide range of processes for the separation of actinides and fission products, both wet and dry, were presented. The second was at the Paul Scherrer Institute, Switzerland, in March 1992, on the topic of accelerator-based transmutation. Again, there was a wide variety of concepts discussed, together with presentations on data acquired, data needs and models that could be applied.

The specialist meetings held so far have been found most useful by the participants. There had been considerable discussion in which the results achieved in earlier and current work in the USA and Europe had been used to illuminate the more recent work in the vigorous programme being pursued by Japan. There had been a wealth of suggestions for different flow sheets with which to improve the manipulation of actinides into different product streams and for alternative reactor and accelerator-driven transmutation of actinides and fission products.

In November 1992, the Argonne National Laboratory hosted the second NEA International Information Exchange meeting. The papers presented indicated that one common thread was the need for guidance on research needs. A number of emerging important issues were identified during the meeting, including the legal background, the incentives and the implications for the whole fuel cycle in different countries.

It was concluded that an attempt to compare systems studies, or proto-systems studies, already in progress should form a central part of the third NEA Exchange meeting. This was regarded as the very first phase in the approach to a more co-ordinated systems study that would seek to identify benefits and penalties of adding P&T to the nuclear fuel cycle. A careful preparation, given the many diverse approaches and criteria in use, was believed to be essential in setting the framework for the comparison of the studies.

The Nuclear Science Committee and the NEA Data Bank have also carried out several programmes related to Partitioning and Transmutation. You will hear more about this work from the NSC's Vice-Chairman Dr. Matsuura.

The NEA's first stage of the P&T Information Exchange Programme was intended to run for 5 years and will thus be concluded with this meeting. As you are no doubt aware, the Nuclear Development Committee has decided during its June 1994 meeting to extend this stage for a further 3-year period and approved a specific project proposal to perform comparisons of systems studies.

You will be primarily addressing, during this meeting, ways of evaluating a limited number of different P&T systems and their integration into the nuclear fuel cycle. Other areas of interest would certainly be the effects of progress made with various national P&T activities, technical advances, economic assessments and the overall P&T objectives in relation to environmental considerations.

Partitioning and Transmutation is in the stage of development. We must collect all the necessary information which will allow us to fully understand its technical feasibility, the related costs and its environmental impacts. For this type of activity, the role of international co-operation is indispensable for guaranteeing the quality of the results and ensuring proper co-ordination and optimal resource utilization. The NEA and its Committees have the experience required and will be ready, if our Member countries wish us to do so, to undertake all the necessary work.

Before concluding my remarks I would like to underline that both past and present NEA P&T activities were assisted by financial contributions provided by the Government of Japan which was generous enough to provide further assistance to support the NEA's approved 1995 programme.

I thank you all for coming, and I particularly wish to thank the team of speakers, as well as, our hosts who have, I am sure, laid the basis for a successful meeting. I wish you all an interesting, instructive and profitable three days.

NEA/NSC ACTIVITIES ON NUCLEAR TRANSMUTATION

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Abstract

The Nuclear Science Committee (NSC) was set up at the end of 1991, to replace the former three science committees; the committee on reactor physics, the nuclear data committee and the Data Bank Committee. While continuing many projects started under the former committees, the NSC has started new activities according to new needs. Among them, the NSC has taken on different activities related to the transmutation of actinides under collaborations with NDC and the RWMC. Various concepts on transmutations are proposed, but at present one whose feasibility has yet to be demonstrated. In this context, the NSC decided, as a first step of approach to this area, to form a task force reviewing the physics aspects of various proposals on transmutation concepts. The task force requested about 30 specialists working for transmutation to inform the physics aspects on their concepts. Based on the comparison of the results of the calculation of transmutation rate of each concept, there seems to be significant discrepancies between those concepts. As the next step, a benchmark analysis is proposed for a set of common systems of calculation in order to understand and to assess their physics concepts.

1. Background of NSC activities

The NSC was set up at the end of 1991. It replaced the former three science committees. They are, the committee on reactor physics, the nuclear data committee, and the data bank committee.

The NSC was asked to take a very wide view in its choice of topics to study. The activities of former three committees had boundaries of physics and nuclear data. But the NSC was asked especially to identify major scientific issues in areas outside such traditional boundaries. At the same time, it was particularly emphasized to establish good working relations with other NEA committees, such as the NDC, the RWMC, the CSNI etc.

From the above mentioned point of view, one of the essential roles of the NSC is to correspond systematically and comprehensively to various needs of scientific information identified by other committees. Therefore, the international information exchange meeting, like this, will give very good chance for the NSC to identify the scientific issues at present and in future.

2. A first step of approach and related activities

While continuing many projects started under the former committees, the NSC has started new activities according to new needs. Among them, the NSC has taken on different activities related to the nuclear transmutation under collaborations with the NDC and the RWMC. The nuclear transmutation is in itself an interesting concept, and will form an essential component of the NSC's activity. Various concepts on transmutations are proposed, but at present any one feasibility has not been demonstrated. A very large number of studies still need to be carried out.

Observing the historical background of the NSC, the achievements of the former three committees, and the international position of the NSC, effective use of the following two scientific tools is most substantial for activities of the NSC. The one is the creation of data bases, and the other is the organization of inter laboratory comparison exercises in member countries. The role of the Data Bank is essential in this type of activities. The creation of nuclear data base on nuclear transmutation is one of the most important activities of the NSC at present and in future as well.

As for the transmutation concepts using fission reactors, most of nuclear data could be obtained from the data base which have been created in the history of reactor development. The other typical concepts of transmutation are based on accelerator driven processes. Many new nuclear data are requested for better evaluation of accelerator driven concepts. The typical examples of them are the nuclear data of high energy proton above several hundred MeV and those of intermediate energy neutrons above several MeV. The NSC has been intensively promoting the creation of nuclear data base for these new requirements.

As a preliminary step of inter laboratories exercises on transmutation concepts, the NSC decided to form a task force reviewing the physics aspects of various proposals on transmutation concepts. The task force requested 30 specialists working for transmutation studies to inform their physics aspects. An overview of the information has been compiled as NEA/NSC/DOC (94)11 by the task Force.

Based on the comparison of the results of the calculation of transmutation rate of each concept, there seem to exist significant discrepancies between the concepts.

As for reactor based systems, for example, there is a significant difference of Cm-244 burnup characteristics among fast reactor based systems. The reason of difference has not been identified.

As for accelerator based systems, only one calculated results has been reported on mass balance of minor actinides between the beginning of equilibrium cycle and the end of it. This might be partly due to calculational difficulties of taking account of neutrons having higher energy more than 20 MeV. Therefore, it is suggested that a next step of study is necessary.

3. A proposal of next step

As an example of the next step, a benchmark analysis is proposed by using a common set of system specifications and nuclear data. The results of benchmark will be substantially helpful to understand and to assess the performances of the system, more precisely. In addition to that, the task force members have suggested that the following issues should be studied in terms of transmutation concept, they are

- radiotoxicity after transmutation,
- safety features of transmutation systems, and
- nuclear data of transuranium nuclides.

In order to challenge these issues, the NSC needs further and stronger cooperation with other NEA committees. This meeting will give a lot of stimulations to activities of the NSC.

IAEA ACTIVITY ON PARTITIONING AND TRANSMUTATION OF ACTINIDES AND FISSION PRODUCTS

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ABSTRACT

In 1990, the IAEA received a request from Member States to review the status of research and development on partitioning and transmutation of actinides and fission products. In response to this request the Advisory Group Meeting (AG) was held in the fall of 1991. AG advised the Agency to play an active role in coordinating international activities in this area. A series of meetings that followed identified considerable interest among many Member States and international organizations in the P&T options as a potential complement to the reference concepts of the back-end of nuclear fuel cycle. Inherent difficulties for the Agency to actively explore this programme were identified including non-proliferation concerns from some Member States about partitioning technology and possible duplication of effort in other international organizations, especially OECD/NEA. But, there remain fundamental questions to be addressed on the objectives of and motivations for P&T and it is clear that some common international understanding would be necessary. In order to contribute to the solution of this problem, and considering the existence of programmes being implemented by OECD/NEA, the Agency has initiated a new CRP entitled "Safety, environmental and non-proliferation aspects of partitioning and transmutation of actinides and fission products" (1994-1998). This presentation will explain about this Agency's new CRP and how the Agency's work is co-ordinated with other international activities.

1. Introduction

There is considerable interest among many Member States in the Partitioning and Transmutation (P&T) of long-lived radionuclides as a potential complement to the reference concept of the closed nuclear fuel cycle comprising: fuel fabrication, energy generation, intermediate storage of spent fuel, reprocessing, plutonium use in fuel and disposal of solidified high-level waste (HLW) in a deep geologic repository.

P&T is a complex issue involving safety, technological, economic and public acceptance aspects. Until now, there is no clear indication that P&T of actinides and long-lived fission products offers a more safe and effective waste management option. Current national (China, France, Japan, Russia) and international (OECD/NEA and CEU) programmes on P&T tend to be more concerned with the development of techniques for the separation of actinides, preparation of fuel and targets and for transmutation of actinides and long lived-fission products rather than with the studies of safety implications of P&T.

The IAEA's involvement with the problem of partitioning and transmutation of actinides and fission products goes back to 1976 when the Agency carried out a Co-ordinated Research Programme (CRP). The results of the CRP were published in a Technical Report Series No. 214 in 1982. The conclusion

of this report on P&T was rather negative. In 1990, the Agency received a request from some Member States to re-activate the programme. The Advisory Group Meeting (AGM) held in the autumn of 1991 advised the Agency to play an active role in coordinating international activities in this area. A series of meetings (two Consultant Meetings (CS) and one Technical Committee Meeting (TCM)) that followed identified considerable interest among many Member States and international organizations in the P&T options as a potential complement to the reference concepts of the back-end of nuclear fuel cycle.

The meetings also gave the opportunity to review the current status and progress of national and international programmes on P&T, to identify the most important directions of national and international co-operation. The meetings provided advice regarding the IAEA programme on P&T.

2. Main results of IAEA meetings on P&T

There is a scientific consensus that the current waste management concept provides an adequate protection to the population and the environment by sufficient confinement of radionuclides. According to the experts' opinion the purpose of P&T is to reduce the long-term effects of radiotoxicity of actinides and long-lived fission products but cannot eliminate the need for a geologic repository. In some countries (France, Japan, Russia) it has already received political or institutional backing as a complementary future strategy to the current fuel cycle. The specific role of P&T in the nuclear fuel cycle needs to be defined further within the general trend to minimize waste and the growing concern about a maximum degree of safety in the very long term.

A general guidance with respect to hazard reduction by P&T cannot be given since the local geological, hydrogeological and confinement conditions of proposed repositories vary from country to country. However, qualitative indications will be very useful to establish an order of priority in the radionuclides to be studied regarding the hazard reduction involved.

It was emphasized that from the point of view of potential hazards (potential hazard involves the source term without taking into account the geological barriers), the actinides are the most important nuclides to be investigated in a P&T concept, however, some long-lived mobile fission products constitute the main residual hazard (residual hazard means the radionuclides released to the biosphere) over a long term period of time.

The following are main conclusions of meetings concerning the progress being made in the fields of partitioning, of fuel and target developments and of transmutation.

Partitioning

Two main fields of work are involved in the partitioning of plutonium, minor actinides and long lived radionuclides: wet separation methods that are implemented in association with the PUREX process; pyrometallurgical processes, which may follow the PUREX process or completely replace it.

Significant progress has been achieved in reducing the losses of plutonium from the PUREX process and in the improvement of neptunium and technetium separation through flow sheet alterations. Progress has also been made with the partitioning of other elements.

Fuel and Target Development for Transmutation

The technical feasibility of the use of U-Pu and U oxide fuels for the recycling of minor actinides in a fast reactor has been proven to some extent. Oxide, metal alloy and inert matrix fuels and targets containing MA and Tc-99 are fabricated, at a laboratory scale, and scheduled for irradiation. Concepts of nitride and molten salt fuels have been proposed for the advanced transmutation systems, e.g. actinide burners and accelerator driven systems.

Transmutation

An overall reduction of the radiological hazards requires the development of new technologies, e.g. with an accelerator driven transmutation system. Only with a major break-through in technology are we likely to reach the expected goal.

3. Role and involvement of IAEA

The participants of IAEA meetings stressed that OECD/NEA and CEU have managed international P&T programmes. However, a number of countries are not yet covered by these international bodies, therefore the IAEA might play a significant role by coordinating the efforts of these countries with OECD/NEA and CEU countries.

Based on the recommendation of experts from Member States and considering the existence of technologically oriented programmes being implemented by OECD/NEA and CEU, the IAEA established a complementary programme on the safety, environmental and non-proliferation aspects of P&T that could be beneficial in assisting Member States.

A series of topics are to be addressed as:

1. the definition of hazard criteria;
2. the evaluation of hazard reduction;
3. the definition of a priority list of nuclides to be considered.

Since each of the three above-mentioned topics are strongly dependent on the geological nature of the repositories and on their technological implementation it is deemed necessary to widen the scope of the P&T discussion to the general waste management issue involved with such an option. It is, therefore, suggested to associate plutonium and waste management experts in the P&T discussions.

In the mainstream of this programme, IAEA with the P&T, Pu, and Waste Management expert groups will be able to make a comprehensive evaluation of the following tasks:

4. establish a worldwide inventory of the radionuclide source term;
5. identify the possible fundamental safety benefits of a series of P&T options and scenarios;
6. evaluate the waste conditioning processes in the perspective of a future transmutation option;
7. identify the influence of secondary waste arisings on the net safety benefit of the overall P&T processes;

8. examine the non-proliferation implications of an extended fuel cycle activity over a long period of time.

This recommendation became a basis of a CRP on P&T initiated by the IAEA in 1994.

4. **Co-ordinated research programme Evaluation of the Safety, Environmental and Non-Proliferation Aspects of Partitioning and Transmutation of Actinides and Fission Products", 1994 - 1998**

Scientific scope and programme goals

The CRP will promote the exchange of information on the results gained by different countries in order to clarify the objectives and motivation of P&T and to obtain a common international understanding from the standpoint of safety and non-proliferation. The results of this programme will also give the opportunity to Member States to define the scope of further research and development work required in this field. The intended beneficiaries of the proposed programme are both the policy makers and the research programme managers in the fuel cycle field.

In the framework of the CRP the radionuclides hazard is to be studied in order to identify the critical nuclides to be considered in a P&T strategy and to quantify their radiological importance in a global nuclear fuel cycle analysis. A priority list of radionuclides for P&T is to be established according to the hazard definition.

The necessary extent of P&T for achieving the radiological hazard reduction should be clearly defined. It may turn out that not all long-lived radionuclides will have to be partitioned to the same degree and hence their contribution to the disposed waste will have a different effect on the overall hazard reduction. The achievable goals in hazard reduction by different P&T systems will be examined.

The construction and operation of various nuclear facilities needed for P&T may involve increasing risk for the personnel and additional detriment to the environment. The safety aspects of the modified fuel cycle are to be realistically assessed.

Non-proliferation implications of different P&T systems will be assessed in order to define the most promising and effective schemes in this regard.

Programme Subjects

A list of proposed subjects is given below:

- a) **Critical Radionuclides for P&T**

These studies will define the hazard criteria and consider the hazards of different radionuclides in HLW. As a result of the studies a priority list of critical radionuclides will be established for partitioning and transmutation.

b) **Radiological Hazard Reduction by P&T**

The aims of the radiological hazard reduction will be defined and the necessary extent of P&T for achieving the hazard reduction will be studied. Attainable hazard reduction by different P&T systems will be assessed. For different partitioning processes under development, the influence of secondary waste arisings and the quantification of unseparated radionuclides which must be sent to a repository will be evaluated.

c) **Safety assessments of different P&T processes**

These studies will include investigations of the operational safety aspects of different P&T systems including the management of the secondary waste in order to assess the safety of the modified fuel cycle.

d) **Non-proliferation aspects of P&T processes**

These studies will be aimed at the potential of P&T for the technical contribution to the non-proliferation regime.

**RESEARCH ACTIVITIES ON PARTITIONING AND TRANSMUTATION
SUPPORTED BY THE EUROPEAN COMMISSION**

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ABSTRACT

The research activities supported by the European Commission in the field of Partitioning and Transmutation (P&T) are summarized in this paper together with the main results obtained so far. These activities are related to experimental work on partitioning of long-lived radionuclides from high level liquid waste, fuel and target development and an overall strategy study to assess the potentialities of P&T for the safety of the management and storage of radioactive waste. Particular emphasis is put on the strategy study, which already gives some elements of answer concerning the potential radiotoxicity, the cost, the quantity of transmuted radionuclides, the time necessary for transmutation and the required separation efficiencies between minor actinides and lanthanides. Finally, the main conclusions of this study raise some of the questions, which remain to be solved before implementing P&T.

INTRODUCTION

The fourth five-year shared-cost research and development programme on "Management and Storage of Radioactive Waste 1990-1994" has been adopted by the Council of Ministers of the European Communities, at its meeting on 15 December 1989. The study of the potentialities of transmutation of long-lived radionuclides has been included in this programme following a request of the European Parliament. Besides, research work on partitioning and transmutation (P&T) is carried out at the Joint Research Centre of the European Commission in Karlsruhe, the Institute for Transuranium Elements (ITU).

The implementation of P&T involves research in three areas: (i) partitioning of long-lived radionuclides from the high level waste, (ii) development of fuel and targets containing these long-lived elements in view of their (iii) transmutation in various burners (fission reactors and accelerator driven transmutation devices). At present, the European Commission supports experimental work on partitioning both in the framework of the shared-cost programme and at ITU, fuel and target development at ITU and an overall strategy study on the potentialities of P&T for nuclear waste management as a shared-cost action.

This paper summarises the main research activities in the field of P&T supported by the European Commission and highlights some of the results obtained so far. Particular attention is paid to the strategy study, because the main theme of this 3rd NEA International Information Exchange Meeting on P&T is system studies. The detailed results obtained in these studies are presented by the different

research institutions involved in the course of this meeting. The activities described in this paper only represent a part of the research effort on P&T in the European Union, since some member States have their own national programme.

CHEMICAL SEPARATION OF LONG-LIVED RADIONUCLIDES

In the framework of its shared-cost activities, the European Commission has placed research contracts with CEA Fontenay-aux-Roses (F), the University of Reading (UK), KfK Karlsruhe (D) and ENEA Saluggia (I) to investigate experimentally the partitioning of actinides from high level liquid waste (HLLW). In the present studies, this operation is carried out in two steps: (i) removal of actinides and lanthanides from HLLW resulting from reprocessing of spent nuclear fuel in the PUREX process; (ii) partitioning between actinides (An[III]) and lanthanides (Ln[III]) using soft electron donor extractants.

CEA FaR is developing for step (i) the DIAMEX process, which uses diamides as extractants, and synthesizing with the University of Reading malonamides, which seem to be the most promising of this class of extractants [1]. Concerning step (ii), this university has synthesized tritertiary-butyl-PTZ and CEA FaR has obtained with this compound a better selectivity (>10) and a larger extraction efficiency than TPTZ for the Am[III]/Eu[III] separation in slightly acidic (pH>1) medium.

KfK Karlsruhe is studying different combinations of soft donor extractants and organic solvents to achieve step (ii). It has synthesized sulfur or mixed sulfur/nitrogen donor extractants. Factors of separation of Am[III] from Eu[III] of the order of 30 - 70 have been obtained with these extractants, but the extraction efficiencies remain insufficient.

ENEA Saluggia has recently started again to work on partitioning. It is using either Ph₂Bu₂ or CMPO for step (i) and the Talspeak process for step (ii) to extract the lanthanides.

On the other hand, CEA Cadarache (F), the Universities of Barcelona (E), Belfast (UK), Mainz (D), Parma (I), Twente (NL) and Strasbourg (F) and EHIC Strasbourg are involved in a research contract with the European Commission to synthesize and test new macrocyclic extractants (crown-ethers and calixarenes). The main aim of this contract is to selectively remove caesium, strontium and actinides from medium level liquid waste (MLLW) to decontaminate them to the extent that they can be disposed of in a near surface site. The research work is divided into three areas. Firstly, the macrocycles are synthesized by the Universities of Barcelona, Belfast, Mainz and Parma. Secondly, the extraction efficiency of the new compounds is determined by experiments of extraction and transport through supported liquid membranes by CEA Cadarache, EHIC Strasbourg and the University of Twente. Thirdly, the stability of the complexes between cations and macrocycles is simulated using softwares for molecular mechanics and dynamics by the Universities of Strasbourg and Twente and CEA Cadarache; the University of Parma is providing X-ray crystal structures of the complexes as starting points for molecular modelling. The main results obtained so far are: a calix[4]arene-crown-6 in the 1,3-alternate conformation synthesized by the University of Parma has a Cs/Na selectivity 100 times higher than that of the best current extractant for caesium; the large selectivity of this molecule has been explained by molecular mechanics and dynamics computations in the University of Strasbourg; the results obtained for Cs extraction from simulated MLLW have been confirmed with real HLLW; the Universities of Belfast and Mainz have synthesized new functionalized calixarenes, which can extract An[III] and Ln[III] more efficiently than CMPO.

The Institute for Transuranium Elements has been studying the extraction capabilities and the radiation stability of TRPO, a trialkyl phosphine oxide synthesized in China. Experiments were carried out with real HLW from reprocessing of WAK commercial spent fuel. TRPO showed excellent extraction properties for the actinides, the lanthanides and also technecium and high stability in the presence of α , β and γ radiation [2]. Similar tests are underway for other extractants like CMPO and DIDPA. In addition, a battery of centrifugal extractors has been installed in one of the chemical hot cells to have a better assessment of the different partitioning processes.

FUEL AND TARGET DEVELOPMENT

The Institute for Transuranium Elements prepared oxide fuels containing minor actinides for irradiation tests in the fast reactor PHENIX in France (SUPERFACT experiment). The analysis of the irradiated fuels enabled to determine the transmutation rate of minor actinides and the incurred occupational dose during handling of this material.

ITU has started a collaboration with CEA (F), ECN (NL), EDF (F) and KfK (D) in September 1992 to set up joint experiments for the study of materials for transmutation, including the fabrication and characterisation of fuels and samples, their irradiation and their in-pile behaviour [3]. The group is called EFTTRA (Experimental Feasibility of Targets for Transmutation). At present, it has been decided to study the transmutation of technecium (metal), iodine (compound) and americium (in an inert matrix). Some of the irradiation tests have already started in PHENIX and in the high flux thermal reactor HFR (NL).

STRATEGY STUDY ON THE POTENTIALITIES OF P&T FOR NUCLEAR WASTE MANAGEMENT

In the framework of its shared-cost actions, the European Commission has placed contracts with CEA (F), Siemens (D), ECN Petten (NL) and more recently with AEA Technology (UK) and Belgonucléaire (B) on a strategy study of Partitioning and Transmutation to assess its benefits for the safety of the management and storage of radioactive waste [4]. The main results obtained so far in these contracts are summarised below.

1)- Potentialities and costs of partition and transmutation of long-lived radionuclides

This study is carried out by CEA [5]. Its main objectives are to analyse the potentialities of a strategy for the management of radioactive waste aiming at reducing the inventory of long-lived radionuclides with P&T and to assess its technological requirements and costs. Reference scenarii without and with conventional reprocessing and scenarii using P&T are compared to assess the potentialities of P&T. The partitioning processes, the radionuclide inventories, the potential radiotoxicities and the costs are assessed for the different scenarii. As the main results of this study are described in detail in [5], only the results concerning potential radiotoxicity are discussed here.

Radiotoxicity has been chosen as a measure of the potential detriment of the waste resulting from the different scenarii analysed. This approach does not take into account the various barriers, natural and engineered, which prevent or delay the migration of the radionuclides disposed of in a deep underground repository. This may show the P&T option more attractive than it is in reality, where annual dose rates would be computed.

Reference and P&T scenarii

The three reference scenarii are:

- R1, with a reactor park having pressurised water reactors (PWR) solely, producing a total power of 120 GW, which is close to the present European capacity, and burning uranium oxide (UOX); the fuel cycle is open without reprocessing;
- R2, with the same reactor park as in R1, but burning UOX and MOX (uranium and plutonium oxide) fuel; the fuel cycle is closed with PUREX reprocessing of UOX and MOX; the losses during reprocessing are 0.3% for U and 0.5% for Pu;
- R3, similar to R2 until 2020; fast reactors (FR) are progressively installed after 2020; the losses during FR fuel reprocessing are 0.9% for U and 0.25% for Pu.

Three scenarii are considered for partitioning and transmutation, two with available technologies, RP1-1 and RP1-2, and one with futuristic technologies, RP2:

- the RP1-1 scenario is compared to the R2 scenario; the transmutation of neptunium and americium starts from 2010 in PWRs in homogeneous or in heterogeneous mode; in homogeneous mode, the UOX + actinide fuel is reprocessed as the standard UOX fuel; the losses during reprocessing are 0.3% for U, 0.5% for Pu, 5% for Np and Am and 100% for Cm; in heterogeneous mode, the specific targets containing either neptunium or americium oxide are irradiated during 5 years and are not reprocessed;
- the RP1-2 scenario is compared to the R3 scenario; as the minor actinide partitioning starts in 2010, Np and Am are stored before being recycled in FRs after 2020 either in homogeneous or in heterogeneous mode; in homogeneous mode, the fuel is reprocessed as the standard FR fuel; the actinide losses during reprocessing are the same as in the RP1-1 scenario; in heterogeneous mode, neptunium and americium oxide targets are irradiated during 15 years (3 cycles) and are not reprocessed;
- the RP2 scenario is similar to the RP1-2 scenario until 2030; CAPRA type FRs are progressively starting operation after 2030 to transmute neptunium in homogeneous mode and americium, technetium and iodine in heterogeneous mode; the fuel and targets are reprocessed with losses of 0.1% for U and Pu, 0.5% for Np, Am and Cm and 10% for Tc and I; curium is placed in interim storage.

Potential radiotoxicity inventory

The potential radiotoxicities are assessed for ingestion and mainly deal with all heavy radionuclides and three long-lived fission products (^{99}Tc , ^{129}I , ^{135}Cs). They are calculated for cooling times between 10 and 10^7 years from 2100. Two radiotoxicity inventories are considered: (i) a "waste" inventory for nuclear materials to be disposed of; (ii) a "cycle" inventory, which includes (i) and nuclear materials present in the fuel cycle (plants, storages, reactors and waste,....).

The time evolution of the potential radiotoxicity computed for the "waste" inventory in the case of the scenarii involving PWRs only (R2, homogeneous and heterogeneous RP1-1 scenarii normalised with respect to the R1 scenario) is shown in Fig. 1. The same is given in Fig. 2 for the incineration in FRs (R3, homogeneous and heterogeneous RP1-2 scenarii normalised with respect to R1).

Concerning the reference scenarii, there is a larger decrease in radiotoxicity for the R3 than for the R2 scenario. The reduction factor is larger than 5 for the R2 scenario between 10^4 and 10^5 years due to the recycling of plutonium. In the short term (10 - 10^3 years), the radiotoxicity is mainly due to ^{244}Cm , then to ^{241}Am . In the long term (10^3 - 10^5 years), there is a paramount contribution of ^{243}Am , its daughter ^{239}Pu and ^{240}Pu . In the very long term ($>10^5$ years), the radiotoxicity is dominated by ^{237}Np .

The reductions in radiotoxicity with respect to the R2 (resp. R3) scenario are nearly the same for both RP1-1 (resp. RP1-2) scenarii. There is an improvement by a factor of 6 between 10^2 and 10^3 year cooling time because of the decrease of Am inventory and between 5×10^5 and 5×10^6 year cooling time due to the smaller content in Np. For the short time periods (<100 years), the gains are offset by the increased production of Cm.

For the RP2 scenario, the radiotoxicity is reduced by more than a factor of 10 with respect to the R2 scenario for time periods smaller than 100 years, because curium is put in interim storage and is therefore not considered as a waste. For the other time periods, the reduction factor for the radiotoxicity is between 10 and 30.

The computation of the potential radiotoxicity of the "cycle" inventory shows that there is no gain for both RP1-1 (resp. RP1-2) scenarii with respect to the R2 (resp. R3) scenario because of the irradiated fuels and targets in storages and reactors. The radiotoxicity is increased for the R2 and RP1-1 scenarii up to 100 year cooling time due to the larger production of curium.

Cost assessment

The implementation of P&T of U, Pu, Np and Am with available technologies (homogeneous RP1-1 and RP1-2 scenarii) leads to a global cost increase of the overall fuel cycle between 10 and 50% with respect to the reference case where U and Pu are recycled only (R2 and R3 scenarii). This increase mainly reflects the larger reprocessing and minor actinide fuel fabrication costs for the P&T scenarii.

2)- Transmutation of long-lived radionuclides in fast reactors

Siemens has analysed the possibilities of fast reactors (FR) for transmutation of actinides and long-lived fission products [6]. The reference fast reactor is EFR with a thermal power output of $3600\text{MW}_{\text{th}}$, MOX fuel and a 1m core height. The possibility of enhancing transmutation by neutron spectrum hardening via leakage increase (smaller core height, smaller units) has been studied. Metallic and oxide fuel and homogeneous or heterogeneous insertion of waste are compared. The safety behaviour of the different cores is evaluated by the sodium void effect and the Doppler effect.

The main results of this study are summarised below.

Transmutation of minor actinides (MA)

The compromise between keeping the sodium void effect at an acceptable level and maximising the MA transmutation could be obtained in a FR with a large diameter core and a reduced height (0.7m). Such a core would be able to take 860 kg of MA and to transmute 65 kg of MA per year, which represents the production of about three PWRs.

The MA transmutation rate for oxide or metal fuel is nearly the same with a transmutation half-life of 11 years, taking into account the limitation of ^{238}Pu build-up of 5% of the total Pu imposed at present

by reprocessing. There is however a slight advantage of about 15% for the metal fuel concerning the transmuted mass per year because of the higher metal density in metal fuel. The positive sodium void effect is much more favourable for oxide than for metal fuel, but the negative Doppler constant is strongly reduced in metal cores. This is beneficial for loss of heat sink accidents, but a disadvantage in case of a transient overpower. On the other hand, metal fuel has a lower loss of reactivity and a longer residence time for the same maximum fuel burn-up than oxide fuel. There are thus no clear advantages for oxide or metal fuel to transmute MA in a FR.

The MA transmutation efficiency in fast reactors is similar for heterogeneous and homogeneous recycling, when considering the whole core, i.e. the MA containing subassemblies and the standard fuel subassemblies. About 30 to 35% of the initial MA content are transmuted during a subassembly lifetime of 6 years. A large amount of ^{238}Pu builds up in the MA containing fuel at high transmutation yields, which might cause difficulties for reprocessing and fuel fabrication. The safety parameters, sodium void effect and Doppler constant, are much more favorable for heterogeneous than for homogeneous recycling.

As the ratio lanthanides/(Am+Cm) in HLLW from spent UOX fuel reprocessing is about 20-50 depending on the cooling time and as americium and curium have the same valency III as the lanthanides, partitioning of minor actinides is not straightforward. For this reason, the influence of lanthanide admixture to the MA containing fuel on the main parameters of a FR core has been investigated. The lanthanides have a similar influence on the core behaviour as the MA: they increase the sodium void effect and reduce the Doppler effect. The admixture of MA and lanthanides has therefore to be limited to an upper value of 5% of the total mass of heavy metals. This ratio requires separation factors between MA and lanthanides around 30-50 in order to achieve transmutation efficiencies equivalent to those obtained without lanthanides. These conclusions have been confirmed by CEA in the framework of the strategy study on P&T.

Transmutation of long-lived fission products

It is difficult to transmute large quantities of fission products such as ^{99}Tc in the external zones of a FR core. Computations have then been carried out for ^{99}Tc irradiated in special moderated target subassemblies, which are distributed heterogeneously inside the core region. For a 1.2m core height with 84 ^{99}Tc target subassemblies and a thermal power output of $2600\text{MW}_{\text{th}}$, about 166kg of ^{99}Tc can be transmuted per year, which corresponds to the ^{99}Tc production of five to six 1GW_e PWRs. The effective transmutation half-life is around 26 years. The safety parameters of such a transmutation device are in the scope of conventional FR layouts. The presence of target subassemblies inside the core region even leads to a sodium void effect reduction. A macrocell study shows that, with an appropriate design of the moderated ^{99}Tc subassemblies, a moderator volume of up to 20% does not disturb too much the fuel pin power distribution in the vicinity of the moderated subassemblies.

When replacing some of the ^{99}Tc target subassemblies by ^{129}I ones, the transmutation rate of ^{99}Tc is not affected and about 22kg of ^{129}I could be transmuted in a FR with a transmutation half-life of around 44 years.

3)- Nuclear data libraries for transmutation studies

The four main aims of the work carried out by ECN Petten are:

- the preparation of a nuclear data base for transmutation of actinides and fission products by inspecting the European JEF-2 library and by paying special attention to long-lived fission product capture and (n, 2n) data;
- the assessment of the ORIGEN nuclear data library for transmutation studies;
- sample burn-up calculations for a few scenarii using the ORIGEN code and the updated ORIGEN nuclear data library;
- investigation of transmutation of long-lived fission products (^{129}I and ^{99}Tc): a possibility is neutron capture in a very high thermal neutron flux reactor such as the European High Flux Reactor (HFR) at Petten.

The main achievements of this study are presented below.

The cross section data bases have been updated for PWRs and FRs with data from the JEF2-2 and EAF-3 libraries. Three-group cross sections for the ORIGEN-S fuel depletion code and one-group cross sections have been calculated.

For PWRs, the cross sections strongly depend on burn-up, because the variations of the nuclide densities cause changes in the neutron spectrum and resonance integrals. The FR cross sections only show a slight dependence on burn-up. For both types of reactors, the cross sections at average burn-up are used for the updated data bases.

Burn-up computations for PWRs show that the nuclide densities obtained from the new cross section data base agree with those derived from the regular cross section updating within 20% and even much less than 10% for most nuclides.

A strategy study on transmutation of long-lived fission products in different reactors is being performed [7,8]. Computations indicate that the transmutation half-life of ^{99}Tc (resp. ^{129}I) is about 40 years (resp. 20 years) with a rate of 38 kg/year (resp. 46 kg/year) in heavy water reactors. These figures have to be compared with the annual production of ^{99}Tc (20 kg) and ^{129}I (4.6 kg) of a 1000 MWe LWR. With a special subassembly in the Petten high flux reactor, the calculated transmutation half-lives for ^{99}Tc and ^{129}I are about 8.6 and 5.7 years. In the case of transmutation in fast reactors, the results of computation for ^{99}Tc are in agreement with Siemens.

4)- Benchmark study on the burn-up of actinides in PWR MOX fuel

Belgonucléaire is identifying the possibilities, constraints and limitations of actinide and long-lived fission product recycling in PWRs with MOX fuel. This study includes three parts: (i) homogeneous recycling of MOX containing Pu and Am; (ii) impact on fuel refabrication; (iii) physics of fission product recycling in dedicated assemblies in a PWR core having a larger moderation. The calculations for parts (i) and (iii) are performed in close collaboration with ECN Petten.

The first results are the following. Recycling Am with Pu in MOX fuel limits the number of recycling steps to one instead of two or three [9]. The Pu + Am recycling strategy increases the dose rates by a factor of 4.5. An extra 25 mm steel shielding is required to reduce the dose rates to the values corresponding to Pu recycling only.

5)- Arisings of secondary waste from processes for P&T of long-lived radionuclides

AEA Technology is assessing the secondary waste that arises from different processes for the partitioning of actinides and long-lived fission products and from the fuel and target fabrication [10]. The plant requirements, indicative costs and doses to operators for these processes will be assessed and compared with a reference fuel cycle without P&T. The separation systems under investigation are the CTH, TRPO, TRUEX and DIAMEX processes and a typical non-aqueous (pyrochemical) process.

CONCLUSION

Progress is being carried out in the field of chemical separation; in particular, very efficient macrocyclic extractants have been synthesised and tested by seven universities and one research centre from six countries of the European Union in the framework of the shared-cost actions of the European Commission.

Fuel and targets are developed by the Joint Research Centre of the European Commission in Karlsruhe in collaboration with some research institutions of the European Union.

Concerning the strategy study on the potentialities of P&T performed in the framework of the shared-cost actions, the main results can be summarised as follows:

- compared to scenarii where uranium and plutonium are recycled only, the additional recycling of 95% of americium and neptunium leads to a reduction by a factor of about 6 in the potential radiotoxicity (without barriers) of the waste to be disposed of between 10^2 and 10^3 (mainly due to Am removal) and between $5 \cdot 10^5$ and $5 \cdot 10^6$ year cooling time (mainly due to Np removal); the radiotoxicity of the waste resulting from actinide incineration is smaller for fast reactors than for PWRs; Am and Np recycling increases the global cost of the overall fuel cycle by 10% to 50%;

- computations show that it is possible to transmute the amount of minor actinides produced by about three PWRs with a transmutation half-life of around 10 years in a fast reactor without compromising its safety behaviour; concerning long lived fission products, the ^{99}Tc production of 5 to 6 1GWe PWRs could be transmuted in a fast reactor with a transmutation half-life of about 25 years;

- from safety and burning computations, separation factors between minor actinides and lanthanides around 30-50 are necessary to transmute minor actinides in fast reactors.

This strategy study points out some of the open questions, which remain to be solved before implementing P&T. Among them, efforts should be devoted to partitioning of americium, which is responsible for a large part of the short-term and very-long-term (through ^{237}Np formation) potential radiotoxicity. Great care should be taken that a P&T scenario does not lead to the generation of unacceptable amounts of secondary waste and does not increase the doses incurred by man. Finally, the question of the number of minor actinide recycling steps should be properly taken into consideration (feasability of fabrication and reprocessing of fuel and targets containing minor actinides and unavoidably some lanthanides).

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IMPORTANCE OF DOUBLE STRATA FUEL CYCLE FOR MINOR ACTINIDE TRANSMUTATION

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Abstract

The reactor physics characteristics of MA transmutation systems are compared between LWR, FBR and Actinide Burner Reactor (ABR). The influence of transmutation reaction products on radiation dose in fuel cycle facilities is discussed. The generation of these neutron emitters varies depending on neutron spectrum hardness of a transmutation system. The preferable feature of a dedicated MA transmutation system is compared with recycling scheme of MA in power reactor from the fuel cycle facility view point.

1. Introduction

Various systems and concepts have been proposed for transmutation of long-lived nuclides as an application of reactor technology for high-level radioactive waste (HLW) management. Recently, the application of transmutation concepts for excess Pu burning is being discussed. Pu recycling together with minor actinide (MA) has been proposed considering nuclear proliferation resistance. For the technical judgement of these concepts, most of discussions are based on consideration for such reactor engineering as fuel related matters and reactor performance. However, when the economy of these concepts is considered, fuel cycle related issues shall be taken into account.

In this paper, the influence of MA recycling in various types of reactors on fuel handling is discussed.

2. Comparison of MA transmutation from the reactor physics view points

Transmutation system concepts can be classified in two groups. The one is MA recycling in power reactors, LWR or fast reactors, and the other is MA burning in dedicated systems, actinide burner reactors or accelerator-driven hybrid systems. When MA is recycled in a power reactor, the acceptable amount of MA in power reactor fuel is limited since the addition of MA in fuel has large reactivity effect for reactor performance and major reactor parameters of power reactors are already optimized from safety and economical view points on the other hand. Therefore, addition of MA should not result in large change of these parameters. For the concepts of MA recycling in PWR, MOX-PWR (Pu usage in PWR) and MOX-FBR, the maximum allowable amount of MA will be 0.2, 0.5 and 5wt% of fuel material, respectively.

When a dedicated transmutation system becomes available, the scheme of an entire fuel cycle will be a strata structure fuel cycle. The concept of the double strata fuel cycle consisting of a conventional power reactor fuel cycle and a Partitioning-Transmutation (P-T) cycle is illustrated in Fig. 1.¹⁾ The final HLW from the double strata fuel cycle contains only short-lived and stable fission products. The separate treatment of MA from the commercial cycle will be preferable for the conventional fuel cycle because MA is a strong neutron emitter and the MA recycling in the conventional power reactor fuel cycle will introduce the problem for fuel handling.

As an example of dedicated transmutation systems, the reactor core design parameters of two types of ABRs are shown in Table 1. Fuel material of these ABRs is MA-enriched uranium nitride mixture. One of ABRs is a lead-cooled pin fuel ABR (L-ABR) and the other is a He-cooled particle fuel ABR (P-ABR).²⁾ In these ABRs, neutron energy spectrum is very hard and the core averaged neutron energy is around 720keV. These very hard neutron spectra are very effective for direct fission of MA which has fission threshold at around 600keV.

In Table 2, the transmutation characteristics are compared between P-ABR and power reactors, namely PWR, MOX-FBR and a metal fuel FBR. For the MA transmutation in power reactors, the concentration of MA is assumed as 0.2% and 5% of heavy metal for PWR and FBR, respectively.

In the table, the transmutation ratio is defined as the ratio of MA weight at the end of cycle to that of the beginning of cycle. In this definition, any nuclear reaction such as fission, neutron capture, (n, 2n), etc. can be considered as transmutation reaction and the conversion of Np-237 into Pu-238 is a part of transmutation of Np. On the other hand the MA burnup ratio is defined as the ratio of fissioned MA weight to that at the beginning of cycle. The latter is the real index of transmutation effectiveness and efficiency because only fission is a real transmutation reaction to solve the problem of long-lived MA management.

The transmutation and the burnup ratios of power reactors in Table 2 are the net ones after the MA generation in fuel being deducted. The negative value of burnup ratio of U-PWR implies that new MA generation in fuel prevails over fission of MA which is added in fuel for transmutation. The large discrepancy between the transmutation ratio and the burnup ratio in power reactors indicates the larger conversion of Np into Pu than in ABRs. The net MA burnup per 1GWt a year of ABRs is significantly larger than that of power reactors because in ABRs major fuel material is MA. The support factor which is defined as the number of power reactor units of which MA is transmuted by one unit of a transmutation system is about 10 to 15 for dedicated transmutation systems, while the support factor of FBR proposed so far is between 4 to 6.

In Table 3, the transmutation characteristics of Np-237 are compared between P-ABR, MOX-FBR and PWR. In this analysis, Np-237 is continuously irradiated with neutron flux, of which level is indicated in the table. In Table 3a, the cumulative fission fraction and the fraction of fission which undergoes as neutron capture products of Np-237 are shown as wt% of initial Np-237. In P-ABR which has very hard neutron spectrum, most of fission occurs as Np-237 or Pu-238 and total fission after 6000 days irradiation is 98.6% of initial Np-237. In MOX-FBR, fission of Np-237 is dominant as Np-237, Pu-238 and Pu-239, and total fission in 6000 days is only 75%. In PWR, Np undergoes fission as Pu-239 or 241, and total fission is 92% which is larger than that of MOX-FBR. In Table 3b, residual actinide after irradiation is shown. In PWR, generation of americium and curium from Np-237 is significant. From this table, it is evident that very hard neutron spectrum is favorable for MA transmutation. In PWR or under thermal flux, transmutation or fission of MA is slow at the beginning of irradiation but after the sufficient conversion of MA to fissionable isotopes, transmutation is very efficient. In PWR, however, generation of heavier MA is serious problem.

3. Influence of MA transmutation products on fuel handling at fuel cycle facilities

There are several criteria for engineering judgment of transmutation systems. At present, most of system evaluations are discussed from the reactor engineering view points, namely, transmutation rate, influence of MA on reactor performance, fuel related issues. The other important point for judgement is the influence of transmutation products on the fuel cycle facilities. In Table 4, the increase of heavier MA generation and neutron emission rate in spent fuel are shown for the case of 0.2 wt% MA addition to PWR fuel. Only 0.2wt% of MA addition results in 700 times increase of Cf and 7 times increase of spontaneous neutron emission thereof. To evaluate the effect of these heavier nuclides, most of which are strong neutron emitters, the increase of decay heat, neutron emission and γ -ray intensity was calculated when MA is added to power reactor fuels.

In this analysis, typical power reactors for MA recycling scenario was selected. Power reactors selected were PWR, MOX-PWR and MOX-FBR. The effect was calculated for both fresh and spent fuels of these reactors. In the present calculation, the fraction of MA in the fuels are 0.2wt% heavy metal of PWR fuel and 5 weight % of MOX-PWR and MOX-FBR fuels. The fuel burnup is 45, 45 and 85GWD/HMT, respectively. The cooling time of spent fuel is 10 years. In Table 5, the result of the analysis is summarized. The increase of decay heat and γ -ray emission are at the acceptable level but the increase of neutron emission in fresh fuels is large.

The influence of neutron emission increase on fuel cycle facilities was evaluated for the scenario of MA recycling in these power reactors. Facilities and fuel handling considered are a) fuel manufacturing, b) fresh fuel transportation, c) spent fuel storage, d) spent fuel transportation and e) reprocessing. The evaluation was made for the process where the decay heat and radiation dose are possibly severest.

a) Fuel manufacturing process

The oxide powder mixture process is the one where the quantity of fuel material will be largest throughout fuel manufacturing. The powder mixture machine was simulated as a ball of 0.5mm thick stainless steel wall. The powder density is 2 g/cm³. In the case of PWR fuel, 3 ton of UO₂ powder is contained in a mixture machine of 142 cm diameter. In the case of MOX-PWR fuel and MOX-FBR fuel, 300kg MOX powder is contained in a mixture machine of 66cm diameter. The calculation model is illustrated in Table 6.

The values in the table are the ratios of dose from fuel material powder which contains MA to that without MA. The radiation dose increase in the PWR fuel with 0.2wt% MA addition is prohibitably high as about 10⁴. In the case of MOX-PWR and MOX-FBR fuel, the increase is 50 to 100 times. The significant increase of neutron dose results in the significant reinforcement of radiation shielding in the fuel manufacturing facility for handling fuel even with very small fraction of MA addition.

b) Fresh and spent fuel transportation

Radiation dose and decay heat at a fuel cask were evaluated.

c) Spent fuel storage

Spent fuel storage facility was selected for the evaluation.

d) Reprocessing facility

Radiation dose and decay heat at a feed preparation tank was evaluate. The calculation model is shown in Table 7. The volume of the tank is 25m³ and the wight of heavy metal in liquid solution is 8.75ton which is calculated from the subcriticality density limit of 36g/l. The cooling

time of spent fuel is 4 years. The increase of the radiation dose is 1.2 to 2 times of that of normal fuel.

In Table 8, the influence of MA contained fuel on fuel cycle facilities is summarized for MA recycling in PWR, MOX-PWR and MOX-FBR. The significant radiation shielding reinforcement is required for fuel manufacturing and fresh fuel transportation. The reinforcement of radiation shielding in fuel cycle facilities will result in the increment of fuel cycle cost.

For the fuel handling in fuel cycle facilities of a ABR or an accelerator-driven system, radiation shielding and decay heat removal are much severer problem than that of MA recycling in power reactors since the concentration of MA is very high in these dedicated systems. The fuel cycle facilities for dedicated systems, however, are very compact and the required number of these facilities is small because nitride fuel can be reprocessed by pyrochemical process and the mass flow is small compared with those of conventional fuel cycle.

4. Conclusion

As a result of increase of heavier MA for the MA transmutation scheme in power reactors, the radiation shielding reinforcement will be needed for the fresh fuel handling (manufacturing and transportation). This may cause the cost increase of the of electricity generation. In the case of a dedicated system, the shielding and the decay heat removal are much severer problem than the MA transmutation in power reactors. However, the cost of construction and operation of compact facility even with heavy radiation shielding and remote handling may not be significant compared to that of large scale facility with medium radiation shielding.

The cost comparison of the fuel cycle between MA recycling in power reactors and MA recycling in a dedicated system is inevitable for judgement of transmutation system.

From the socio-techno view point, the confinement of troublesome MA in one closed site will be very important. In this sense, the double strata fuel cycle concept can provide the closed HLW management park concept.

At the time of MA transmutation system selection in the future, the criteria for the selection should be established not only from the reactor or system performance view points but also from the fuel cycle facility view points.

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Table 1 Reactor design parameters of modified Actinide Burner Reactors

	L-ABR ¹⁾	P-ABR ²⁾
Fuel concept material	pin-bundle (64NpAmCm-36U ³⁾) _{1,0} N ⁴⁾ _{1,0}	coated particle (65NpAmCm-35U ³⁾) _{1,0} N ⁴⁾ _{1,0}
MA initial loading,kg	918	2870
MA/U	588/330	1865/1005
Reactor power, MWth	180	1200
Coolant material	Lead	Helium
Neutron flux, 10 ¹⁵ n/cm ² · sec	3.1	6.6
Core averaged mean neutron energy, keV	700	700
Reactivity (% Δk/k)		
Coolant-void reactivity/core	-1.3	—
Doppler reactivity/core (Δt=300°C)	-0.01	-0.01
Kinetic parameters		
β _{eff}	2.6 × 10 ⁻³	2.6 × 10 ⁻³
L _p , sec	1.3 × 10 ⁻⁷	1.5 × 10 ⁻⁷
Cycle length, full-power days	550	300
MA burnup, %/cycle	11	13

- 1) L-ABR: MA nitride fuel with lead cooling burner reactor
- 2) P-ABR: MA particle fuel burner reactor
- 3) 90% enriched uranium
- 4) ¹⁵N enriched

Table 2 Comparison of MA transmutation in various reactors

	MA Burner Reactors	Power Reactors		
	P-ABR	U-PWR	MOX-FBR	LMR ¹⁾
Output (MWt)	1200	3410	2600	2632
Cycle length ²⁾ (EFPD)	300	850	1368	900
Core averaged				
Fast neutron flux (× 10 ¹⁵ n/cm ² · s)	5.9	0.37	4.1(3.0)*	6.1(4.1)*
Mean neutron energy (keV)	720	thermal	480	490
MA loaded (kg)	1865	180 ³⁾	1450 ³⁾	1200 ³⁾
MA transmutation ratio ⁴⁾ (%/cycle)	18.8	9.0	33.5	29.8
MA burnup ratio ⁵⁾ (%/cycle)	13	-23.8	8.8	8.3
MA transmutation(kg/1GW · year)	292	1.7	40.9	44.5
MA burnup (kg/1GW · year)	202	-4.4	10.7	12.4

- 1) Metal fuel FBR
 - 2) Effective fuel irradiation time
 - 3) Concentration of MA in fuel ; 0.2% for U-PWR, 5% for MOX-FBR and LMR
 - 4) MA transmutation ratio={MA(BOC) - MA(EOC)}/ MA(BOC)
 - 5) MA burnup ratio=(MA fissioned-MA generated) / MA(BOC)
- Values of 4) and 5) are those for the equilibrium cycle in ABRs, and average values of 1 to 10th cycle for power reactors.
- *) Values in the parentheses are for the outer core

Table 3 Comparison of transmutation of Np-237 between different neutron spectra of ABR, MOX -FBR and PWR

Table 3a ²³⁷Np Fission
(unit:% of initial ²³⁷Np)

Reactor	Flux (×10 ¹⁵)	Cycle No.	Cumulative Fission	Fissioned as					
				Np-237	Pu-238	Pu-239	Pu(0+1)	Am	Cm
P-ABR	8.4	1	13.2	10.6	2.4	0.13	<0.01	<0.01	<0.01
		10	87.1	35.0	36.9	12.1	1.01	0.02	<0.01
		20	98.6	36.1	41.0	15.5	2.20	0.09	0.02
MOX-FBR	3.3	1	3.4	2.80	0.57	0.02	<0.01	<0.01	<0.01
		10	44.0	15.1	21.4	6.24	0.22	<0.01	<0.01
		20	74.7	18.1	35.1	15.9	1.26	0.04	<0.01
U-PWR	0.37	1	1.8	0.41	0.24	0.70	0.03	<0.01	<0.01
		10	65.1	1.61	5.35	39.4	13.9	0.26	0.38
		20	91.6	1.71	6.70	51.4	21.2	0.47	2.05

Table 3b ²³⁷Np Capture
(unit:% of initial ²³⁷Np)

Reactor	Flux (×10 ¹⁵)	Cycle No.	Residual Actinide	Capture to					
				U	Np-237	Pu-238	Pu-239	Am	Cm
P-ABR	8.4	1	86.8	0.05	71.3	14.3	0.90	<0.01	<0.01
		10	12.9	1.74	3.23	4.40	2.33	0.03	<0.01
		20	1.4	0.53	0.13	0.21	0.17	0.02	<0.01
MOX-FBR	3.3	1	96.6	0.04	85.1	11.0	0.37	<0.01	<0.01
		10	56.0	5.22	20.0	22.2	7.12	0.03	<0.01
		20	25.3	6.34	4.06	7.61	4.55	0.15	0.01
U-PWR	0.37	1	98.2	0.07	75.8	19.9	1.87	<0.01	<0.01
		10	34.8	4.43	6.36	14.4	2.92	0.82	1.59
		20	3.3	2.29	0.58	1.80	0.38	0.33	1.73

One cycle ; 300 days irradiation and 3 years cooling

Table 4 Effect of MA addition to power reactor fuel handling

Reactor/Fuel	Ratio of value of MA added fuel to that of normal fuel		
	Decay heat	Neutron emission	Photon intensity
U-PWR (0.2wt%) ^a			
fresh fuel(U235:4wt%) ^b	3.6×10^3	8.3×10^4	1.3×10^3
spent fuel(45GWD/t) ^c	1.5	4	1
MOX-PWR (0.5wt%)			
fresh fuel(Pu:6.5wt%)	1.4	4.8	1.2
spent fuel(45GWD/t)	1.5	1.7	1
MOX-FBR (5wt%)			
fresh fuel(Pu:30wt%)	2.2	1.0×10^2	2.1
spent fuel(80GWD/t)	2.8	19	1

a: minor actinides(MA) fraction in fuel (HM weight %)

b: fuel enrichment

c: fuel burnup (cooling time : 10 years)

Table 5 Effect of MA addition in PWR fuel

(values : per ton of HM)

Item	Reference PWR	MA-PWR	(MA-PWR)/(Ref.PWR)
Nuclide (g)			
Np	469	918	2.0
Am	162	276	1.7
Cm	38	296	7.7
Bk	3.4×10^{-7}	1.7×10^{-4}	510
Cf	3.5×10^{-7}	2.4×10^{-4}	690
α -activity (10^5 Ci)	0.29	1.3	4.6
(α, n) (10^6 n/s)	6.3	36	5.8
Spnt. fission(10^9 n/s)	0.49	3.5	7.1

MA addition : 0.2% of HM

U enrichment : 3.2%

Burnup : 33000MWD/T

Irradiation : 847 days

Cooling : 150 days

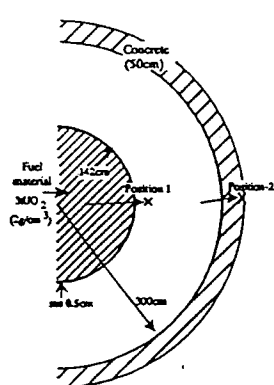
Table 6 Effect of MA on radiation dose of fuel manufacturing facility
 (values: ratio of dose from fuel material with MA to that without MA)

Fuel	Position of dose	Neutron	γ	Total
U-PWR	1	8.3×10^4	7.9×10^2	9.6×10^3
	2	8.3×10^4	3.3×10^3	2.8×10^4
MOX-PWR	1	48	6.5	41
	2	48	26	45
MOX-FBR	1	100	12	79
	2	97	86	96

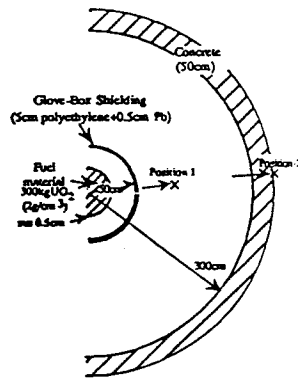
Position-1: 1m from powder mixing machine for PWR

1m from Glove Box Shielding for MOX-PWR and MOX-FBR

Position-2: Outer surface of 50cm thick concrete placed at 3m from the center



Calculation Model for PWR



Calculation Model for MOX-PWR and MOX-FBR

Table 7 Effect of MA on radiation dose at feed preparation tank of reprocessing plant
(values: ratio of dose from fuel material with MA to that without MA)

Fuel	Neutron	γ	Total
U-PWR	4.4	1.8	2.0
MOX-PWR	1.2	1.2	1.2
MOX-FBR	---	---	$\sim 2^*$

Position : Outer surface of concrete wall (2m thick)

Fuel material: 8.75MTHM (360g U+Pu/ℓ, volume 25m³)

* Estimated value from the case of MOX-PWR

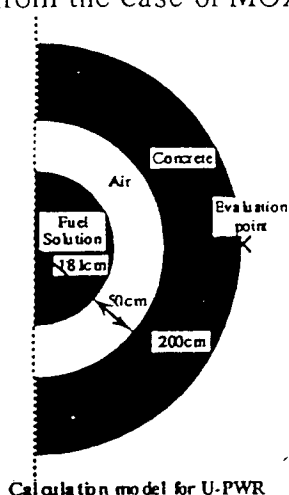


Table 8 Effect of MA on shielding design of fuel cycle facilities
when MA added to power reactor fuels

Reactor \ Process	Fuel Manufacturing	Fresh Fuel Transportation	Spent Fuel Storage	Spent Fuel Transportation	Reprocessing
U-PWR	S	S	N	M	M
MOX-PWR	S	S	N	M	M
MOX-FBR	S	S	N	M	M

S: Significant effect, radiation dose increases by order of magnitude

M: Minimum effect, radiation dose increases factor of two or less

N: Negligible effect

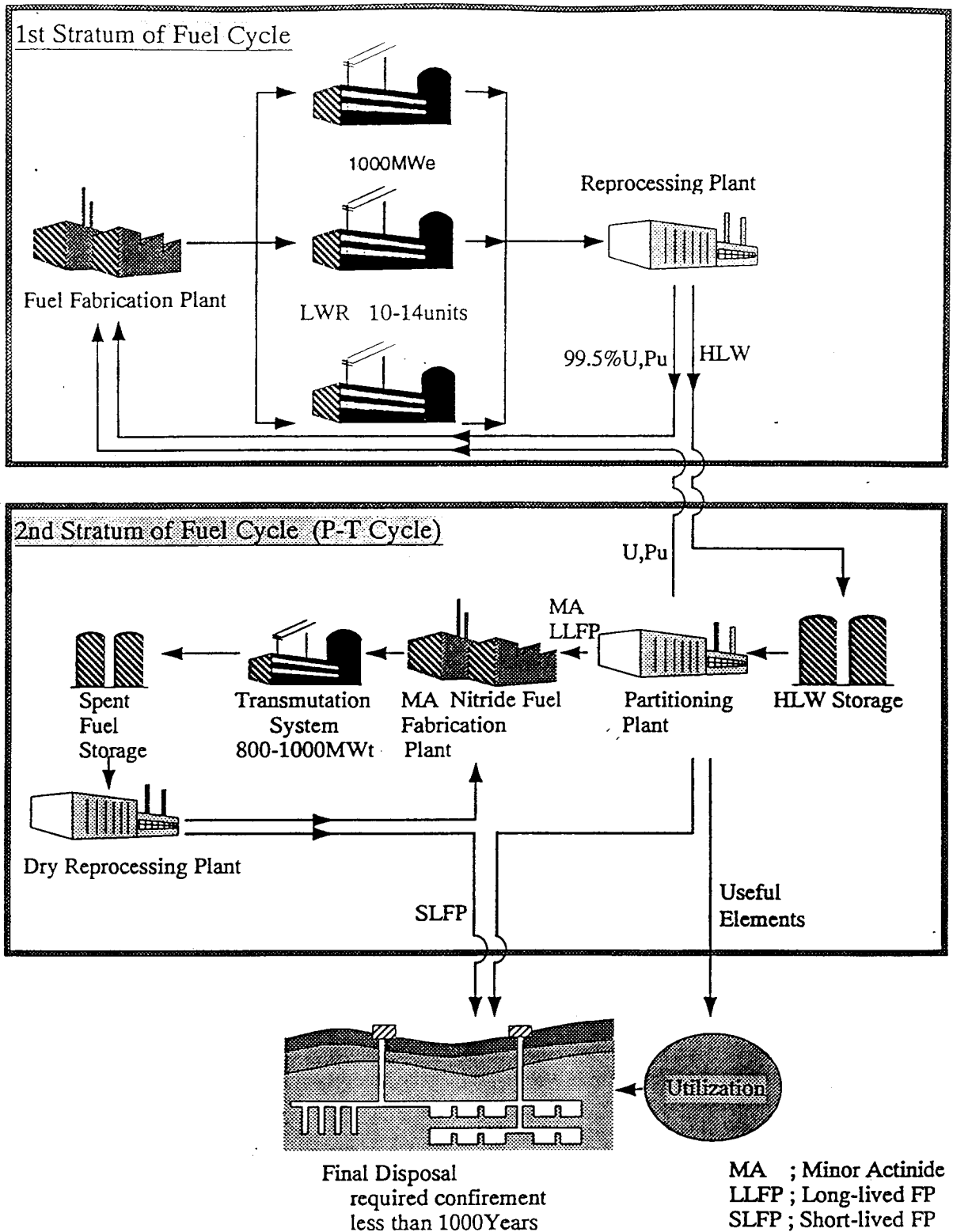


Fig. 1 JAERI's Concept of Double Strata Fuel Cycle for Complete HLW Management

IMPACT OF HIGH BURNUP IRRADIATION AND MULTIPLE RECYCLE OF PLUTONIUM AND MINOR ACTINIDES ON THE FUEL CYCLE ACTIVITIES

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ABSTRACT

The paper shows the impact of increasing burnups and recycle of MOX fuel in thermal and fast reactors. Reprocessing of the targets for multiple recycling will become increasingly difficult as the burnup increases from 4 to 7 At % in LWR-MOX fuel and 12-20 % in fast reactor fuel.

Multiple recycling of Pu + MA in fast reactors is a feasible option which has to be studied very carefully: Pu, Am and Np levels decrease as a function of the recycle number, Cm accumulates and gradually transforms into Cm-245, 246 and 247. Long cooling times are necessary with aqueous processing.

At each recycle step PUREX reprocessing will produce a concentrated HLW solution which contains all the fission products, minor actinides and U-Pu residues. These solutions are the source terms for partitioning operations.

The presently developed partition operations DIDPA, TRUEX, DIAMEX and TRPO do extract quantitatively the actinides from the concentrated waste solution. The Am-Cm fraction is a mixed actinide-lanthanide product stream which has to be treated for partial lanthanide removal.

New fuel fabrication technologies will have to be developed in order to cope with increased Pu-238 levels. The preparation of fuel or targets containing Am-Cm is the biggest challenge to be overcome.

1. Introduction

The recent evolution in the nuclear fuel cycle management is influenced by a number of important technical and political facts which have changed the traditional outlook of this nuclear activity.

- There has been a general tendency in reactor fuel management to increase the burnup of irradiated UO₂ fuel from 33 to 43 Gwd/T and even burnups beyond that value are considered.
- The reprocessing capacity has doubled but is not yet capable of keeping pace with the production of spent fuel, increasing furthermore the backlog of spent fuel. A number of countries have modified their position towards reprocessing as reference fuel cycle option.
- The use of separated Plutonium has shifted from Fast Breeder Reactors to Light Water Reactors. The potential market for LWR-MOX fuel is very large but depends on the availability of LWR-MOX fuel fabrication plant-capacity.

- The quantity of separated Plutonium exceeds the present usage trend and the potential arrival of high quality Plutonium from military origin with an outspoken proliferation risk puts the emphasis on Plutonium rather than on Minor Actinides.

- Due to technical and institutional reasons the geologic disposal approach has further been shifted into the future and there is time to develop technologies to decrease the potential toxicity of Actinides and certain fission products.

2. Evolution of quantities of actinides and their radiotoxicity as a function of burnup

2.1. UO_2 fuel

Most of the earlier radiotoxicity - and actinide - inventory calculations were made on the basis of UO_2 fuel irradiated up to 33 GWd/T. [1], [2].

This burnup rate has since long time been surpassed by operational power plants. The present standard burnups are close to 43 GWd/T and this results in an increase of the fuel irradiation time, an increase in actinide generation per unit of weight of fuel and an increased residual heat. By doing so the specific fuel cycle costs, the residual Pu and the residual fissile contents decrease sharply. Table 1 gives an overview of this tendency.

Table 1
Influence of increasing burnup on Pu inventory of UO_2 fuel [3]

Burnup GWd/THM	THM/GWe- year (*)	kg Pu/THM	Annual Plutonium inventory		
			Total kg/GWe	Fiss. %	Fissile kg/GWe
33	33.5	9.17	307	69	212
40	27.6	10.24	283	66.5	188
50	22.1	10.86	240	63.2	151
60	18.4	11.74	216	58.8	127

(*) 33 % electric yield - to be multiplied with the load factor (0.75 to 0.85) to obtain net values per year.

With a load factor of 80 % the total Pu inventories range from 246 to 173 kg Pu/GWe - 0.8 year. The radiotoxicity of the discharged spent fuel does not follow this trend due to an important increase of the Pu-238 and Cm-244 content of this fuel. Table 2 shows the evolution of the radiotoxicity as far as this property is equated to the α content of the fuel.

Table 2
Radiotoxic inventory of highly irradiated UO₂ fuel in Ci α/THM
7 years after discharge from the reactor

Burnup GWd/THM	Plutonium		Np-237	Am-241	Cm-244	α Total
	Total	Pu-238				
33	3 213	2 360	0.15	1 310	1 490	6 050
40	4 430	3 480	0.40	1 500	2 700	8 750
50	6 720	5 740	0.51	1 950	5 700	14 300
60	9 270	8 220	0.64	2 430	11 600	23 500

By comparing Table 1 with Table 2 it is striking that while the total Pu content is decreasing the radiotoxicity increases when expressed in total α per GWe year.

Burnup	Total Ci α / GWe-year	Net Ci α / 0.8 GWe-year
33	2.02 10 ⁵	1.61 10 ⁵
40	2.41 10 ⁵	1.92 10 ⁵
50	3.16 10 ⁵	2.52 10 ⁵
60	4.31 10 ⁵	3.45 10 ⁵

There is a slightly more than linear increase in the total α activity released per GWe-year and 80 % of this value per installed GWe for a mean load factor of 80 %.

The reprocessing plants which have to process high burnup fuel will have to cope with a steadily increasing α toxicity due essentially to Cm-244, Pu-238 and to a lesser extent to Am-241. The half-lives of these radionuclides are respectively 18.1, 87.7 and 432 years. Delayed reprocessing will only partially influence the Cm-244 contribution, the other two strong α emitters do not decay appreciably within a technological time frame of e.g. 20-30 years.

2.2. MOX fuel

Recycle of recovered Pu from reprocessed UO₂ fuel has shifted from FBR's to LWR's. The industrial recovery of plutonium from MAGNOX (low burnup) and more recently from UO₂ fuel (33 GWe/T) has led to the creation of a considerable stock of separated plutonium. According to well informed agencies (OECD, IAEA) this stockpile amounted to 92 THM Pu at the end of 1992.

The large reprocessing units (La Hague, THORP) produce together 28 THM Pu/year at full capacity. The degradation of the plutonium spectrum due to the decay of Pu-241 into Am-241 is the main drawback for keeping a stockpile unused apart from the economic penalties.

The typical isotopic composition of different types of plutonium used industrially at the MOX fuel fabrication plant of BELGONUCLEAIRE (Dessel, Belgium) are shown in Table 3.

Table 3
Typical isotopic compositions of plutonium processed at BN for making MOX fuel

Isotope	A	B (gas-graphite reactor)	C (LWR)	D (LWR)	E (LWR recycled)
Pu-238	0	0.05	0.5	1.5	2.3
Pu-239	94.0	79.0	43.1	58.5	48.6
Pu-240	5.45	17.0	40.0	25.0	27.3
Pu-241	0.50	3.5	5.9	10.0	14.8
Pu-242	0.05	0.45	10.5	5.0	7.0

The incentive to use the separated plutonium in LWR-MOX is very high since this is at the present time the main outlet for plutonium. The previous application in fast breeder reactors is presently at a very low gear and the facilities have been either closed or reoriented to LWR or Pu incineration purposes.

By blending plutonium from different origins it is possible to prepare MOX fuel with a reasonable high fissile plutonium content (65-70 %). The potential occurrence of excess plutonium from military origin, with its very high fissile content (Table 3, type A), on the nuclear fuel market would greatly improve the flexibility of MOX use in LWR's and increase the recycle frequency and the time span during which plutonium would contribute to the LWR-fuel cycle.

Two nuclides are limiting the use of recycled plutonium in fresh MOX fuel: Pu-238 and Am-241. In manually operated MOX fuel fabrication plants a concentration level of 1.5 % Pu-238 is acceptable; above that level special neutron shielding has been installed.

The Am-241 content depends on the initial Pu-241 content and the time interval between reprocessing and fuel fabrication. There is no technical procedure to influence the ingrowth once the PuO₂ powder has been delivered to the fuel fabrication plant. The unbalance between the plutonium output of the large LWR-UO₂ reprocessing plants (La Hague, THORP) and the available fuel fabrication facilities is undoubtedly the most severe constraint on the industrial use of MOX fuel.

Two strategies could be pursued to solve this serious problem:

- develop Pu purification units to remove Am-241 from old plutonium stocks,
- design and operate fully shielded and remote handled MOX fuel fabrication facilities.

In the first option a solution can be found for the use of old plutonium stocks. The second option is much more difficult to realize but offers perspectives for the production of MOX fuel with a significant minor actinide content. The economic and radiological impact of each of the two options will determine which one has most merits to be implemented on industrial scale.

Because of the increased fuel fabrication costs the irradiation of MOX fuels to higher burnups was assumed from the beginning. Burnups of 40 to 60 GWd/T are currently being considered.

Since the aimed burnup determines the necessary initial Pu_{Fiss.} enrichment and as the Pu_{Fiss.} content of the discharged UO₂ fuel varies according to its burnup and cooling time (see Table 1), this complex interaction will result in a total plutonium concentration which generally varies from 6 to 8 % Pu total.

After irradiation the composition of spent MOX fuel is approximately shown in Table 4.

Table 4
Indicative spent MOX fuel composition (7 years cooling) [4]

Burnup GWd/T	33	40	50
Initial total Pu %	5.03	5.58	5.58
End concentration Pu %	3.56	3.8	3.50
Plutonium isotopic composition % after 7 years			
Pu-238	2.7	2.6	3.0
Pu-239	39.0	40.8	37.4
Pu-240	34.0	32.4	32.8
Pu-241	14.5	13.6	13.8
Pu-242	9.8	10.3	12.7
Minor Actinide content kg/THM			
Am-241	1.95	2.56	2.42
Am-243	1.07	1.05	1.28
Cm-244	0.37	0.428	0.635
Cm-245	0.03	0.05	0.08

These data are only approximative and need further certification since the initial composition plays a very important role in the production rate of the heavy minor actinides. Experimental verification of the actinide composition based on post irradiative analysis of discharged MOX fuel with a very good irradiation history should be carried out in order to quantify with more accuracy the most critical nuclides: the plutonium isotopes in particular Pu-238, the minor actinides especially Am-241 and Cm-244. The ARIANE programme proposed by BELGONUCLEAIRE is a very good initiative in this direction.

Nevertheless a rough calculation based on these actinide yields shows that the overall α -load from MOX fuel is much higher than that of UO_2 fuel at the same burnup. Table 5 shows the global values.

Table 5
Radiotoxic inventory of highly irradiated MOX fuel in Ci α /THM 7 years after discharge

Burnup GWd/THM	Plutonium		Minor Actinides		Total α load	
	Total	Pu-238	Am-241	Cm-244	Total α	Total/GWe-y
40	$2.07 \cdot 10^4$	$1.69 \cdot 10^4$	$8.70 \cdot 10^3$	$3.45 \cdot 10^4$	$6.40 \cdot 10^4$	$1.76 \cdot 10^5$
50	$1.92 \cdot 10^4$	$1.81 \cdot 10^4$	$8.27 \cdot 10^3$	$5.12 \cdot 10^4$	$7.90 \cdot 10^4$	$1.75 \cdot 10^6$

Expressed per 1 GWe year discharged fuel the α radioactivity to be handled in a conventional reprocessing facility is about 5 to 7 times higher with MOX than with the corresponding UO_2 fuel as can be deduced from comparing the data of Table 5 with those of Table 2.

However in the near future only 30 % of the reactor core will be loaded with MOX fuel. By "diluting" 1 MOX fuel element with 2 UO_2 fuel elements the increase of the α radioactivity load is reduced to a factor 2 to 3.5 in the reprocessing plant operations compared to the equivalent LWR UO_2 fuel.

3. Source term evolution in multiple recycling

3.1. Recycle of UO_2 -fuel

The possible recycle of reprocessed U (U_{Repro}) has been discussed in an earlier paper [5]. The most important conclusion yet, is that in the present economic circumstances only a small fraction will be recycled. With a major U_{Repro} stream as waste, instead of product, the separation of Np-237 from HLW cannot be justified on the basis of a radiotoxicity assessment because U-234 and U-236 are present in U_{Repro} .

The HLW solution resulting from LWR- UO_2 fuel reprocessing is the first source term for minor actinide partitioning. Quantitative separation of the Am-Cm fraction before vitrification would result in a substantial decrease of the residual toxicity of vitrified HLW.

By systematically recycling the plutonium, occluded in the spent LWR- UO_2 fuel we achieve a first "concentration" of the actinides' mass with a factor of 5 at least.

After irradiation the total Pu content has furthermore been reduced by 25 to 30 % depending on the burnup but the spent MOX fuel becomes a 3 to 5 times more concentrated actinide source term. If no reprocessing of MOX fuel is envisaged in the near future on an industrial scale it is the first intermediate actinide source term which has to be taken into account in the comparative evaluation of the long term toxicity of different scenario's. If industrial reprocessing of MOX fuel is considered in a medium term the resulting Pu product will become more enriched in Pu-238 and the fuel fabrication technology will have to be adapted due to increased α activity, heat and neutron emission.

3.2. Recycle of MOX fuel in LWR's

Recycle of MOX fuel in LWR's has been investigated for self generated plutonium by WIESE [4]. The fissile Pu content decreases from 63 % in fresh MOX to 48.7 % in spent MOX fuel. This partially depleted plutonium is subsequently mixed with fresh Pu from LWR- UO_2 reprocessing at each recycle. By following this approach multiple recycling of MOX is not excluded if the fissile Pu content is gradually increased to counter effect the steady increase of Pu-242 up to 25 %. The total Pu enrichment must in this hypothesis be increased from 6.32 % in the first recycle to 8.9 % at the 5th recycle with an equivalent fissile Pu content of 3.7 to 4.4 %.

However the minor actinide radioactive inventory resulting from the repeated reprocessing of spent MOX recycle fuel increases very sharply especially Cm-244. From an analysis of the above published data it appears that the Pu consumption increases from 20.8 kg/THM to 26.6 kg/THM at the 5th recycle. However the minor actinides are produced at a rate of 4.8 kg in first generation MOX to 8.9 kg/THM at the 5th recycle. There is a net total actinides consumption of ~ 100 kg in 6 cycles per THM.

By examining the radiotoxicity evolution it appears that the decrease of Pu-238 and the increase of Am-241 are more or less in equilibrium as shown in Table 6, but the generation of Cm-244 is the overwhelming phenomenon.

Table 6
Radiotoxicity evolution of MOX fuel (50 GWd/T) in repeated recycling (Ci/T) after 7 years

	MOX	1st recycle	2nd recycle	3rd recycle	4th recycle	5th recycle
Pu-238 ($\times 10^3$)	+ 0.87	- 5.62	- 8.7	- 9.8	- 11.7	- 12.6
Am-241 ($\times 10^3$)	+ 8.2	+ 9.5	+ 10.7	+ 12	+ 12.7	+ 13.4
Am-243 ($\times 10^3$)	+ 0.25	+ 0.35	+ 0.42	+ 0.51	+ 0.56	+ 0.62
Cm-244 ($\times 10^3$)	+ 51.3	+ 69.4	+ 82.2	+ 95	+ 102	+ 110
Ci α -Balance ($\times 10^3$) per THM	+ 60.7	+ 73.6	+ 85	+ 97	+ 103	+ 112
Heat emission KW/THM due to Cm-244	1.7	2.35	(2.8)	3.23	(3.5)	3.76

3.3. Recycle in fast reactors

As the recycle in LWR-MOX gave rise to a very much increased radiotoxicity and to a disturbing Cm-244 concentration level it was examined what could be obtained by recycling in fast burner reactors. A computation was performed with the ORIGEN-2 programme to assess the effect of increased burnup per cycle and to investigate multiple recycling of LWR-MOX spent fuel in a fast reactor core.

The initial conditions of loaded fuel were as follows:

67 % depleted UO₂

33 % Trans Uranium Elements:

29.4 % Pu		3.6 % Minor Actinides (MA)	
% Pu isotopic composition		% MA composition	
Pu-238	3.08	Np-237	5.09
Pu-239	37.25	Am-241	49
Pu-240	33.62	Am-243	30.8
Pu-241	17.1	Cm-244	15.18
Pu-242	8.95		

Two levels of mean average flux were considered:

3.8 10^{15} n cm⁻² s⁻¹ to reach a burnup of 200 MWd/T

2.44 10^{15} n cm⁻² s⁻¹ to reach a burnup of 129 MWd/T after a reactor cycle operation of 1846 EFPD (~ 5 years).

The single cycle with a burnup of 200 MWd/T resulted in a Pu inventory drop of 26.6 %. The values for the minor actinides varied in burnout: Am 27.5 %, Np 30.5 %, whereas the Cm level increased with 63.4 %.

For the more conservative burnup of 129 MWd/T a multiple recycle scenario with 4 cycles was investigated. Each of the cycles lasted for 1846 EFPD and between each cycle a cooling time of 10 years was taken into account. The results of this calculation are summarized in Table 7.

Table 7
% Depletion of TRU's by multiple recycling in a fast reactor

4 cycles of 1846 EFPD ; Intermediate cooling time: 10 years ;
Burnup per cycle: 129 MWd/T; Flux: $2.44 \cdot 10^{15} \text{ n cm}^{-2} \text{ s}^{-1}$

	Cycle	Cooling	Cycle	Cooling	Cycle	Cooling	Cycle	Cooling
	1	1	2	2	3	3	4	4
Pu	19.5	21.7	14.1	15.3	13.2	14.3	13.2	14.3
Am	11.4	-17	29	16.1	29.8	17.7	29.21	16.5
Np	21	12	22	13.3	23.1	16.3	23	17.15
Cm(*)	-44.3	4.4	-42	3.8	-39	4.1	-34.6	5.7
(*) negative depletion is enrichment.								

By combining the individual depletions per cycle we obtain a global residual inventory of
50 % for Pu
47 % for Am
47 % for Np
83 % for Cm.

Whereas an actinide burnup of 129 MWd/T constitutes an overall depletion of 54.5 % in 4 cycles, there is a good correlation between the overall actinide consumption due to burnup and the individual nuclide depletions except for Cm. It is however striking that the 10 years cooling time between each cycle is particularly effective for Cm depletion ; without cooling periods the inventory would have increased by a factor of 3.75.

The above results have to be analyzed from radiotoxicological point of view and be investigated for each important α emitter i.e. Pu-238, Am-241, Am-243 and Cm-244.

The results of this computation are shown in Table 8 which shows for the most significant nuclides the inventory of α emitters in Ci/THM.

Table 8
Radiotoxicity of recycled MOX fuel in fast reactors after 10 years cooling,
expressed in Ci α /THM (same conditions of Table 7)

Elements	Cycle			
	1	2	3	4
Pu-(238, 239 240)	1.8 10 ⁵	1.59 10 ⁵	1.35 10 ⁵	1.07 10 ⁵
Am-(241, 243)	0.76 10 ⁵	0.59 10 ⁵	0.43 10 ⁵	0.3 10 ⁵
Cm-(244)	3.25 10 ⁵	2.82 10 ⁵	2.54 10 ⁵	1.95 10 ⁵
Totals	5.8 10 ⁵	5.0 10 ⁵	4.3 10 ⁵	3.32 10 ⁵
Relative toxicity versus initial charge	0.84	0.72	0.62	0.48

In the short term toxicity, which is considered here, Np-237, Pu-242 and Cm-245 ... do not contribute in any significant manner. The main conclusion is that the radiotoxicity of the MOX-mixture actinides, is reduced in the same proportion as the global TRU inventory. Cm-244 still plays an important role but does not increase with recycling as it is the case for LWR-MOX recycle (see Table 6). Fast reactor irradiation is an adequate way to reduce the actinide inventory but it takes a long time to achieve a significant reduction. Indeed the reactor irradiation takes 5 years, cooling time 10 years and processing 2 years. Globally each cycle would last 17 years and the actinide reduction obtained is 50 % after 4 cycles (68 years). One way to speed up the incineration pace is by increasing the burnup. The above mentioned fuel inventory would have been reduced to 30 % if a burnup of 200 MWd/t would have been reached in the fuel. However this limit of the attainable burnup depends on the cladding material of the fuel pins and the specific power output of the irradiated fuel. Since multiple recycling involves multiple reprocessing it has to be examined which burnup level is acceptable for aqueous processing.

4. Status of multiple recycle fuel processing

The residual power released by spent fuel is determined by the TRU content, the burnup and the cooling time. Reprocessing of LWR-MOX fuel after 7 years cooling has to cope with an increased residual heat load compared to LWR-UO₂. Standard 33 GWd/T- LWR-UO₂ fuel has an heat output of 1.3 KW/THM, this value increases to 3.27 KW/THM for LWR-MOX fuel of 40 Gwd/T and 4.16 KW/THM at 50 Gwd/T. As mentioned in section 2.2, the impact of this increased heat generation has generally been mitigated by dilution of LWR-MOX fuel with equivalent LWR-UO₂ fuel.

With fast reactor fuel the heat problem is much more important even with a cooling period of 10 years: the increased burnup (to e.g. 180 GWd/T) results in a 7 KW/THM heat generation due to the fission products; the high concentration levels of Pu-238 and Cm-244 produce an additional heat load of 14 KW/THM. Due to this very high decay heat of ~ 21 KW/THM spent fuel, full size fuel assemblies

cannot be chopped in present generation bundle shears. Up to now spent FR-MOX fuel assemblies are disassembled prior to individual pin chopping or multi pin chopping. Some tons of FR-MOX fuel with a burnup up to 180 Gwd/T have nonetheless been reprocessed successfully at Dounreay and Marcoule. The radiation resistance of the conventional extractant TBP is very well known and well understood. The solvent withstands irradiation doses of 30 Wh/l if the contact times are short (pulse columns, centrifugal extractors). The conventional separation U Pu/Fiss. product + actinides can be performed with aqueous processing methods but the solvent will have to be washed more intensively.

Once the raffinate is separated and the so called HAW is sent subsequently to the acid recovery and the waste evaporator the specific heat problem becomes more stringent.

Indeed the volume reduction between HAW and its concentrate HAWC is generally a factor of 10-15 i.e. the ~ 15 KW/THM decay heat is confined within a 350 to 500 l volume which would correspond to 30 W/l or more. All operations on this type of effluent will be difficult unless sufficient dilution is provided before a chemical process is started. It would be preferable to carry out the partitioning processes on the HAW raffinate before its concentration to HAWC, but this approach can only be realized in a, on purpose, designed waste processing and acid recovery system.

If shorter cooling times are to be considered new separation methods will have to be envisaged. Pyrometallurgical methods might be used as first rough separation technique.

5. Critical overview of the actinide partitioning methods [6]

There are four aqueous methods which have up to now been developed for the separation of actinides from high Active Waste Concentrate (HAWC).

The best known method is the american TRUEX process based on the extractant CMPO, the second one is the japanese DIDPA method, the third process is being developed in France under the name DIAMEX and last but not least there is the TRPO method initially developed in China.

5.1. The TRUEX process

CMPO is a very powerful extractant for all III, IV and VI valent actinides from 2 to 4 M HNO₃ solutions. It is compatible with the PUREX process using TBP as extractant and displays the same radiolytic stability.

The following conclusions can be drawn with regard to the TRUEX process;

- CMPO mixed with TBP allows the extraction of the most important actinides, except Np, and the process is directly applicable to HAW solutions and to certain types of HAWC with relatively low salt content.
- The decontamination factors are of the order of $4 \cdot 10^4$.

- CMPO is such a powerful extractant that quantitative elution of extracted actinides, especially U and Pu, is difficult.
- CMPO is an expensive product and it needs additional purification to eliminate some interfering impurities e.g. POPPA.
- Am extracted by CMPO can be recovered quantitatively but it is accompanied by Cm and the Rare Earths. The TRUEX process has to be complemented by an Am-Cm/RE separation (TALSPEAK) and in the end by a Am/Cm separation.
- In order to reduce the production of secondary effluents, the process ought to be modified by introducing salt free reagents (hydrazine, oxalates, hydroxylamine, ...).
- In presence of high salt concentrations third phase formation is observed.
- The TRUEX process does not generate secondary effluents but the subsequent steps (Am/Cm/RE separations) produce a lot.

5.2. DIDPA method

The DIDPA actinide separation method was developed by JAERI as an improvement of the HDEHP method initially worked out by ANL and tested on HAW solutions by JRC Ispra. The DIDPA method makes use of Di-isodecylphosphoric acid instead of HDEHP.

- The DIDPA method needs a preliminary acid destruction step, down to 0.5 M HNO₃, prior to any actinide extraction.
- DIDPA is compatible with TBP and allows the quantitative extraction of all actinides from 0.5 M HNO₃ HAW.
- The decontamination factor is about 10⁴.
- The Am-Cm-RE fraction has further to be separated with HDEHP according to the TALSPEAK process. The resulting Am-Cm fraction still contains 10 % of the accompanying rare earths and further separation is feasible but very tricky.
- The incorporation of the Np separation into the initial process made it rather complex.
- The amount of secondary effluents is of the same order of magnitude as that of the TRUEX method.

5.3. DIAMEX process

The DIAMEX process is a dual process which relies on the one hand on the electrochemical oxydation of Am III into Am IV and on the other hand on the extractive properties of Diamides.

- Initially the mono-amides were investigated as a possible substitution of TBP. The purpose was to reduce the solid residue from solvent incineration.
- The diamides DMDOMA and DMDBMA are good extractants for all actinides from acid solutions (1-3 M HNO₃) but less powerful than CMPO or DIDPA.
- Among the diamides tested DMDBTDMA has the most attractive properties as actinide extractant in acid solution but has to be considered as a compromise between chemical properties and physical characteristics.
- The most attractive part of the present development is the selective separation of oxidized Am(IV) complexed into a phosphotungstate cage by di-n-octylamine extraction.
- The diamides are in general very viscous solvents with relatively slow kinetics and display emulsifying properties in contact with aqueous solutions.

5.4. TRPO process

This process was initially invented in the People's Republic of China and further tested at the Trans Uranium Institute at Karlsruhe.

- The TRPO extraction is similar to the TOPO extraction process which has been widely used in the mining industry during several decades.
- Though relatively new in the field of partitioning this method seems very promising because of the excellent kinetics and the complete extraction reversibility.
- The decontamination factors range from 10³ to 10⁴.
- The actinide loading capacity of TRPO is much larger than that of CMPO.
- TRPO (Tri-n-octylphosphine-oxide) is fully miscible and compatible with TBP and does not form any "third phase" with TBP-Kerosene.
- The AM-Cm-RE fraction can be selectively eluted from loaded TRPO without U-Pu cross contamination.
- Certain fission products (Zr, Mo, Ru, Tc) contaminate the actinide fraction.
- The TRPO is probably the best and most promising of the actinide partitioning methods but must be complemented as the previous methods with Am/Cm/RE separation steps.

5.5. Am/Cm separation

- The largest experience has been accumulated during the production of Am and Cm isotopic heat sources. The methods developed by ORNL and SRL for the separation and purification of Am and Cm by high pressure ion exchange are undoubtedly the most extensive experimental achievement. However these methods produce huge amounts of secondary wastes.

- The most promising approach for Am/Cm/RE separation is the peroxidation to Am IV which can be followed by selective extraction, as proposed in the DIAMEX process.

6. Fuel and target fabrication

- Reprocessing of MOX fuel produces a Pu product which contains increasing amounts of Pu-238. This fact will be of utmost importance in the fuel fabrication facilities.

Table 9 gives the evolution of the Pu-238 in fresh fuel for LWR-MOX 50 Gwd/T throughout the 4 recycles and the equivalent situation for FR-MOX 129 Gwd/T.

Table 9
Evolution of the % Pu-238 in fresh Pu fuel during recycle in LWR and FR's

	R ₁	R ₂	R ₃	R ₄
LWR-MOX 50 Gwd/T 7 y discharge	3.11	3.45	3.71	3.85
FR-MOX 129 Gwd/T 17 y discharge	3.08	3.90	4.68	4.74

The increase of Pu-238 is higher in FR-MOX fuel but the amount of energy extracted from the fuel is 2.5 times higher. The fuel fabrication facilities must be designed to treat in the future fuel types with 4 to 5 % Pu-238.

- The preparation of Am and Cm containing fuel is the greatest challenge in this field since the amounts of Cm-244 determine the total heat load and neutron dose emitted by the spent fuel. Recycling them for actinide destruction will require a fully new technologic approach which has not yet been experienced even on small scale.

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