

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

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ABSTRACT

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

INTRODUCTION

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

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

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

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

CHEMICAL SEPARATION OF LONG-LIVED RADIONUCLIDES

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

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

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

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

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

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

FUEL AND TARGET DEVELOPMENT

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

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

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

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

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

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

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

Reference and P&T scenarii

The three reference scenarii are:

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

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

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

Potential radiotoxicity inventory

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

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

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

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

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

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

Cost assessment

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

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

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

The main results of this study are summarised below.

Transmutation of minor actinides (MA)

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

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

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

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

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

Transmutation of long-lived fission products

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

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

3)- Nuclear data libraries for transmutation studies

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

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

The main achievements of this study are presented below.

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

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

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

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

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

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

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

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

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

CONCLUSION

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

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

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

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

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

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

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

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