

## **SESSION 1**

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## WELCOME AND INTRODUCTORY REMARKS

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Good morning Ladies and Gentlemen,

It is a great pleasure for me to welcome you and deliver the opening address on the occasion of the 4th OECD/NEA International Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation.

Nowadays, nuclear power has become a very important energy source and accounts for about 17 % of gross electric power in the world. From the view point of stable energy supply in the future, it is strongly expected that nuclear energy could play a more important role as a reliable energy source on a large scale, once we overcome the basic problems, such as technical and economical difficulties.

Because of plentiful oil production and of low oil prices thereof, people have tended to use oil as if it is an inexhaustible resource. But oil and other fossil fuels are not only for energy but are also as raw materials for various essential products of daily uses. Increase in consumption of fossil fuels is believed to cause global warming, which is one of those global environmental problems to be resolved urgently. Nuclear energy is able to cope with these global problems. The success of nuclear energy use will greatly influence global society in the 21st Century.

For these reasons, Japan intends to guarantee its energy security by carrying forward research and development efforts for commercial implementation of nuclear fuel cycle which includes secure supply of uranium resources, Pu utilization and radioactive waste disposal. For the matured nuclear fuel cycle, the establishment of ways of appropriate management and disposal of radioactive waste, especially high-level radioactive waste, is one of the most important tasks.

Therefore, Japan is implementing significant research and development efforts for realization of consistent high-level radioactive waste disposal measures. In 1995, in order to proceed systematically with high-level radioactive waste management and to promote understanding on the part of Japanese Public for implementing the high-level radioactive waste disposal, the Atomic Energy Commission set up a round table conference for

discussing social and economic aspects of disposal. In the same year, the Advisory Committee on fuel cycle backed management was also set up for reviewing and deliberating on planning and technical issues of research and development of high-level radioactive waste disposal.

At the same time, Partitioning and Transmutation is deemed in Japan to be quite an interesting subject from the perspectives of potential utilization of resources and possible alleviation of environmental burden caused by long-lived radioactive waste. The R&D program is jointly carried out by the collaborative efforts of relevant organizations.

Partitioning and Transmutation is one of the options for processing high-level radioactive waste before final disposal. Significant efforts are needed to implement this concept. I believe this information exchange meeting is very useful for international collaboration in pursuing efficient R&D.

I am also a strong supporter of basic research. This type of work must continue at all times because policy changes can occur in the future and basic research has a long lead time. It is not inconceivable that our study of, for instance, star formation and nucleosynthesis can give us insight into better and novel methods of transmutation which can, in turn, help solve the problems of high-level radioactive wastes.

Therefore, it is a great pleasure and pride for us to host the 4-th OECD/NEA Information Exchange Meeting in Japan.

I would like to conclude the opening address hoping fruitful discussions and also for the promotion of nuclear energy developments in each country of the participants.

**OPENING REMARKS BY MR. G.H. STEVENS**  
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Ladies and Gentlemen,

It is a great privilege and pleasure to welcome you on behalf of the NEA to the 4th International Information Exchange Meeting on Actinide and Fission Product Separation and Transmutation. It is a particular pleasure for me as it was here that the first of these meetings was held in 1990, and I treasure my memories of that occasion.

At a number of recent meetings related to nuclear energy but not specifically to nuclear science or to waste management, I have noticed a tendency to believe that there does exist some magic spell that will somehow relieve us of the difficulties of managing highly radioactive wastes. I, too, wish that there could be such a "spell" but I do not believe in alchemy. I do believe that patient research and ingenuity in development will improve our capabilities to meet the challenges of long-term waste management within a cost-competitive fuel cycle. I see no reason to modify the position that has been taken since the outset of this Programme, that successful application of Partitioning and Transmutation would not replace the need for geological disposal for high-level waste.

Since the NEA was invited to take up this topic in 1988, the interest in it has grown in several of our Member countries. The task is one of long-term scientific research, but it is recognised that certain short- or medium-term benefits could also be derived. There is quite a rich network of bilateral agreements on P&T between OECD countries. However, judging from the number of participants who have come a long way to this meeting there is a clear view that substantial benefits can be achieved from wider international activities and co-operation. I am very glad to see so many friends and colleagues from around the globe.

There are also some newcomers to this forum and I am glad about that too. Mainly for their benefit let me describe briefly the NEA's role and responsibilities in the field we are here to discuss. When the NEA agreed to work on the Japanese proposal for an international activity, it was entrusted to the Nuclear Development Committee. They agreed that it would be sensible to sponsor an information exchange. This led to the first Mito City meeting in 1990. That generated the idea of holding specialist meetings on particular scientific aspects. Subsequently the establishment of the NEA's Nuclear Science Committee made it more practical to hold such meetings under that committee. The Secretariat ensures close co-operation between these committees. The topic is one of great interest to the waste managers and this is recognized in the reporting of the activities we make to the Radioactive Waste Management Committee and in the nomination by it of one of its members, M. Lefèvre, as their special liaison person.

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 some means of taking an integrated view of the expected benefits and possible disadvantages of including P&T in the

nuclear fuel cycle. Among other results of such an approach would be 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.

These views were carried forward at the third NEA Information Exchange meeting, hosted by the CEA at its Cadarache site in December 1994. Several participants from 11 countries, together with Russia, the IAEA and the European Commission attended the meeting which primarily focused on P&T strategic systems studies. The meeting provided a solid base for approaching a more co-ordinated NEA project, which has started in early 1996, on the benefits and penalties of adding P&T to the nuclear fuel cycle.

At that meeting, interesting papers were also presented on national policy orientations and on scientific and related data aspects. There was a wealth of wide ranging ideas and suggestions for future activities. It was noted that real progress since the Argonne meeting had been achieved in three fields:

- chemistry of separation;
- experimental irradiation of actinide-based fuels and targets had been launched in the frame of national or international programmes;
- preliminary cost estimates were presented for a P&T scenario in the frame of the EU strategic systems study.

There is a clear need for objectives against to which to measure potential benefits of P&T. Discussion at the Cadarache meeting indicated that a final set could not yet be established. Therefore, working hypotheses should be adopted for goals which can be adjusted as knowledge improves. This remains our approach.

In addition, the NEA Nuclear Science Committee (NSC) and the NEA Data Bank are co-ordinating a number of technical activities in the field of P&T. The NSC task force on different transmutation concepts has issued specifications for a calculation benchmark comparing activities from various fuel cycles after passage through different reactor and accelerator concepts. The NSC is also preparing a state-of-the-art report on the separation chemistry of actinides. An important part of the technical activities is related to nuclear data questions for transmutation applications. The data activities cover both conventional reactor and intermediate energy applications, and include the compilation of a high priority request list for nuclear data, as well as the compilation of basic experimental data themselves. The Data Bank is also setting up a collection of evaluated intermediate energy nuclear data libraries and is conducting nuclear model code comparison to validate the computational methods used.

I am looking forward to hearing in this meeting of progress in ways of evaluating a number of different P&T systems and their integration into the nuclear fuel cycle. I have no doubt that we shall also have lively discussion on the scientific ideas and results that have been coming out in the last two years. Please forgive me if I have a biased view on these matters, but I firmly believe that exchanges of information and opinion in this sort of meeting is one of the most effective ways to carry forward R&D in this complex and, in some ways, politically sensitive area. I am grateful for the support that the Japanese Government has given the NEA in order that we can continue to be sufficiently active in this field.

I know that organising a meeting like this is hard work, so I particularly wish to thank our Liaison officer Dr. Mukaiyama and his colleagues in the STA, JAERI, PNC and CRIEPI, who have, I am sure, laid the basis for a very successful and stimulating meeting. Thank you all for being here. I wish you all an interesting, instructive and profitable three days.

# RECENT TOPICS IN THE FIELD OF RADIOACTIVE WASTE MANAGEMENT IN JAPAN

Tateo ARIMOTO

## **1. Introduction**

Thank you Mr. Chairman for your kind introduction. Distinguished guests, and Ladies and Gentleman, I'm very honored and glad to have this opportunity to make a speech here today.

On behalf of the Science and Technology Agency, I'd like to present here, recent topics in the field of radioactive waste management.

## **2. Policy Development for the Geological Disposal of High Level Radioactive Waste**

The Atomic Energy Commission of Japan put forward a road map for the geological disposal of the high-level radioactive waste in its "Long-Term Program for Research, Development and Utilization of Nuclear Energy" issued in June 1994. The Program attached particular importance to the program of the management of radioactive waste, especially to ensure the smooth implementation of disposal of high-level radioactive waste. The Program stipulates that the high-level radioactive waste generated through the reprocessing of spent fuels be disposed of in geological formation after vitrification and temporary storage for cooling (for about 30-50 years). The role and responsibility of the related organizations concerned with the geological disposal have been confirmed and the schedule and procedures are shown for the implementation of the disposal.

### **3. Current Activities for Radioactive Waste Management**

Along with the basic philosophy and mile stones set at the Long-Term Program said above, the Atomic Energy Commission set course to promote the discussion and preparation of high-level radioactive waste disposal in September 1995. The Atomic Energy Commission set up "Special Committee on the Disposal of High-Level Radioactive Waste". The Special Committee will make a wide range of study including social and economic aspects in order to pave the way for public understanding and approval of specific measures for actual implementation of the disposal of high-level radioactive waste. "Advisory Committee on Nuclear Fuel Cycle Backend Policy", which was set up by the Atomic Energy Commission last year, is intensively studying and deliberating technological items concerning disposal such as formulation of a research and development plan on geological disposal of high-level radioactive waste.

The special Committee has produced general consensus to promote implementation and research and development for disposal of high-level radioactive waste positively by our own responsibility. On the other hand, the Advisory Committee has started its discussion to produce a research program concerning the high-level radioactive waste disposal forwarding year 2000, when Power Reactor and Nuclear Fuel Development Corporation will publish a report on the results of research and development as said above. The Advisory Committee is now discussing how to evaluate the report. The Advisory Committee is also considering the implementation system as well as research and development for the other radioactive wastes and the decommissioning of nuclear facilities etc. in the future.

Steering Committee on High-Level Radioactive Waste Project (SHP) was established in May 1993. SHP is responsible to prepare measures to encourage the establishment of organization to execute the disposal of the high-level radioactive waste under the public understanding and cooperation. The recent activities of SHP are as follows:

- considering what kind of organization is the most appropriate to conduct the disposal project
- developing mid- to long- term, public relation strategy
- investigating a policy to collect the fund for the disposal project
- investigating the plan for regional developments for site incentive

SHP published the first interim report in May 1996, which includes coexistence with the host communities, define the nature of and funding for the implementing entity including its legal and regulatory aspects, and public information programs and activities for promoting public understanding and acceptance.

#### **4. Underground Research Laboratory**

On 28 December 1995, the Power Reactor and Nuclear Fuel Development Corporation (PNC) signed an agreement with the prefectural and local governments concerned with the construction of an underground research laboratory at Mizunami city in Gifu prefecture, central Japan. It is the first underground laboratory to be able to provide undisturbed conditions of deep underground in Japan, which is of great advantage compared with in-situ test facility using mining galleries. It is expected to conduct a comprehensive geoscientific research, and provide fundamental knowledge of groundwater and rock mass in deep underground to the research and development for geological disposal.

The research projects in the deep underground research laboratory, which will last 20 years, are expected to be executed with the participation of researchers from not only other research organization concerned in Japan but also ones abroad.

#### **5. Partitioning and Transmutation Technology**

The Partitioning and Transmutation technology, which would reduce the environmental impact of the disposal by utilizing useful nuclides in the high-level radioactive waste, is considered to be future technology in the Long-Term Program. The Japan Atomic Energy Research Institute (JAERI), the Power Reactor and Nuclear Fuel Development Corporation (PNC) and other organization such as the Central Research Institute of Electric Power Industry (CRIEPI) are carrying out the basic research and development of these technologies. Research and Development activities of this technology are being continued according to the Long-Term Program for Partitioning and Transmutation, which was published by the Atomic Energy Commission in 1988. The activity which follows this program is called OMEGA project (Options for Making Extra Gains of Actinides and Fission Products generated in Nuclear Fuel Cycle). The check and review based on the progress of these activities will be carried out in the second half of this decade and consideration will be given to how to further proceed with development of this technology.

Now, it's time to close my presentation. Thank you very much for your attention.



## **RESEARCH ACTIVITIES OF THE EUROPEAN COMMISSION ON NEW FUEL CYCLE CONCEPTS**

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### **ABSTRACT**

This paper presents the objective and the research work of the eight projects partly funded by the European Commission in the field of new fuel cycle concepts, which includes three areas of work: (i) strategy studies, (ii) partitioning techniques, (iii) transmutation techniques. This field is part of the specific programme on Nuclear Fission Safety of the Framework Programme for the European Atomic Energy Community (1994 -1998).

## INTRODUCTION

In its fourth five-year shared-cost research and development programme on "Management and Storage of Radioactive Waste 1990-1994", the European Commission (EC) included the study of the potentialities of partitioning and transmutation (P&T) of long-lived radionuclides. The activities partly supported by the EC were dealing with experimental work on partitioning of long-lived radionuclides from high level waste (HLW) and an overall strategy study on the potentialities of P&T for nuclear waste management. Besides, research work on P&T, mainly on partitioning experiments and fuel and target development, was carried out at the Joint Research Centre of the European Commission in Karlsruhe, the Institute for Transuranium Elements (ITU).

The main results and conclusions of the strategy study have been presented elsewhere [1]. This study has led to an order of priorities for the radionuclides to be recycled. Plutonium should have the first priority because of its very large radiotoxicity and of proliferation risks. At present, the main issue is to improve the burning rate of plutonium in reactors to decrease its stock. Americium is the most radiotoxic actinide after plutonium up to 50,000 years and also contributes to the potential radiotoxicity of HLW through neptunium formation beyond 50,000 years. The problem to be solved is to find a selective partitioning process for americium only from the liquid HLW coming from PUREX reprocessing, which does not generate unacceptable amounts of secondary waste. When removing all plutonium and americium from HLW, the resulting waste will become less radiotoxic than uranium ore after 1000 years. Fast reactors appear at present to offer the best prospects for plutonium and americium incineration, when compared to light and heavy water reactors and high temperature reactors.

Nuclear Fission Safety is one of the specific programmes of the Framework Programme for the European Atomic Energy Community (1994-1998). It has been launched by the European Union on 15 December 1994. Exploring new fuel cycle concepts is part of this programme with three research tasks: (i) strategy studies, (ii) partitioning techniques, (iii) transmutation techniques. Eight research proposals to be partly funded by the EC have been selected in 1995 by the Commission on the basis of an evaluation made by independent experts. The objective and the research work of these projects are briefly described in this paper.

## STRATEGY STUDIES

Four projects cover the strategy studies.

### Evaluation of Possible P&T Strategies

The first project is a global assessment of different possible P&T scenarios and of the technical feasibility of P&T techniques and of advanced fuel and target fabrication. Eleven European laboratories are participating to this study, which is coordinated by CEA Cadarache (F). Five scenarios have been defined. The first one is the reference scenario with one-through cycle for pressurised water reactors (PWRs) with uranium oxide ( $UO_2$ ) fuel. The four other scenarios have the same common basis from 2000 to 2040: the reactor park consists of PWRs with  $UO_2$  fuel and PWRs with a mixture of 70% of  $UO_2$  fuel and 30% of mixed oxide (MOX) fuel; plutonium (Pu) is recycled once, i.e. spent  $UO_2$  fuel is reprocessed to recover Pu for MOX fuel fabrication and spent MOX fuel is placed in interim storage. From 2040, spent MOX fuel is also reprocessed following the requirements in Pu, minor actinides are partitioned and recycled in homogeneous mode for neptunium (Np) and heterogeneous mode for americium (Am) and curium (Cm). A PWR loaded with 30% MOX fuel is replaced every year by an over-moderated PWR with 100% MOX fuel in the second scenario and by a CAPRA type fast reactor (FR) incinerating Pu in the third scenario. The fourth scenario starts like the third one until a shutdown of nuclear energy is decided: special actinide burners are then built. The beginning of the fifth scenario is similar to the third one; at a certain time, fast breeder reactors are coupled to the network with the objective of having only FRs as burners and breeders in the reactor park.

The consequences of the use of fuels with high Pu content in PWRs and FRs on partitioning will be assessed: the dissolution of spent MOX fuel in nitric acid becomes more difficult, as the content in  $PuO_2$  increases. Reprocessing of MOX with 45%  $PuO_2$  and uranium free fuel and the effect of inert matrices will be studied. Both enhanced "PUREX based" and/or pyrometallurgical processes will be investigated.

Transmutation in PWRs and FRs will be assessed by computing actinide consumption and core safety parameters for advanced MOX fuels (highly enriched in Pu and/or containing Np) and/or specific targets containing americium and/or curium. Pu consumption will be optimised, while keeping the core safety parameters at a reasonable level. The moderator-to-fuel ratio of full MOX fuel cores in PWRs will be increased to maximise the Pu consumption. "U-free" PWRs and FRs will be studied.

The fabrication processes of the advanced fuel and target will be investigated: sol-gel fabrication and MOX fuels adapted to enhanced moderation and full core loading. The consequences of the presence of more radioactive nuclides in the fuel on the dose rates received by the workers during the different steps of fuel fabrication and handling will be evaluated.

Finally, the long term risk and residual dose to man from different waste to be disposed of in an underground repository will be compared: spent PWR UO<sub>2</sub> fuel at high burn-up, spent PWR MOX fuel, vitrified HLW, actinide depleted vitrified HLW, separated actinides conditioned in advanced matrices, spent FR MOX fuel after multiple recycling. The retardation efficiency of three geological barriers (clay, hard rock, salt) will be investigated.

### **Supporting Nuclear Data for Advanced MOX Fuels**

Belgonucléaire (B) coordinates this project, which involves six European research institutions altogether. The objective of this project is to provide the strategy study above with more accurate nuclear data for the scenarios aiming at reducing the waste toxicity in MOX recycling schemes either in PWRs or FRs.

The accuracy of strategy studies involving the use of PWRs with enhanced moderation and full MOX loading will be assessed. The experimental basis is provided by the analysis of the composition of MOX fuel after irradiation at very high burn-up (80 GWd/t) in the overmoderated BR3 reactor. Experimental data are also available for MOX fuel irradiated in the Saint Laurent B1 PWR. These data will be compared with the results of calculations using the European JEF 2.2 library. Sensitivity and uncertainty analyses will be performed on the basis of the JEF 2.2 library and the EAF transmutation and uncertainty file.

A similar exercise will be performed for FRs. The experimental data are provided by the measurements of the isotopic composition of samples irradiated in Phenix and KNK-II in Karlsruhe. These data will be interpreted with the European JEF 2.2 library.

A critical analysis and intercomparison of the nuclear data files for the isotopes <sup>240</sup>Pu, <sup>242</sup>Pu and <sup>241</sup>Am will be performed especially in the range of resonances.

The corrected nuclear data will be introduced into working libraries for transport and radionuclide inventory computations. The computational techniques of the different partners will be compared to integrate all results in a common data base. Finally, the uncertainty margins of the isotopic compositions will be estimated for the P&T strategy studies.

### **Thorium Cycles as Nuclear Waste Management Option**

The third project is coordinated by ECN Petten (NL) and involves six other European laboratories. The objective is an assessment of the thorium (Th) fuel cycle to limit nuclear waste production and to burn waste. This study will cover the major aspects of the thorium fuel cycle, i.e. mining, fuel fabrication, reactor operation, reprocessing, waste disposal and non proliferation.

The work on mining consists of a study of the quantities of tailings for available thorium minerals, the extraction performances and the composition of radionuclides in the extraction waste. The health impact on workers, the potential radiotoxicity of the generated waste and possibly the residual short and long term risks to the public will be also calculated.

The different types of thorium oxide fuels will be reviewed with special emphasis on the radioactivity of fresh and recycled fuels. Mixed oxide fuels will be considered with uranium, protactinium (Pa) and plutonium.

Three types of thorium fuelled reactors will be assessed: a PWR core with minimum actinide waste production; a PWR to burn as much as possible plutonium and minor actinides (MA) (Np, Am, Cm); a FR as an energy generating system with low actinide production or with high capability to burn Pu and MA. Limits imposed by reactor safety parameters, neutron economy and maximum achievable burn-up will be taken into account.

Concerning reprocessing, the THOREX process will be reviewed on the basis of previous experience with special attention to partitioning of Th, U, Pu and Pa.

The long term residual risk of geological disposal of the different HLW generated by the three types of reactors considered above will be computed. The basic migration parameters of Pa and Th will be measured in order to assess their behaviour in two generic geological environments (clay and granite).

The technical proliferation characteristics of Th and U based fuel cycles will be compared. The basic problems (neutron emission and heating rates) associated with the fissile  $^{233}\text{U}$  and  $^{239}\text{Pu}$  isotopes will be addressed.

### **Impact of the Accelerator-based Technologies on Nuclear Fission Safety (IABAT)**

Kungliga Tekniska Högskolan (S) is coordinating this research contract, which involves a total of eleven European research institutions. The possibilities of Accelerator Driven Systems (ADS) for safe energy production, minimum waste production and nuclear waste transmutation will be assessed in this project. The objectives of the IABAT project are to perform system studies on accelerator driven hybrid systems, to assess accelerator technology, to obtain basic data on nuclear reaction cross-sections and on radiation damages at the spallation target walls and to study the radiotoxicity of the ADS fuel cycles.

The system studies on ADS will address several items: a safety and economical assessment; the physics of molten salt systems with fast and thermal neutron spectrum; the stability, time response and dependence of dynamics on subcriticality; a 2D/3D neutron kinetics study; an actinide incineration system with liquid lead as an actinide carrier; spallation target optimisation with respect to transmutation efficiency; estimation of the spallation product yields and of the radiotoxicity of the spallation target.

The technological aspects of accelerator technology related to construction, proton current and energy levels will be assessed. A cost analysis will be performed for linear and circular accelerators with an energy of e.g. 1 GeV and a current of 10 mA.

As the high energy transport codes are not very reliable in the 100 MeV energy region, basic nuclear data are necessary. Evaluated nuclear data files will be assembled for protons on lead and isotopes for energies up to 200 MeV. Some cross sections above 20 MeV will be measured and validated for neutron and proton induced reactions (e.g. fission of Pb, Bi and  $^{238}\text{U}$  and transmutation of some nuclides). The measurement of yields for  $^{233}\text{U}$  thermal fission and  $^{232}\text{Th}$  fast fission will be performed. The radiation damages at the lead spallation target enclosure walls will be estimated.

Different aspects of possible fuel cycles for ADS will be addressed. The radiotoxicity of the residual waste and the non-proliferation resistance of the reprocessing technology will be investigated for a LWR waste transmutation system based on molten salt fuel and thermal neutron spectrum. Different thorium fuel options will be considered for an ADS with liquid lead coolant/carrier. The radiotoxicity of the thorium based ADS fuel cycle will be evaluated for thermal, epithermal and fast systems.

## **PARTITIONING TECHNIQUES**

Experimental work on partitioning is carried out in two projects.

### **New Partitioning Techniques (NEWPART)**

Seven European laboratories are participating to this project, which is coordinated by CEA Marcoule (F). The objective of this project is to develop processes for the separation of minor actinides from very acidic aqueous solutions containing HLW without generating secondary solid waste, while avoiding the problems encountered previously (e.g. radionuclide precipitation and difficulties in the back-extraction of actinides from the solvent).

To meet this objective, two principles have to be applied: (i) the molecules (extractants, diluents, aqueous soluble reagents) used in the processes must be totally incinerable, once they have been spent; in other words, they are converted into gases, which can be released into the atmosphere; (ii) the very acidic liquid HLW to be processed must not be neutralised.

There are two possible routes for the separation strategy:

- route N° 1 has two extraction cycles, the first one to co-extract the actinides and the lanthanides from the very acidic liquid HLW, and the second one to separate the actinides from the lanthanides in aqueous acid solutions, 0.05 to 0.5 mole/L. Some work will be done on the Am/Cm separation.

- route N° 2 has a single cycle, which extracts selectively the actinides, leaving the fission products and lanthanides in the very acidic aqueous solution (> 2 mole/L). This route corresponds to the second step of route N° 1, except that the separation is done at a higher acidity, which makes it much more difficult from an experimental point of view. Its basic criteria must be carefully established using molecular design, synthesis and testing of new extractants.

It is proposed to use weak bases such as diamide reagents for the first cycle of route N° 1 and to develop the DIAMEX process further. For the second cycle of route N° 1 and for route N° 2, tripyridyltriazine (TPTZ) derivatives and new heterocyclic N donor atom extractants will be investigated.

### **Extraction and Selective Separation of Long Lived Nuclides by Functionalized Macrocycles**

This research project involves CEA Cadarache (F) as coordinator and eight European universities. Its objectives are to synthesise and test new macrocycles for the selective extraction of strontium and actinides from MLW (after a sufficient decontamination, the resulting waste could be disposed of in surface repositories) and for the partitioning of trivalent actinides (Am, Cm) from trivalent lanthanides in HLW.

The work programme of this project has three different items: (i) synthesis of macrocycles; (ii) measurement of the extracting properties of the macrocycles; (iii) molecular modelling and X-ray and NMR structures of the complexes formed by the extractant and the radionuclide.

The synthesis of various extractants such as bis-crown ether and calixarene derivatives will be investigated for the removal of strontium. Calixarenes bearing phosphine oxide and diamide groups will be synthesised for the extraction of actinides. Macrocycles (calixarenes, resorcarenes, and others) with soft and hard donor groups will be studied for the actinide/lanthanide separation.

The extracting properties of these compounds will be thoroughly investigated by determining the complexation constants, measuring the distribution coefficients of the nuclides of interest and carrying out transport experiments through supported liquid membranes.

These experimental results will be compared with molecular mechanics and molecular dynamics simulations to understand the selective complexation of cations and to provide guidance for the synthesis of macrocycles. The measurement of NMR and X-ray structures of the complexes between the macrocyclic extractant and the cation, both as crystal and in solution, will provide additional data for modelling.

## TRANSMUTATION TECHNIQUES

Two projects are dealing with transmutation techniques. The neutrons are produced in a nuclear reactor in the first project and by spallation using an accelerator in the second one.

### Joint EFTTRA Experiment on Am Transmutation

This project aims at demonstrating the feasibility of  $^{241}\text{Am}$  transmutation in a high thermal flux reactor. For this purpose, an irradiation experiment of a target containing  $^{241}\text{Am}$  embedded in an inert matrix (e.g. cerium oxide or spinel) will be performed in the High Flux Reactor (HFR) at Petten.

The project includes different steps: fabrication of the target (stainless steel cladding encapsulating pellets made of a mixture of americium oxide and inert matrix); preparation of the irradiation facility; irradiation of the target during one-and-half year; non-destructive analyses (i.e. visual inspection,  $\gamma$ -ray spectrometry and tomography, neutron radiography); destructive post-irradiation examination (after de-cladding, the sample will be prepared for microscopic examination and a part of it sent to chemical analysis to determine its isotopic composition and hence the transmutation rate); interpretation of the results.

The work will be carried out by the EFTTRA (Experimental Feasibility of Targets for TRANsmutation) group, which consists of six European laboratories and institutes. The coordinator is ECN Petten (NL).

### Neutron Driven Transmutation by Adiabatic Resonance Crossing (TARC)

This project is coordinated by CERN and involves five other European research institutions. Its main objective is to develop both theoretically and experimentally a new method, the adiabatic resonance crossing (ARC), which enables to enhance strongly the capture rate of neutrons by the radionuclides to be incinerated. An experimental test will be carried out with a neutron spallation source driven by the CERN proton synchrotron. The transmutation of the radionuclides exposed to the neutron flux will be detected by the de-excitation  $\gamma$ -rays and the decay of the daughter nuclei with electronic methods. A first test will be made with  $^{99}\text{Tc}$ , for which the predicted cross sections are sufficiently accurate.

The peak cross sections for neutron energies corresponding to the resonance region are much larger than for the other neutron energies. For instance, the cross section of  $^{99}\text{Tc}$  is 4000 barn at the peak of a resonance at 5eV, but it is only 20 barn at thermal energy. Access to the resonance region can be achieved by using a transparent medium where the neutrons lose their energy in very small decrements following successive nuclear encounters (i.e. the lethargy is minimal) and their absorption is very small. In such a medium, the neutron flux per unit energy is correspondingly larger and the neutron energy decreases slowly through the resonances, leading to a very large probability of capture during crossing. Lead appears to be one of the best elements which satisfy these criteria.

All the measurements will be performed in a large lead volume with a spallation target (initially the lead itself) and several types of detectors. The radionuclides to be transmuted will be introduced in the lead assembly through thin channels. A narrow hole brings the proton beam from the CERN proton synchrotron deep inside the structure. Protons create neutrons after interaction with the lead.

The first task is to construct the lead assembly and to equip the proton beam line with instruments. Then, different types of experiments on ARC will be carried out. The transport properties of lead for neutrons at different energies will be studied. An appropriate formalism and new time-efficient Monte Carlo simulation methods for parallel computers will be developed for ARC. Finally, a conceptual design of an incinerating device based on ARC will be made.

## **CONCLUSION**

The three areas of the research activities on new fuel cycle concepts are covered with more emphasis on strategy studies and balance between the two experimental areas. These research activities gather different scientific fields from basic chemistry (partitioning) to nuclear physics and accelerator technology. About forty different European research institutions are participating to the projects, which have started in 1996. It is expected that, at the end of the specific programme on Nuclear Fission Safety in three years from now, the scientific community will have a clearer picture on the possibilities offered by the different options investigated to reduce efficiently the radiotoxicity of nuclear waste.

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## **REFERENCE**

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## 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 CRP entitled "Safety, environmental and non-proliferation aspects of partitioning and transmutation of actinides and fission products" (1995-1998). The documents on accelerator-driven systems and on the status of transmutation studies in non-OECD countries are being prepared.



## 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.

5. **Status Report on Transmutation of Actinides in non-OECD Countries** (to be published in 1996)

The preparation of a Status Report on Actinide Transmutation in Advanced Nuclear Reactors in non-OECD countries was initiated in 1994. Its objective is to review the status of research activities in non-OECD countries and to discuss relevant programmes on transmutation development in these countries.

Participating Countries: Belgium, China, Czech Republic, France, India, Japan, Republic of Korea, Russian Federation.

6. **Status Report on Accelerator Driven Systems** (to be published in 1997)

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

Participating Countries and Organizations: France, Japan, Russian Federation, Sweden, USA, EC-JRC (Ispra Site), CERN.

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

This CRP was established in 1995. It will examine the different fuel cycle options in which Pu can be recycled with Th to eliminate of Pu, or replace Pu with materials that are more acceptable to the public. Potential of Th-matrix will be examined through computer calculations. Each participant can choose his own cycle, and the different cycles can be compared through certain predefined parameters (e.g., annual reduction in Pu inventory). The toxicity accumulation and a transmutation potential of Th-based cycles for current, advanced and innovative nuclear power reactors, including hybrid systems, will be investigated.

The CRP consists of two parts (Benchmarks):

**Part 1: Calculation of the isotopic composition, cross- sections and fluxes for a typical PWR-cell loaded with (Pu-Th)O<sub>2</sub> - fuel, as a function of the fuel burnup.**

8 countries are participating in the Benchmark.

Data set was distributed by IAEA in 1995 and all participants have already sent us the results of calculations. As all results of calculations are available, RCM is planned to be held in October 1996 in Vienna to discuss the results of calculations and to approve the next stage of the programme.

**Part 2: Accelerator Driven Systems (ADS) - Neutronic Calculations**

Goal of Stage 1 of this Benchmark was defined as a verification of reactivity swing during burnup, as well as some important reactivity effects for a fast spectrum ADS ( <sup>233</sup>U - <sup>232</sup>Th fuel) with an external (spallation type) neutron source at different subcriticality levels. 9 countries and 1 International Organization (CERN) are participating in this Benchmark. The final data specification was distributed in July 1996 and RCM is planned for February-March 1997.

## EFTTRA IRRADIATION EXPERIMENTS FOR THE DEVELOPMENT OF FUELS AND TARGETS FOR TRANSMUTATION

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### ABSTRACT

The EFTTRA collaboration (Experimental Feasibility of Targets for Transmutation) between CEA (France), ECN (The Netherlands), EDF (France), FZK (Germany), IAM and ITU (European Commission) was launched in 1992, with the aim of performing joint experiments for the study of materials for the transmutation. Irradiations have started in parallel in the Phénix fast reactor in France, and in the high flux thermal reactor HFR in the Netherlands. One of these experiments, concerning technetium and iodine, has been completed ; post-irradiation examinations of the Tc metallic samples are performed by ECN, CEA and ITU, and a summary of the last results is presented. The other on-going EFTTRA experiments are described, with a report on the application of fabrication methods for matrices with up to 10% americium content. Finally, some considerations on the strategies for americium are given.

## INTRODUCTION

In the field of the research on nuclear wastes management, the problem of the long-lived radioactive nuclides requires careful consideration. The possibility of separating and transmuting these long-lived radioactive nuclides, with the aim of reducing the radiotoxicity of the final waste, has to be investigated. To carry out such experimental or theoretical investigations in the frame of an international collaboration presents many advantages, one of them being to split the costs of the studies. Therefore, the decision to form the EFTTRA (Experimental Feasibility of Targets for Transmutation) group, devoted to technical problems, together with European partners, was taken [1].

## EFTTRA PROGRAMME AND RESULTS

The EFTTRA programme presently consists of the study of the transmutation of Tc-99 (metal), and of the development of materials (inert matrices) for the transmutation of americium. One experiment on Tc (EFTTRA-T1) concerned also iodine.

### Technetium

Metallic Tc rods have been produced and welded in 6 cylindrical target capsules (3 for HFR and 3 for Phénix), at ITU, for irradiation in HFR and in Phénix. The irradiation of 3 target capsules in HFR, an experiment called EFTTRA-T1, was completed in 1994, and "the first results of the post-irradiation examination at ECN, which have been presented earlier [2-4], show that the burn-up is about 6.4 % and that no swelling of the rods has occurred. Part of these irradiated samples have been sent to ITU and CEA for comparative examination ; the results from the three laboratories will be discussed and interpreted in common, and a detailed report will be published. The observations at ITU and CEA confirm the results obtained earlier at ECN that the irradiation had no significant effect on the structure of the material, and no swelling was detected. It will be checked if this good behaviour of technetium metal remains for higher burnup (see below, EFTTRA-T2). The distribution of the Ru concentration along the radius of the sample, determined at ITU by EMPA (Electron MicroProbe Analysis), showed the same sharp decrease as for the ECN sample, from ~ 13 % at the surface, down to 8 % at 0.1 mm depth and 6 % at 1mm depth (diameter of the samples : 4,8 mm). The value obtained at the surface of the sample is slightly below the ECN value : this can be due to the very steep slope of the concentration curve near the surface, or to the orientation of the sample. No significant variation of the Ru concentration was found along the axis of the samples. The PIE observations are in excellent agreement with the results of detailed KENO Monte-Carlo calculations, as performed at ECN [5].

Two technetium rods have been re-packed at ECN for a re-irradiation in the HFR. This irradiation (EFTTRA-T2) was started in February 1996 and will last for about 500 full power days during which a burn-up of more than 20% will be achieved.

The irradiation of the 3 other Tc target capsules is planned in the fast reactor Phénix (EFTTRA-F2) ; depending on the schedule of the Phénix reactor, the irradiation should start in 1997. The samples will be placed in the radial blanket of the reactor, in a thermalized neutron flux ; the moderator is CaH<sub>2</sub>. The experiment aims at a transmutation of 15%.

### Iodine

The iodine capsules (containing natural I-127) of the EFTTRA-T1 irradiation were examined at ECN [4,6]. The capsules containing CeI<sub>3</sub> and NaI did not show any degradation and the burn-up was 5-6 %, the capsules containing PbI<sub>2</sub> were heavily corroded and leakage had occurred. Taking also into account the information from the fabrication of the targets, NaI seems to be the best of the three candidate materials. However, because the necessity of iodine transmutation is still under discussion and the implementation of the transmutation of iodine in the fuel cycle is very complicated (large inventories, long transmutation half lives, vented pins concepts to release Xe pressure), the EFTTRA cooperation will stop its efforts on this element.

### Inert matrices

The heterogeneous recycling of Am supposes that targets for the transmutation, with a high percentage of americium (10 to 40%), are produced, irradiated, and reprocessed. The choice of a suited inert matrix depends on its thermodynamic, physico-chemical, and mechanical properties, and on its behaviour under irradiation or during cooling.

The preliminary studies are done in the laboratory, for a first selection of matrices [7,8]. Techniques have to be developed for the fabrication of the materials ; if reprocessing by the Purex process is envisaged, the solubility of the matrix in nitric acid, possibly after crushing of the material, is an important factor : MgO was found to be the most easily soluble. Once the matrix has been fabricated, a range of parameters has to be

measured : the ideal matrix material should have high thermal conductivity and melting point temperature, low creep and swelling properties, low activation coefficient, low neutron absorption, and should not react with the cladding or the coolant. Radiation damage can also be studied out-of-pile, by ion implantation. Table 1 shows the main criteria and the present situation on the evaluation of candidate (oxide) matrices. A similar study is now underway for nitride matrices.

For the study of the behaviour under irradiation of the candidate matrices, several experiments are planned.  $UO_2$  may be added to the samples, to simulate the presence of americium oxide. The EFTTRA-T2 experiment, already mentioned in the above section on Tc, also includes the irradiation in HFR of samples of  $Al_2O_3$ , YAG ( $Y_3Al_5O_{12}$ ) and spinel ( $MgAl_2O_4$ ). A shorter, parallel irradiation, with also a sample of  $CeO_2$ , started at the same time in HFR (EFTTRA-T2bis), and should give a quicker indication of the behaviour of the matrices in a high thermal neutron flux environment.

The start of the EFTTRA-T3 experiment, in which a selection of inert matrices is mixed with  $UO_2$ , 20% enriched in U-235, is planned for the end of November 1996. Table 2 gives a list of the samples which have been selected for this irradiation.

Details on the EFTTRA-F1 and EFTTRA-F1bis experiments, also called Matina 1 and Matina 1bis, in the Phénix fast reactor have been given earlier [8-10] ; changes in the programme may occur, depending on the operation planning of the Phénix reactor, and of possible new orientations in the strategy.

#### Americium targets

The same preliminary studies on fabrication, properties, and reprocessing, have to be done with the matrix material containing americium. This was started with the AmO - MgO system [11].

The irradiation in HFR of a sample of americium oxide embedded in a spinel matrix, the EFTTRA-T4 experiment, is now under preparation. This experiment is partly financed by the European Commission, through the shared cost actions of the Framework Programme 1994-1998 on Nuclear Fission Safety. The samples have been produced by ITU, using an impregnation method developed in the frame of a research programme of the Institute on innovative fabrication techniques for the fuels of tomorrow ; a publication is under preparation, which will give detailed information on the technique. Dose measurements have been performed on the samples, giving indications on the protective measures to be considered for the future developments of the technique ; it can already be stated that the impregnation technique elaborated at ITU presents two main advantages in this respect, compared to the classical powder mixing method : the low amount of wastes produced, and the possibility to remotely control the process, i.e. to produce the samples with a good radiological protection of the operator.

The samples, containing 10 wt% of Am-241, will be encapsulated in a stainless steel cladding. The irradiation is planned to start in September 1996 ; the duration will be about 400 full power days, with a fluence of the order of  $4 \times 10^{26} \text{ m}^{-2}$ . According to the calculations [12,13], the actinide density in the sample could be reduced by 35 %.

### UNDERLYING STRATEGIES

The efforts of the EFTTRA group are concentrated on the technological aspects of the transmutation, namely the development of targets for the transmutation of technetium and americium. However, the connection of our programme with the studies on strategies cannot be denied : it has to be consistent with the possible options for the future, and the final choices will be made taking also into account our results on the technical practicability of the proposed solutions. As such, EFTTRA is participating in the shared cost action of the European Commission on the Possible P&T Strategies, where it will contribute to the work package on the assessment of the feasibility of advanced fuel or target fabrication.

In most of the countries using nuclear energy, no strategy has been chosen yet for the long term, concerning the back-end of the fuel cycle, i.e. the fate of the plutonium and of the minor actinides. Different scenarios have been defined, which could be classified according to the recycling scheme, considering the available reactor park : the CEA studied 4 scenarios, PWRs only (recycling of Pu and minor actinides in MOX fuel), PWRs and recycling in fast reactors, PWRs with Pu monorecycling and subsequent recycling in fast reactors, or fast reactors only. Sometimes, separate dedicated devices for the transmutation are proposed, like for the Japanese "double-strata" system [14], based on fast reactors or hybrid (accelerator) systems. The scenarios include also the possible evolution of the nuclear energy in the coming years : decrease, stability, or increase (possibly with the development of new reactor technology). The final aim of the study of the various scenarios is an appreciation of their impact on the reduction of the radiotoxicity of the end waste disposal. For each scenario, many elements have to be investigated, like the consequences on core operation, on safety, on cycle operation ; the reactor parameters have to be optimised, and the materials tested. There are also other items entering in the appreciation of the value of a scenario :

- should it be a strategy for today, with the present technology, or a solution for the future ?
- weight of the economic factor



- capacity of reducing the losses from reprocessing
- is there a request for reprocessing, or is once-through permitted?
- technological possibility to go to high burn-up
- can the reprocessing technique be different from PUREX ?

Concerning the field of interest of the EFTTRA group, i.e. the transmutation of americium, Table 3 shows the spectrum of envisaged possibilities. The advantage of an inert matrix is the reduction of the production of plutonium isotopes ; but a UO<sub>2</sub> matrix is easier to reprocess, has the best behaviour under irradiation, and it has been shown that the extra radiotoxicity produced is negligible. If the once-through solution is adopted, there will be no reprocessing, but very long irradiation times will be necessary, with the problem of finding suitable materials.

## CONCLUSION

Before a definitive choice of the right strategy for the future can be made, the study of the different scenarios must be pursued, in order to have enough elements of decision available. EFTTRA is participating in this effort, with a range of complementary irradiation experiments, both in a thermal and in a fast neutron spectrum, including the fabrication tests, the determination of the properties of the candidate materials, and the post-irradiation examination programme.

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Table 1 : Evaluation of candidate inert matrices for the transmutation of minor actinides

### Candidate oxide matrices for M.A. transmutation in a PWR

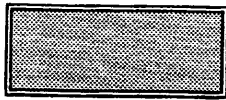
Technical steps	MgO	MgAl <sub>2</sub> O <sub>4</sub>	Al <sub>2</sub> O <sub>3</sub>	CeO <sub>2</sub>	Y <sub>2</sub> O <sub>3</sub>	Y <sub>3</sub> Al <sub>5</sub> O <sub>12</sub>	CePO <sub>4</sub>	ZrSiO <sub>4</sub>
Manufacturing	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes
Basic properties	Yes	Yes	Yes	Yes	Yes	Yes ?	Yes	No
Neutron activation	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes
H <sub>2</sub> O compatibility	No	Yes	Yes	Yes	No	Yes	?	?
In pile behaviour	Yes	Yes ?	No	?	?	Yes ?	?	?
<b>PWR compatibility</b>	<b>No</b>	<b>Yes ?</b>	<b>No</b>	<b>Yes ?</b>	<b>No</b>	<b>Yes ?</b>	<b>?</b>	<b>No</b>

### Candidate oxide matrices for M.A. transmutation in a Fast Reactor

Technical steps	MgO	MgAl <sub>2</sub> O <sub>4</sub>	Al <sub>2</sub> O <sub>3</sub>	CeO <sub>2</sub>	Y <sub>2</sub> O <sub>3</sub>	Y <sub>3</sub> Al <sub>5</sub> O <sub>12</sub>	CePO <sub>4</sub>	ZrSiO <sub>4</sub>
Manufacturing	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes
Basic properties	Yes	Yes	Yes	Yes	Yes	Yes ?	Yes	No
Neutron activation	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes
Na compatibility	Yes	Yes	No	No	Yes	Yes	No	No
In pile behaviour	Yes	Yes ?	No	?	?	Yes ?	?	?
<b>FR compatibility</b>	<b>Yes</b>	<b>Yes ?</b>	<b>No</b>	<b>No</b>	<b>Yes ?</b>	<b>Yes ?</b>	<b>No ?</b>	<b>No</b>

Table 2 : EFTTRA- T3, irradiation of candidate inert matrices in HFR Petten

level 1	CeO <sub>2</sub> + UO <sub>2</sub>	CeO <sub>2</sub>	Y <sub>2</sub> O <sub>3</sub> + UO <sub>2</sub>	Y <sub>2</sub> O <sub>3</sub>
	CePO <sub>4</sub> + UO <sub>2</sub>	CePO <sub>4</sub>	Y <sub>2</sub> Al <sub>5</sub> O <sub>12</sub> + UO <sub>2</sub>	Y <sub>2</sub> Al <sub>5</sub> O <sub>12</sub>
level 2	MgAl <sub>2</sub> O <sub>4</sub> + UO <sub>2</sub> (sol-gel)	MgAl <sub>2</sub> O <sub>4</sub> (sol-gel)	MgAl <sub>2</sub> O <sub>4</sub> + UO <sub>2</sub>	MgAl <sub>2</sub> O <sub>4</sub>
	YN + UN	YN	ZrN + UN	ZrN



also for CAPRA-programme

Table 3 : Possible strategies for the transmutation of americium

	thermal spectrum		fast spectrum	
	once-through	reprocessing	once-through	reprocessing
inert / heterogeneous	few % Am	> 20% Am	X	> 40% Am
UO <sub>2</sub> / heterogeneous	X	> 20% Am	X	> 40% Am
MOX / homogeneous	X	few % Am	X	few % Am

# A COMPARATIVE SYSTEMS-ANALYSIS APPROACH ON FUEL CYCLES WITH PARTITIONING AND TRANSMUTATION

by L.H. Baetslé

## Abstract

A system's analysis approach aims at the definition of the optimum strategy in future P&T research and development activities. Since all nuclear electricity production leads to the production of waste, a general discussion must lead to an agreement on the risk analysis assessment. Two concepts are in opposition: the intrinsic radiotoxicity versus the site specific risk analysis.

The objectives of the P&T systems analysis are:

- provide a synthetic and comparative overview of current projects
- investigate the feasibility of the proposed approaches
- bridge the gap between waste management issues and P&T options
- select promising developments for further international cooperation.

The fuel cycle with P&T should be compared with the currently practised fuel cycles: the once-through fuel cycle with direct disposal and the conventional fuel cycle with U+Pu recovery and MOX use in LWRs and disposal of vitrified waste.

In the once-through cycle the criticality issue and the long term stability of spent fuel are the most important radiological safety aspects to be assessed.

The conventional fuel cycle reduces Uranium mining needs and the resulting radiologic impact of the mill tailings. The short term increase of dose to man in LWR-MOX fabrication and the longterm impact of depleted and reprocessed uranium should also be discussed.

The separation and recycling of Minor Actinides from HLLW will reduce the intrinsic residual radiotoxicity of vitrified waste, but may not influence the risk analysis.

Separation of actinides and a recycling strategy to stabilize the total actinide inventory is probably the most ambitious goal of P&T in the present circumstances.

The longlived fission products can only partially be eliminated either by dilution e.g. I129, or by transmutation (Tc99), in dedicated very specialised facilities and at very high cost.

The advanced fuel cycle with a complete set of waste recycling steps should be analysed from comparative cost benefit point of view. Fully new technologies e.g. actinide burner reactors and accelerator-driven-transmutation facilities can only be assessed in a generic way since their development is still in their infancy. A very extensive set of parameters might influence the outcome and the credibility of the comparative analysis.

Adaptation of existing technologies will be more easily approached: a.o. reprocessing of high burnup fuel from LWR-MOX and FR-MOX, fabrication of concentrated actinide targets and fuels especially those containing Pu238, Am241, Cm244., processing losses etc but are fundamental issues in the judgment on P&T.

The realization of a comprehensive systems analysis study will require contribution from all involved countries and require the participation of a multidisciplinary task force.

## 1. DEFINITION OF PARTITIONING AND TRANSMUTATION

**Partitioning** is a complex series of Chemical and /or Metallurgical operations, intended to separate selected radiotoxic nuclides or groups of nuclides from the bulk of radionuclides occluded in the spent fuel elements which are periodically discharged from a nuclear reactor. The separated nuclides or group of nuclides can be stored as such or transformed into new fuel elements or irradiation targets.

**Transmutation** is the general term covering as well elementary nuclear transmutation as a result of single neutron capture or more complex phenomena as fission of heavy nuclei, spallation and other nuclear reactions involving neutrons produced in nuclear reactor cores or in subcritical nuclear facilities connected to high energy proton accelerators. The ultimate aim of the transmutation is to reduce the radiotoxicity by transforming the initial nuclide either into short lived radionuclides or into stable isotopes.

## 2. ROLE OF P&T IN THE FUEL CYCLE SCENARIOS.

### 2.1 The Once Through Cycle (OTC)

The OTC is the scenario by which all spent fuel discharged from a nuclear reactor is stored in engineered facilities till sufficiently cooled to be transferred to a permanent disposal site in deep geological formations.

The OTC scenario implies that with continuing nuclear power throughout the world the quantity of Uranium mining remains at present day levels, and that the residual fissile material content of the spent fuel (1% Pu 0.8%U235) is lost for ever.

The main longterm potential radiotoxicity is associated with the actinide elements particularly the TRU nuclides (Pu, Np, Am, Cm...) which constitute over periods of hundred thousands of years the radiological source term within the geosphere. However the intrinsic insolubility of actinides in deep geological formations reduces the effective contribution of the TRUs to the longterm dose to man.

The fission products (FP) are in the short term the most limiting factor in designing the repository facilities due to the decay heat emission which increases proportionally with the burnup. After some 300 to 500 years the major part of the FP have decayed except for some longlived nuclides (Cs135, Tc99, I129, Zr93...) with half lives of hundred thousands to million years. Some of them are relatively mobile in the geosphere and contribute to the dose to man in the vicinity of repository structures or may even enter a worldwide circulation.

The radiologic impact of the OTC can only be controlled by man made barriers which are as longlived as the radiological source term they confine. The very long time periods involved require a full public acceptance of such scenario which goes far beyond the reference lifetime of a geologic repository i.e. 10,000 years.

At the present time there is no worldwide agreement on the time intervals for confinement of high level radioactive wastes in a geologic repository. Periods of 100000 years and longer have in the past been considered as a necessary or a wishful target but no internationally accepted confinement periods have been established.

**The working group might investigate the consequences of the duration of the confinement period in a geologic repository on the requirements for a P&T approach.**

A number of specific regulatory and safety aspects are associated with the OTC scenario:

-the maximum inventory of spent fuel is (in the US) limited to 70000 THM per repository.  
-there is an inherent criticality potential associated with the quantity of fissile material in the repository

-the longterm heat emission on the surrounding geosphere due to the TRUs.

Indirectly the OTC scenario implies the continuous production of mill tailings which accumulate at the mining sites.

By not recycling fissile or fertile materials the energetic significance of the nuclear energy in the global world energy economy is rather limited. **P&T is incompatible with OTC.**

## **2.2 Conventional Fuel Cycle with Reprocessing and U- Pu recycling. (CFC)**

Since natural Uranium contains only 0.72% of fissile U235 isotope, the recycling of U -Pu from spent fuel through the CFC has been from the beginning of the nuclear era the standard scenario of nuclear energy production. Since 1972 this policy has been rejected by the US administration for economic and non-proliferation reasons.

The CFC scenario incorporates the "reprocessing" step which aims at the recovery of U and Pu from dissolved fuel elements and the transfer of FP and Minor Actinides (M.A.=Np,Am,Cm) to the High Level Liquid Waste(HLLW) which is stored for a number of years till vitrification of this highly active solution. The glass-blocks are stored in engineered facilities till their transfer as HLW to a geological repository.

By proceeding according to this CFC scenario the major fraction (99.7%) of the U and Pu streams are extracted and only a very minor fraction of the so called "major actinides" are transferred to the HLLW (and consequently to the HLW) and eventually to the geologic repository.

Partitioning of Minor Actinides (M.A.) from HLLW in order to further reduce the radiological potential of HLW has been studied since the 1970ies. Initially the R&D activities were focussed on the quantitative removal of MA in order to eliminate the need for any longterm storage or final disposal in geologic formations. This option was abandoned, because unrealizable. However if the public and/or political acceptance of very long term disposal of HLW could not be obtained, the removal of MA from HLLW is a technical solution which might reduce the residual radiotoxicity of the HLW. With increasing burnup the generation of MA becomes more and more important.

**The addition of a MA Partitioning module to the standard reprocessing plant would in such a case be the most obvious step to be added to the current CFC.**

Countries with a reprocessing infrastructure (France,UK,Japan,Russia,China) and their associated partners could in a medium term realize a partial partitioning scenario by which the actually produced HLW would be practically free from longlived TRUs.

However the question arises what to do with the recovered U,Pu, and TRU fractions ?

The countries which chose to reprocess their spent fuel did this with the main purpose to recover the major actinides (U and Pu), to save on fresh uranium purchase (20%) and to use

the residual fissile components of the spent fuel (1% U235, 1% Pu) corresponding to about 25 % of the regular SWU expenses in the uranium enrichment step.

For a number of decades the Pu recycling was envisaged in a Fast Reactor (FR) option but for technical, economic and political reasons this longterm option of nuclear energy production has been slowed down and sometimes even put to an end.

The stock of Pu already accumulated at the reprocessing plants and which was intended to be used in LMFBRs became redundant in a cheap uranium market economy.

Some years ago the recycling in LWRs of Pu recovered in the reprocessing operations became an industrial practice. The use of LWR-MOX in a CFC option got industrial significance in Western-Europe where increasing quantities of PuO<sub>2</sub> were transformed into LWR-MOX fuel and irradiated in specially licensed reactors in France, Germany, Switzerland and Belgium.

**The reuse of Pu is to a certain extent a first step in a global P&T scenario which has to be brought into broader perspective of reuse of resources and reduction of the radiotoxic legacy from the nuclear age.** The recycling of Pu as LWR-MOX is a posteriori a step to recover the expenses spent during the conventional reprocessing step by producing a fraction of the electricity in a subsequent reactor core. But from radiotoxic point of view the overall gain is rather limited since only ~ 25% of the recycled Pu is fissioned and about 10% is transformed into a longterm radiotoxic MA source term.

Spent LWR-MOX is for the time being the most important radiotoxic source term which has been reduced in overall mass (a factor of about 5) but not significantly in total radiotoxicity. Multiple recycling of LWR-MOX is theoretically possible in LWRs but the resulting radiotoxicity increases throughout the subsequent recycling campaigns.

In order to reduce the global radiotoxicity it is necessary to transfer the TRU content of spent LWR-MOX fuel into a FR-MOX fuel cycle scenario which becomes a part of the next-generation scenario: the AFC.

In the meantime retrievable storage of spent LWR-MOX fuel is the most appropriate fuel cycle option till a fast Burner reactor technology will become industrially available.

Reprocessing of spent LWR-MOX fuel in view of recycling the TRUs in a FR energy scenario is conceptually possible in present reprocessing plants but these are equipped with extraction facilities for U+Pu recovery and transfer the MA fraction to the HLLW. Since the MA generation in spent LWR-MOX fuel is much higher than that in LWR-UO<sub>2</sub> fuel the radiotoxicity of HLLW resulting from LWR-MOX reprocessing is much more important.

**The potential radiotoxicity of HLLW produced during reprocessing of LWR-MOX fuel would require an additional TRU separation module to significantly reduce the residual radiotoxicity of HLW in comparison with spent LWR-MOX fuel. The same technology as that proposed for HLLW from LWR-UO<sub>2</sub> would be adequate.**

The recovery of U from spent fuel was from the beginning of the nuclear era an obvious option which was followed because the extraction in the PUREX process was carried out with TBP. For many decades U has been recovered during reprocessing but very little of this stockpile has been reused in subsequent reactor loadings.

Reprocessed U contains some disturbing radioisotopes (U234 and U236) which are the mother isotopes of natural decay chains with radiotoxic decay nuclides. Any delay in recycling reprocessed U leads to the buildup of the (4n+2). Radium chain from U234 with outspoken residual radiotoxicity due to Ra226. The buildup of U236 in reprocessed U, has economic consequences, since it is a neutron poison which decreases the reactivity of reprocessed U versus natural U. For all these reasons and particularly because fresh mined U became so cheap, reprocessed U has not been recycled on an industrial scale and might become a medium level waste type which cannot be disposed of in surface repositories.

If there is no medium term solution to this potential U waste stream, the residual radiotoxicity of reprocessed U will surpass that of Np237, one of the MA.

Recovery of Np237 from the U- Pu product stream which is technically possible in the PUREX process, might this way become unnecessary from radiotoxic point of view. This problem ought to be investigated from many viewpoints to end up in a recommendation for the elaboration of the AFC scenario. Indeed one of the TRU nuclides with the longest half life (2,140,000 years) is Np and very few natural barriers are capable to confine this radiotoxic element over such an extended time period.

The quantitative removal of Am241,243 from HLLW resulting from the reprocessing of LWR-MOX is without any doubt the most important task to be investigated, since these nuclides determine to a major extent the potential radiotoxicity of this type of HLW within the time interval of 500 to 4000 years. Part of the Np 237 problem is solved when Am241, its mother isotope, has been quantitatively separated from HLLW before vitrification.

**As a conclusion we might say that the recovery of TRUs from spent LWR-MOX fuel would be a first significant step in implementing the P&T option for LWR fuel. The costs incurred in this option might become prohibitive unless the dose-benefit/cost-increment ratio remains acceptable in comparison with the Pu extraction and recycling.**

### **2.3. Advanced Fuel Cycle with TRU recycling. (AFC)**

P&T has to play an essential role in the future AFC which is intended to reduce as much as possible the residual radiotoxicity of the Waste streams. The ideal AFC scenario with P&T will be treated in this note as a long term objective to be reached, knowing that economic constraints will damp the enthusiasm for this futuristic option.

**Any AFC scenario must rely on the use of reprocessing of spent fuel as an indispensable first step.**

A comprehensive AFC scenario with **Partitioning and Transmutation** comprises the following steps:

- Conventional reprocessing of LWR-UO<sub>2</sub> fuel.
- Separation of MA from HLLW resulting from LWR-UO<sub>2</sub> reprocessing.
- Fabrication of MA targets for heterogeneous irradiation in LWRs.
- Separation of certain fission products with long half lives if required for the disposal step.
- Quantitative recycling of U and Pu into LWR-MOX fuel. (single or multiple recycling)
- Reprocessing of spent LWR-MOX fuel in adequate facilities (higher Pu inventory).
- Separation of MA from HLLW and conditioning of individual elements. (Np, Am, Cm)
- Fabrication of FR-MOX fuel with limited MA content.
- Irradiation of FR-MOX fuel in Fast Burner Reactors. (very high burnup)
- Reprocessing of spent FR-MOX fuel in specially designed and licensed facilities.
- Quantitative separation of all TRUs from the spent FR fuel solution.
- Revision of the fission product management: Tc99 separation (head end, HLLW)
- Investigation of the radiological impact of the minor long-lived Fission Products.
- Impact of Platinum metals separation on the overall economics of Partitioning.
- Multiple recycling of FR-MOX fuel with major TRU content until significant depletion.

The most important of these steps will be briefly commented:



### 2.3.1 Conventional reprocessing of LWR-UO<sub>2</sub> fuel.

The current separation factors obtained for major Actinides (99.7%) are in a first approach sufficient to reduce their content in HLLW. The only improvement which might have a significant influence on the longterm toxicity and waste management is the reduction of Medium Level Waste (MLW) which is not vitrified before disposal and occupies a rather large volume compared to HLW.

### 2.3.2 Separation of MA from HLLW.

The current reprocessing technology does not separate quantitatively all the MA. Am and Cm (+ shorter lived TRUs, Bk, Cf...) are quantitatively (>99.5%) transferred to HLLW but Np is partly transferred to the High Active Feed (HAF) solution and partly directed towards the HLLW.

The partitioning of Np needs further chemical extraction steps to insure a quantitative transfer to one single waste or product stream, from which further separations could be performed. Partitioning of all MA from HLLW is presently under investigation in many laboratories throughout the world. (Japan, France, China and some minor nuclear countries) and was studied formerly in the US national laboratories (ANL, ORNL, Hanford...).

Several processes have been studied at the conceptual level and tested in hot facilities among the most important are: the TRUEX, DIDPA, TRPO and DIAMEX processes.

The systems analysis should emphasize the merits and drawbacks of each of these approaches in existing reprocessing facilities or the adaptations to be made to the current flowsheets and their translation into technological modifications to be implemented in present facilities or to be designed in future reprocessing plants with incorporated Partitioning steps.

**Separation of Am(+Cm) is from radiotoxic point of view the first priority**, but the Am(Cm) fraction contains all the Rare Earth (RE) elements which are in terms of quantity, 10 times more important than actinides. (12.4Kg RE compared to 1.27 Kg MA). In order to obtain a TRU fraction with 90% purity a separation factor of 100 is required for the RE fraction. A 99% purity involves a separation factor of 1000, which is a technical limit for elements such as the Lanthanides and the Actinides, with very similar chemical properties.

### 2.3.3. Fabrication of MA targets for heterogeneous irradiation in LWRs.

In the medium term only thermal reactors and particularly LWRs are available for irradiation of MA targets. Fabrication of irradiation targets with industrially representative quantities of MA are difficult to prepare even in pilot-scale hot-cell facilities.

Experience has been gained in the production of isotopic heat sources, but the radiologic context and the ALARA limitations to be expected from regulatory bodies on industrial activities are probably very different from what has been done for military- and space applications.

The presence of large quantities of Am<sup>241</sup> accompanied by 1 to 10 % RE will require fully gamma shielded and remote operated fabrication facilities. The presence of 5% Cm<sup>244</sup> in an Am<sup>241-243</sup> target will multiply the degree of technical complexity due to the additional neutron shielding resulting from the spontaneous fission rate and from the alpha-n reaction in oxide-type isotopic targets.

The report should emphasize the order of magnitude of advanced fuel fabrication capacity required to cope with the potential output from an industrial advanced reprocessing plant, equipped with an Am(Cm) separation module. Recycling of such targets to achieve a significant TRU depletion in the target could be discussed in connection with the issue of multiple recycling of fuels and targets (see section 2.3.9) in fast reactors.

#### 2.3.4. Separation of long-lived fission products.

A number of radiologically important fission/activation products play a potentially important role in the assessment of a geologic repository. In order of importance the following nuclides have to be assessed: the fission products Tc99, I129, Cs135, Se79, Zr93, and the activation product C14.

**Tc99** is a fission product with a half life of 210,000 years which occurs as Tc (TcO<sub>2</sub>) metal in the insoluble residues and as Technetate ion in the HLLW solution. In order to effectively address the longterm radiotoxicity problem both fractions ought to be combined before any nuclear action is taken towards depletion by transmutation. The similarity between Tc and the Platinum metals and the nature of the separation methods (pyrometallurgical techniques) makes this partitioning operation very difficult. Tc displays only a radiotoxicological hazard when submitted to oxidative underground conditions (Tuff, Yucca Mountains). In reducing deep underground aquifers the migration of Tc99 is negligible.

**I129** is separated from the HLLW during the conventional reprocessing operations. The separated fraction can either be stored on a specific adsorbent or discharged into the ocean. Since I129 has a half life of 16 million years it will in any scenario enter into a worldwide dispersion in the geo- or biosphere. I129 is one of the critical nuclides when considering landbased repositories of spent fuel. In a worldwide dispersion scenario its radiotoxic importance is rather limited.

**Zr93** and **Cs135** are two longlived (1.5 resp. 2 million years half-life) nuclides occurring in spent fuel. Separation of these radionuclides from the other fission products for eventual transmutation is almost excluded since they are accompanied by other radioisotopes which are very radioactive (Cs137) or are present in much larger quantities (736 g Zr93 with 3327 g Zr per THM). In order to effectively reduce the radiotoxic potential by neutron irradiation a series of isotopic separation processes ought to precede any target fabrication and this route is presently considered as an almost impossible endeavour from economic point of view.

**C14**, with a half life of 5730 years, is a difficult case because it can potentially enter into the biosphere through its solubility in groundwater and play an important radiotoxicological role because of its incorporation into the biochemical life cycle. Its role in the longterm radiotoxicity is dependent on the physico-chemical conditions occurring in deep underground aquifers or in water unsaturated geospheres.

Some radionuclides discussed in this section ought to be examined in depth by the working group in order to establish **their risk and potential radiotoxic role** in comparison with the TRUs. Their radiotoxicity is between 1000 and 100,000 times less important than TRUs but their contribution to the long term risk is predominant because migration to the biosphere may be much more rapid and generate in the very long term a certain radiation dose to man.

### 2.3.5 Quantitative recycling of U and Pu into LWR-MOX fuel.

Up to now it has been assumed that in the CFC scenario all major actinides (U+Pu) were recycled and used in a next fuel loading scheme: LWR-MOX or FR-MOX. For reasons explained above this has not yet been realized and large quantities of reprocessed U and recovered Pu are still in the engineered storage facilities.

The management of Pu has recently been examined by a OECD-NEA working group and the conclusions should be used to establish the most probable scenario for the recovery and future use of this very controversial spent fuel constituent.

Three alternatives are open -CFC with partial use of U+ Pu as LWR-MOX

- CFC with use of U+ Pu as feedstock for FR-MOX

- AFC with quantitative recovery of U and Pu for LWR-MOX followed by its transfer to FR-MOX at a later stage.

The working group should assess whether the AFC scenario should preferably be situated as a follow up of the LWR-UO<sub>2</sub> reprocessing step or whether it is indicated to transfer all the Pu first to the LWR-MOX step before starting a FR-MOX recycle. If quantitative recovery of all major actinides is not a viable route, it will have a direct impact on the overall separation factor to be achieved for TRUs in general and MAs in particular.

Recycling, or not, of U has a direct influence on the significance of separating Np from spent fuel solutions. The degree of Pu recycling will directly affect the radiotoxic significance of TRU partitioning.

### 2.3.6 Reprocessing of LWR-MOX

Up to now the reprocessing of LWR-MOX has been done by diluting the LWR-MOX fuel with LWR-UO<sub>2</sub> fuel according to the ratio it occurs in the reactor-core. (UO<sub>2</sub>/MOX=2). Reprocessing of spent LWR-MOX can be performed industrially if the reprocessing plant has been licensed for the treatment of increased Pu concentrations and a much higher total Pu inventory.

The radiotoxicity of spent LWR-MOX is ~ 8 times higher than that of LWR-UO<sub>2</sub>. Conventional reprocessing will remove U+Pu which accounts for about 30% of the total alpha activity the residual 70% enters into the HLLW stream and is composed of Cm<sup>244</sup> and Am<sup>241+243</sup>.

**In a perspective of P&T it would be indispensable to remove the TRUs from the HLLW before vitrification.** The techniques to be used are in principle the same as for the LWR-UO<sub>2</sub> fuel (see 2.3.2), but the higher alpha activity level will interfere with the extraction because of increased radiation damage.

Another option is to store the spent LWR-MOX fuel for example during 50 or more years and to let Cm<sup>244</sup> decay (18 years half life) to Pu<sup>240</sup> before carrying out the reprocessing. The chemical extraction processes are after the "cooling" period much easier as the alpha decay heat is reduced by a factor of 7 or more.

### 2.3.7 FR-MOX Fuel fabrication with limited TRU content.

The largest industrial experience has been gained in the FR-MOX fuel fabrication since for several decades FR programmes were undertaken in many nuclear countries. The fabrication of

FR-MOX fuel with 15 to 25 % Pu has been realized routinely. But the Pu quality used for these purposes was derived from low burnup UO<sub>2</sub> fuel with low Pu<sup>238</sup> and Pu<sup>241</sup> contents..

In a perspective of the use of advanced FRs (CAPRA) still higher Pu concentrations are envisaged. ( up to 45%)

In the mean time the burnup of spent LWR-UO<sub>2</sub> fuel has increased to 45 MWd/THM and that of LWR-MOX may reach 50 MWd/THM. The isotopic composition of Pu resulting from the reprocessing of such fuels is seriously degraded. High Pu<sup>238</sup> and Pu<sup>242</sup>, low Pu<sup>239</sup> and Pu<sup>241</sup>. A thorough study of the issues involved was published in 1989 by OECD-NEA and is still valid as source book for the working group on P&T. (PLUTONIUM FUEL An Assessment )

**The recycling of FR-MOX containing high Pu<sup>238</sup> levels and limited amounts of MA is still more difficult and requires the design and construction of remotely operated fuel fabrication plants.**

For homogeneous recycling of MA in FR-MOX, admixtures of 2.5% Np<sup>237</sup> and/or Am<sup>241</sup> are currently studied. Since Np<sup>237</sup> is a pure alpha emitter there is no handling problem involved; but the admixture of Am<sup>241</sup> at the 2.5% level will induce a gamma field around the glove boxes or hot cells. However the major interfering nuclide is Pu<sup>238</sup> at the 3% level which is a heat and neutron source (5KWth/THM; 5.10 E8 neutrons/s per THM.)

The FR-MOX fuel fabrication with limited TRU admixture will also be influenced by the degree of separation of the Rare Earths (strong gamma emitters) and last but not least by Cm<sup>244</sup> which will accompany Am<sup>241</sup> and Pu<sup>243</sup> when separated from HLLW. The presence of even small Cm<sup>244</sup> impurity levels will boost the neutron emission of the resulting fresh FR-MOX fuel.

The working group should establish which separation coefficients ought to be obtained from Rare Earths and from Cm<sup>244</sup> to allow for industrial fuel fabrication operations.

Heterogeneous recycling of MA means to avoid the dilution of troublesome nuclides, e.g. Cm<sup>244</sup>, throughout the fuel fabrication step and carry this operation in small but dedicated facilities.

### **2.3.8 Metal Fuel fabrication for ALMRs and advanced fuels for Burner reactors**

In the frame work of the Integral Fast Reactor project a specific fuel fabrication technology has been developed and tested on cold (and hot) pilot scale. At the FFTF facility metal fuel has been recycled by casting a UPu-Zr alloy on laboratory- and hot pilot scale.

It is obvious that these processes are still in the exploratory stage and cannot be considered as proven technology but their potential should be investigated since metal fuel permits to reach very high burnups and has good neutronic characteristics for transmutation of TRUs.

The working group could summarize the results obtained and draft a series of recommendations for further work in the countries (Japan, and possibly the US) involved in this type of pyrometallurgical development.

Very recently the attention was drawn on the potential of Nitride and Carbide fuels for Fast Burner Reactors. (FBuR)

Nitride TRU fuel containing macroscopic quantities of MA can be produced by a combination of an internal gelation method and a carbothermic synthesis. These nitride fuels can be reprocessed by electrorefining methods similar to the technology developed for metal fuel.

A short analysis of the potential and limitations of this type of new fuel cycle should be included.

### 2.3.9. Fast Burner Reactors (FBuR)

In the frame work of P&T the Fast Breeder Reactor (LMFBR) is not a goal in itself because it produces more TRUs than initially present in the reactor. But very much technological experience has been accumulated since thirty years of R&D&D all over the world which can be transferred to FBuR technology.

The decision of the french government to modify the objective of the Super-Phenix (SPX) reactor

will have far reaching consequences on the future FR programme. Gradually the large SPX prototype reactor working with FR-MOX fuel, will be used as a semi industrial tool to investigate the feasibility of accelerated TRU burning. Therefore it is necessary to modify the reactor configuration (removal of blankets) and even to fundamentally change the fuel and core design. The recently launched CAPRA project will primarily investigate accelerated Pu burning technologies (very high Pu concentrations in the fuel), but MA incineration is also considered. In order to reduce as much as possible the TRU formation, U free TRU fuel with inert matrices are receiving increasing attention.

Several years ago from 1988 on, the Japanese government launched new transmutation projects in the framework of the OMEGA project. The Minor Actinide Burner reactor (MABR), the Particulate Bed Burner Reactor (P-ABR). These projects are being further developed but apart from FR-MOX fuel other types e.g. Nitrides and Carbo-nitrides are investigated as possible alternatives.

The IFR project of ANL evolved into an industrial sodium cooled FR prototype design called the PRISM reactor which is supposed to use metallic fuel (UPuZr) with adjacent pyrometallurgical reprocessing facilities to recycle TRU fuel and discard the fission products. Core design studies and core performance analysis are underway to increase the TRU incineration.

The working panel should comparatively investigate:

- the transmutation capacity of TRUs as a function of neutron fluence and spectrum
- the influence of burnup on the fuel behaviour and the cycle time
- the overall mass balance of TRU input and output inventories
- the evolution of the residual radiotoxicity
- the fuel recycle aspects, total cycle inventory, TRU depletion, residual core content.

This analysis would lead to a better understanding of the relative merits and drawbacks of each conceptual design and help the international nuclear community to better grasp the significance and purpose of the many new reactor concepts which were launched in the frame of P&T.

### 2.3.10 FR-spent fuel reprocessing

The reduction of the radiotoxic inventory resulting from multiple recycling of spent FR-MOX or FR-Metal fuel is only possible if repeated reprocessing of FR-spent fuel is part of the AFC scenario. Irradiation of FR-MOX or FR-Metal fuel is carried out till very high burnup levels. Compared to LWR-MOX fuel the target burnups of FR-fuel are 2 to 3 times higher, i.e. burnups ranging from 120 to 180 MWd/THM are currently to be envisaged, and even higher burnup

levels should be attained in a more distant future. But even these very high burnup levels still correspond to a rather small transmutation or incineration %.

After a prolonged irradiation campaign of e.g. 5 years and a burnup of 150 GWd/THM the degree of depletion is only about 15 to 20 % of the initial TRU inventory. In order to reach an overall TRU depletion yield of 90% it is necessary to recycle the FR-spent fuel between 15 to 17 times according to the expected percentage of losses during each of the chemical extraction operations.

Any reprocessing campaign of spent FR-fuel based on the use of the PUREX process or on an equivalent aqueous extraction process cannot be carried out within a short time interval after the discharge from the fast reactor because the decay heat is much too high. Cooling times between discharge and reprocessing are, as a result, increased from 7 to 10 or 12 years.

The duration of the total AFC cycle becomes 15 to 17 years per cycle. The time interval to reach a global depletion of 90% is consequently of the order of 250 years.

Pyrochemical reprocessing of spent FR-fuel was developed in the frame of the IFR project. In this case FR-Metal fuel irradiated at very high burnup is transferred to a "on-site" pyrometallurgical hot-cell for reprocessing. The molten salt bath (CdCl<sub>2</sub>, LiCl, KCl..) is not subject to alpha radiation damage and much shorter cooling times can be envisaged. However this type of reprocessing technology is still in its infancy and will require extensive R&D before reaching industrial maturity in the civil nuclear sector.

The process involves electrorefining of spent metal fuel with a Cadmium anode, solid and liquid cathodes, and a molten salt electrolyte (LiCl+KCl) at 500°C. The heart of the process is a pyrochemical dissolution process of mechanically clad spent fuel.

Pure U (free of Pu) is electro-transported to the solid Cd cathode. The mixture of Pu+ MA with some U is electro-transported to a second but liquid Cd cathode and separated from the bulk of fission products. The Pu+MA fraction is recycled as metal and casted into the new fuel pins.

Similar pyrometallurgical processes are being developed in Japan by the CRIEPI institute and may in a distant future become an alternative to aqueous reprocessing with the PUREX process for highly irradiated FR-Metal /Nitride spent fuel.

The fundamental advantage of the pyrometallurgical reprocessing processes is their insensitivity to burnup of the spent fuel and the possibility to reduce the overall AFC fuel cycle to 6 or 7 years. By doing so, a given depletion yield could be reached in half the time period scheduled for an AFC with aqueous reprocessing. The material problems due to corrosion, the degree of separation of the TRUs and the waste handling are the most important issues which await fully convincing answers before this type of pyrometallurgical process will become a serious contender for the reprocessing of FR-Metal fuel.

**The P&T working group should assess the merits and drawbacks of each of the approaches taking into account that improvements are possible in the PUREX process and that the pyrometallurgical processes are still in their laboratory phase.**

### **2.3.11 Transmutation issues of longlived fission products**

Transmutation of longlived FP is a very difficult task, because the capture cross sections to transmute the radioactive nuclide into a short lived or stable isotopes requires dedicated and large reactor capacities. Thermal reactors and dedicated Accelerator driven Transmutation (ADT) facilities are the only possible choices for carrying out this very expensive endeavour.

From reactor- technological point of view **the transmutation of Tc99** is the easiest approach since the metallic Tc target is transmuted into metallic Ru100, a stable isotope. However the thermal n-gamma cross section for thermal neutrons is only 20 barns (b) which is extremely low for practical transmutation purposes. The transmutation "half life", i.e. the time necessary to deplete the target with 50 percent varies from one thermal reactor to another but amounts to about 30 years in the best conditions. The reactor must be overenriched to compensate the reactivity loss due to the antireactivity of the Tc targets in the fuel assemblies. The simultaneous production of Tc99 by fission in the LWR-UO2 driver fuel decreases the net transmutation rate and necessitates large Tc loadings. The reactor ought to be dedicated for that purpose.

**Transmutation of I129** is from pure neutronic point of view very similar to that of Tc99, but is from chemical-metallurgical standpoint a much more difficult process since the target product is chemically unstable and the neutron capture reaction product is a noble gas : Xe 130 which has to be vented from the irradiation capsule during its stay in the reactor. Any temperature excursion would result in a release of I129 into the reactor off gases.

ADT technologies with extreme high thermal neutron fluxes ( $10E16$  n/cm<sup>2</sup>/s) should be capable of reducing the transmutation half life. This technology is presently in the conceptual phase. However any type of thermal neutron transmutation will be energetically very expensive.

The radionuclides **Zr93 and Cs135 cannot be considered for reactor transmutation** since they have to be isotopically separated from the other Zr and Cs isotopes before being submitted to irradiation.

The transmutation of **C14** has up to now not yet been considered in the P&T context. Theoretically the C14 released from the spent fuel could be removed from the reprocessing off-gases and transformed into a solid target e.g. BaCO<sub>3</sub>, but the cross section of C14 for thermal neutrons is nearly zero.

Transmutation by charged particles in high energy accelerators is a theoretical alternative in some cases, but the practical feasibility and the economy of such approaches is very questionable.

**The working group should evaluate whether the transmutation of long-lived fission products is a realistic way to pursue and assess the dose benefit/ cost increment ratio for a significant (e.g. 90%), source term reduction of Tc99 and I129. Since these radionuclides have very long half lives, the effect on the dose to man is stretched over geologic periods and surpasses by far the reference lifetime of a repository and even the confinement times of natural barriers.**

### **2.3.11 Conclusions on the role of P&T in the AFC option.**

The P&T option within the AFC scenario, as described above, is the most comprehensive approach which can be reasonably proposed and constitutes a very important extension of the fuel cycle activities in comparison with the CFC and a priori with OTC scenario.

The Working Group on P&T should identify the most attractive of the subscenarios to be pursued in the future within a comprehensive and often wishful type of AFC scenario.

# **Transmutation : Physics and Strategies**

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- Much can be said (and has been said) about "criteria".
- However, the simple parameter represented by the activity of the materials stored in a repository (or the "source of potential radiotoxicity") is still widely used. Much debate, not many new suggestions.
- Long-lived fission products : since 1990 (1st OECD Exchange Meeting here in MITO) no much progress towards a clear assessment of the relevance of their associated toxicity.

## Transmutation (with neutrons) :

- everything has already been said !

few clear guidelines :

- fission should be privileged and neutron economy is essential,
- the evaluation of the consequences on the physics parameters of the fuel cycle is crucial. Again, it is better to "transmute" by fission than by capture,
- Np is not a priority : Americium is !
- Cm : no unique strategy is easily available,
- when evaluating benefits/drawbacks of transmutation, the role of the strategy chosen for Pu management is dominant.

## The Physics of Transmutation

- At equilibrium :

$$\frac{dN_J}{dt} = \lambda N_J + S_J \equiv 0$$

( $S_J$  : continuous feed of isotope J to the system)

$\Rightarrow$  equilibrium is readily achieved !

- At each isotope J, one can associate a "neutron consumption/fission"

$D_J$  ( $D_J < 0$  means "production").

One can define a  $D_{\text{comb}}$  for a fuel, which is a mixture of isotopes J with fractions

$\epsilon_J$  :

$$D_{\text{comb}} = \sum_J \epsilon_J D_J$$



- The system neutron balance can be expressed in terms of a (positive or negative) neutron "surplus"  $G$  :

$$G = - D_{\text{comb}} - (CM + L)$$

( $CM$  and  $L$  are the neutrons/fission lost in parasitic captures or by leakage).

- Since  $CM + L$  is generally equal to  $\approx 0.3$  n/fission (for most systems) one can assess what system can allow "transmutation" (fast reactors or thermal reactors with increased fuel enrichment).

- For a source-driven subcritical system :

$$G = S_{\text{ext}} - D_{\text{comb}} - (CM + L)$$

In a subcritical system driven by an accelerator which supplies protons of  $\sim 1.5$  GeV to a target producing  $\sim 40$  neutrons/proton (i.e.  $\text{Max } S_{\text{ext}} \simeq 1 \div 1.5$  n/fission),

$\Rightarrow$  The relations among the subcritical  $K_{\text{eff}}$ , the fraction  $f$  of the fission energy used to feed the accelerator, the proton current  $i$  and the power  $P$  of the subcritical core give for example :

$$\begin{array}{llll} K_{\text{eff}} \simeq 0.95 & f \simeq 10 \% & P \simeq 300 \text{ MWth} & i \simeq 5 \text{ mA} \\ K_{\text{eff}} \simeq 0.9 & f \simeq 20 \% & P \simeq 1000 \text{ MWth} & i \simeq 30 \text{ mA} \end{array}$$

$\Rightarrow$  The surplus of neutrons has a price ( $f$ ). However, can help to provide an alternative option to critical fission reactors for transmutation if :

- fuels very exotic (better  $K_{\text{eff}} \ll 1$  !)
- possible to concentrate all minor actinides in a limited number of dedicated machines ( $\sim 2 \div 5$  % of a total power reactor park).

Transmutation in critical reactors (Homogeneous recycling) :

- Both LWRs and FRs :
  - Reactivity loss over the cycle : reduced
  - Temperature coefficients (and boron effectiveness) : worse
  - Coolant void reactivity effect : less negative (or more positive).
- For LWRs : need of over-enrichment (neutron economy).  
However : Higher moderator-to-fuel ratio is beneficial.
- Maximum allowable MA : 5 % of total HI for FRs - 1 ÷ 2 % for LWRs.

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Heterogeneous recycling :

- Targets at the periphery of the core for minimum perturbation to power distributions.

⇒  $\left\{ \begin{array}{l} \bullet \text{ Performances (mass inventory)} \\ \bullet \text{ Consequences on the physics characteristics of the fuel cycle} \end{array} \right.$

## Mass inventories after 1 recycle

	FR of EFR type (1500 MWe) Homogeneous recycling (content : 2.5 %)	FR of EFR type Heterogeneous recycling in radial blankets (content : 40 %)	PWR-MOX with $V_m/V_F = 3$ (content : 1 %) BU = 4.7 GWd/t	PWR-UOX Heterogeneous recycling in the core BU = 42 GWd/t
<u>Np</u>				
Consumption (Kg/TWhe)	10	13	11	15
Consumption rate (%)	60	60	45	38
Fission rate (%)	27	24	9	3
<u>Am</u>				
Consumption (Kg/TWhe)	9	14	10	8
Consumption rate (%)	45	60	42	70
Fission rate (%)	18	22	6	13



## Consequences on physics parameters of fuel cycle

Type of recycling		HOMOGENEOUS			HETEROGENEOUS
		2.5 % Np	2.5 % Am	2.5 % Cm	40 % Am + MgO
FABRICATION	Actinide content				
	Power	× 1	+ 71 %	× 24	× 14
	Activity	× 1	+ 3 %	× 2	× 19
	Gamma source	× 1	× 4	× 12	× 55
	γ-dose at 1 m	× 4	× 76	× 470	× 1680
	Neutron source	× 1	+ 40 %	× 1700	× 7
	Decay heat	+ 2 %	+ 1 %	- 4 %	÷ 3
	Activity	+ 0.3 %	+ 1 %	- 2 %	÷ 5
	Gamma source	- 0.7 %	- 1 %	- 3 %	÷ 3
	γ-dose at 1 m	+ 6 %	+ 2 %	- 1 %	÷ 2
End of irradiation	Neutron source	- 5 %	× 4	× 6	/
	Decay heat	+ 80 %	× 3	× 6	× 32
	Activity	+ 5 %	+ 13 %	+ 30 %	× 5
	Gamma source	+ 1 %	+ 5 %	+ 7 %	+ 43 %
	γ-dose at 1 m	- 1 %	- 2 %	- 2 %	÷ 4
End of irradiation + 5 years	Neutron source	- 6 %	× 4	× 8	/

## Am transmutation in a FR

- Heterogeneous mode
- Multirecycling ? Large amounts of Cm are produced
- Cm : what strategy ?
  - Temporary storage → decay to Pu (60 ÷ 100 y !)
  - Recycle as for Am - Higher mass MA production
  - Reduce its production :
    - fission Am to 90 ÷ 95 %
    - long irradiation in a high flux (thermalised) at the periphery of a FR.
    - However, high DPA (> 200 DPA NRT ?)
    - Also : optimisation to avoid too small initial Am loadings

## Application to a reactor park

- Whatever the strategy, Pu recycling reduces potential radiotoxicity by factor  $\sim 2 \div 3$  (in particular if FRs are used) : this is a first step in the right direction !
- If Am is recycled, in an equilibrium park :

PWR-UOX	PWR-MOX	FR
70 %	10 %	20 %

the potential source of radiotoxicity is reduced by factors f :

At t (years)	$\Rightarrow$	102	103	104	105	106
f	$\Rightarrow$	71	75	84	140	80
(theoretical f)	$\Rightarrow$	510	480	630	700	570)

if Cm is not put to wastes - Losses : Pu 0.1 % Am, Cm, Np 1 %  
if Cm is put to wastes, f are reduced :

f	$\Rightarrow$	10	11	7	94	32
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## Conclusions

- Data and methods for transmutation :

⇒ Need of improvements (reduction of uncertainties).

However, present uncertainties allow reasonable evaluations.

- Physics analysis (at equilibrium) allows to point out major features and to intercompare different systems/strategies.

- Accelerator-driven systems : a few extra neutrons available, but at a cost. Can help if dedicated (i.e. MA-fuelled) reactors are envisaged in a "double-strata" - type of approach.

- Fast reactors can do most of the job. Limited consequences on the fuel cycle. Transmutation in PWRs : less "attractive".

- Major issue : target development for (once-through) Am irradiation.

⇒ Role of SUPERPHENIX : irradiations foreseen.