Safety and environmental aspects of partitioning and transmutation of actinides and fission products

Proceedings of a Technical Committee meeting
held in Vienna, 29 November–2 December 1993

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FOREWORD

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 reuse in fuel and disposal of solidified high level waste in a deep geological repository.

There is scientific consensus that the current waste management concept provides adequate protection to the population and the environment by sufficient confinement of radionuclides. However, there is also an interest in investigating the possibility of further reducing the source term. The purpose of the P&T option is to reduce the long term radiotoxicity of disposed waste by the separation of minor actinides and long lived fission products from high level liquid waste and transmutation of these nuclides into stable isotopes in nuclear reactors or accelerators. This could significantly reduce the period during which the waste materials require barriers to protect man and environment.

P&T is a complex issue with safety, technological, economic and public acceptance aspects involved. 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, Russian Federation) and international (OECD/NEA and European Commission) programmes on P&T are 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 than with the studies of safety implication of P&T.

Recognizing this, the IAEA organized a Technical Committee Meeting on Safety and Environmental Aspects of Partitioning and Transmutation of Actinides and Fission Products which was held in Vienna from 29 November to 2 December 1993. The meeting, which was attended by twenty-eight participants representing eleven countries and four international organizations, 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 and provided information on the IAEA programme on P&T.

The results of the Technical Committee meeting are presented in this document. The IAEA wishes to thank all participants of the meeting for their fruitful contributions and especially the Chairman, Mr. L.H. Baetsle. The Scientific Secretary of the IAEA responsible for the organization of the meeting and for the compilation of this document was Mr. A. Grigoriev of the Nuclear Materials and Fuel Cycle Technology Section, Division of Nuclear Fuel Cycle and Waste Management.
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SUMMARY OF THE TECHNICAL COMMITTEE MEETING

1. CURRENT STATUS AND PROSPECTS

The objective of the Technical Committee Meeting on Safety and Environmental Aspects of Partitioning and Transmutation was to discuss the studies on fundamental safety aspects of partitioning and transmutation (P&T), to review the current status and progress of national and international programmes on P&T, to identify the most important directions of national efforts and international co-operation in this area and to provide technical advice to the Secretariat regarding the IAEA programme on P&T.

The participants noted the considerable interest among participating Member States and international organizations in the P&T option 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 reuse in fuel and disposal of high level waste (HLW) in a deep geological repository. The once-through nuclear fuel cycle involving direct disposal of spent fuel remains unaffected by the P&T option.

There is a worldwide scientific consensus that the current waste management concept provides adequate protection to the population and the environment by sufficient confinement of radionuclides. According to the participants the purpose of P&T is to reduce the long term effects of radiotoxicity of actinides and long lived fission products but it cannot eliminate the need for a geological repository. P&T is a complex issue with safety, technological, economic and public acceptance aspects. In some countries (France, Japan, Russian Federation) 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.

The participants emphasized the potential use of plutonium as a nuclear fuel resource for light water reactors (LWRs) and fast reactors (FRs), but acknowledged the strong interrelation between plutonium and minor actinides (MAs) during the future recycling processes of actinides in view of their long term hazard reduction. The reduction of the hazard of long lived nuclides has to be achieved gradually: from modest gains (analogous to achievements in air pollution) to significant gains which may involve important economic penalties. 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\(^1\), 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\(^2\) over a long term period of time.

The participants summarized the progress being made in the fields of partitioning, of fuel and target developments and of transmutation in the following way.

\(^1\)Potential hazard involves the source term without taking into account the geological barriers.
\(^2\)Residual hazard means the radionuclides released to the biosphere.
2. PARTITIONING

Two main fields of work are involved in the partitioning of plutonium, minor actinides and long lived radionuclides:

- wet (aqueous) separation methods that are implemented in association with the Purex process by modifying it or extending it by additional separation steps;
- dry pyrometallurgical processes, which may follow the Purex process or completely replace it. Such processes are under investigation in Japan, Russia and the USA.

Two main trends in separation processes are apparent:

- a tendency to separate neptunium, the soluble portion of the technetium, and possibly zirconium, directly during the Purex process;
- recovery of other elements from HLW solution and from off-gas dissolver.

For minor actinides (mainly americium and curium) data have been presented on several extractants. Most of extractants are applied in a two-step process: one step in which MA and rare earth (RE) are extracted together, and then a second step in which MA are separated from RE. The extractants concerned are: carbamoyl phosphine oxide (CMPO in the TRUEX process) (in India, Japan, Russia, the European Commission), dicarbollyde (Russia, the Czech and Slovak Republics, the European Commission), di-ethylhexyl phosphoric acid (HDEHP) (Sweden), di-isodecyl phosphoric acid (DIDPA) (Japan, European Commission), three alkyl phosphine oxide (TRPO) (European Commission, Russia) and diamides (France, European Commission). The main advance has been the attainment of high extraction factors that have been confirmed on fully active solutions, sometimes on a pilot plant scale. The extractants have normally been used in liquid extraction processes, and sometimes in sorption techniques.

This two-step process currently has some drawbacks: it involves the extraction of a large quantity of RE, which is not required for P&T, with a smaller amount of MA which is required. The separation of americium from RE remains difficult, so specific extractants for americium are under development, and this should be pursued further in the future. The aim is to improve the specificity, and also to reduce the production of secondary waste.

Curium presents specific problems: difficulties in aqueous separation, and then, if separated, safety problems in target production and irradiation. The problem of curium needs further consideration.

Behaviour of fission products during reprocessing is variable and complex. In the case of technetium, for example, a substantial fraction of the total inventory occurs as insoluble residue from dissolution, together with the platinum group metals.

Reduction in potential toxicity of waste is dependant on the reduction of losses during reprocessing and the minimization of secondary waste streams arising not only from reprocessing operations but also from the subsequent partitioning and recycling.

Some elements present a specific problem in that their long lived isotopes are accompanied by stable or shorter lived isotopes. Transmutation would require preliminary isotope separation.

For the short lived elements such as strontium-90 and caesium-137, separation might be envisaged to reduce heat dissipation and hence the volume required in a deep geological repository.
Conclusions

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.

3. 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 (KNK II/b and SUPERFACT irradiation experiments). Studies are still in progress for recycling in a LWR, e.g. ACTINEAU project.

Oxide, metal alloy and inert matrix fuels and targets containing MA and $^{99}$Tc 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.

The following aspects are considered to be important for the characterization of the fuels/targets for transmutation of MA and FP:

- capability to withstand stringent irradiation conditions considering both fuel and cladding;
- optimization of loading of MA and FP in fuel from the viewpoints of structural behavior and reactor operation;
- capability to reprocess such fuels/targets;
- handling in fabrication, discharge and storage.

Processes have to be developed for the fabrication of fuels/targets on an industrial scale. They have to meet criteria such as: minimization of losses, radiation protection aspects, criticality and heat generation. Such processes will require dedicated remote handling facilities.

The technical feasibility of the use of these fuels/targets has to be demonstrated through irradiation experiments in proper environments, and post-irradiation examinations for fuel and cladding behavior. Suitable experimental irradiation facilities have to be provided. Such studies are required for high burnup LWR– and FR–MOX fuel (alloys and inert matrices) and for advanced fuel concepts.

Extensive studies would also be required for targets (solid or liquid) for accelerator driven systems, including compatibility studies of structural material for the liquid targets.

Modelling of fuel performance (structural self-shielding, isotope diffusion and fission gas release) during irradiation is required.

Proliferation aspects of such fuels/targets have to be considered. Decrease of proliferation risk is expected by the associated higher radioactivity of the nuclides involved.

Cost evaluation is needed for the fabrication stage of fuels/targets containing MA and FP.

Conclusions

Within the constraints of transmutation efficiencies and partitioning selectivity fuels and targets are being developed to investigate technical feasibility for the proposed P&T schemes.
4. TRANSMUTATION

Transmutation is a measure to reduce the amount of long lived nuclides in waste which are the source of long term potential hazard. The overall transmutation goal, as a means of radiological hazard reduction, has to be seen also from the aspect of the hazard period and, as a consequence, of confinement time.

Many transmutation methods have been proposed. Those can be structured in several ways:

1. Type of device: – fission reactor; – accelerator driven system; – fusion.

2. Type of neutron spectrum: – thermal; – fast.

3. Type of scenario: – power reactor; – special system, symbiotic with the power reactor fuel cycle.

The targets for transmutation are minor actinides and long lived fission products. Degraded plutonium is also a candidate for transmutation because it is a source of MA. In the short term, separated plutonium is a target for transmutation because large stockpile of surplus plutonium should be eliminated.

4.1. Transmutation in thermal reactors

It appears that LWRs do not present a suitable transmutation mode for minor actinides, because the neutron capture cross section is much larger than the fission cross-section for thermal neutrons. In the thermal energy range the build up of heavier actinides is preferred compared to the destruction via fission. Only an increase in neutron flux density up to $10^{15}$ n/cm$^2$.s and well above could give an appreciable destruction rate. The advantage of thermal high flux reactors ($\geq 10^{16}$ n/cm$^2$.s) is that they can definitively transmute fission products such as $^{99}$Tc or $^{129}$I to a satisfactory degree. No thermal high flux reactor with corresponding large irradiation volume in the high flux region exists.

4.2. Plutonium recycle in LWR as MOX

It is foreseen in many countries to recycle plutonium in LWRs as MOX fuel (ca. 30% of the core or even as full MOX loading). This recycling scheme can possibly stabilize the plutonium accumulation. It should be mentioned that if MA, as e.g. Np or Am are admixed to the fuel to an amount of a few percent (1%–3%), the safety characteristics of the LWR plant are not heavily distorted. Because of Doppler or coolant void effects the overall plant behavior will not be changed.

4.3. Transmutation in fast reactor

This reactor system seems to have the highest preference as a transmutation in fast reactors. But it is not yet fully understood how much of the actinides and fission products can be destroyed in such a facility. It was reported that at present there are definite possibilities for transmutation in conventional FR of MA and FP generated in 2–5 LWRs. But the aim is transmutation of MA and FP from 10 or more LWRs. The limitation of burning MA and FPs is due to the worsening of the safety coefficients of Doppler and void effects, except if some extreme designs are assumed as pan-caked cores, etc. However,
some advantage of MA admixture to fuel can be expected in reduction of excess reactivity. This could result in the conclusion that an optimum has to be found in the sense of making fast burner cores smaller in size.

4.4. Limits of reactor transmutation

If the burnup is increased to higher than about 50 000 MWd/t in LWR, then the nuclide composition changes (more $^{238}$Pu, $^{240}$Pu, $^{242}$Pu) with more transplutonium elements especially $^{244}$Cm and Cf. From this list of radionuclides $^{244}$Cm will be difficult to destroy.

Multiple recycling always causes deterioration of the final plutonium composition. Consequently, the plutonium concentration in the fuel increases, otherwise the recycling has to stop. Therefore, the global transmutation rate is decreased. The spent fuel after many recyclings is highly toxic and gives high hazard values despite preceding transmutations. This spent fuel then has to be destroyed by using a high performance accelerator machine.

4.5. Accelerator driven transmutation

Unlike LWRs and fast reactors, accelerator driven transmutation facilities are not yet a technical reality. To increase proton currents by two orders of magnitude from the present record level would be a very challenging task. Some components in beam losses are quadratic in current which may lead to significant radiation hazards. Subcriticality is an important advantage in preventing runaways but that is not the only problem and other ones may be more severe for accelerator driven blankets than for traditional reactors.

During the TCM the observation was made that a machine for the full elimination of MA and FP in spent fuel should be considered. This machine would preferably be an accelerator driven multiplying system, generating fast and/or thermal neutrons of high flux density. The JAERI and BNL proposals for fast neutrons systems have been investigated with respect to burning actinides and fission products (Tc). It was shown that Tc could be burned effectively, but at the same time almost an equal amount is created via fission processes on plutonium in the system, with a total net effect near zero.

Further research is needed to clarify which accelerators with good transmutation rates could be used in the future. The main emphasis should be placed on the destruction of FP, because FP cannot be transmuted to any marked degree in reactors, unless a large excess reactivity can be generated (e.g. heavy water reactor (HWR) or in FR with moderated subassemblies).

Conclusions

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 breakthrough in technology are we likely to reach the expected goal.
LIMITATIONS OF ACTINIDE RECYCLE AND WASTE DISPOSAL
CONSEQUENCES – A GLOBAL ANALYSIS

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Abstract

Recently very much emphasis has been put on partitioning and transmutation (P&T) of minor actinides and long lived fission products as an alternative or additional waste management option. This approach supposes that quantitative recycle of uranium and plutonium can be achieved and that the minor actinides and the fission products constitute the only residual radionuclide stream to be reckoned with in the long term. However the fuel cycle with reprocessing and recycle of plutonium in LWRs is only a partial solution to the decrease of the cumulative plutonium inventory resulting from LWR operation and leaves in the most advantageous case of a double recycle scenario a residual plutonium inventory of ~50% which remains with the LWR-MOX spent fuel or, as a separated (and degraded) plutonium product. The influence of P&T on nuclear waste management strategy is discussed in this paper.

1. INTRODUCTION

Very much emphasis has been put on transmutation of minor actinides and long lived fission products as an alternative or additional waste management option. This approach supposes that quantitative recycle of uranium and plutonium can be achieved and that the minor actinides and the fission products constitute the only residual radionuclide stream to be reckoned with in the long term.

However the fuel cycle with reprocessing and recycle of plutonium in LWRs is only a partial solution to the decrease of the cumulative plutonium inventory resulting from LWR operation and leaves in the most advantageous case of a double recycle scenario a residual plutonium inventory of ~50% which remains with the LWR-MOX spent fuel or as a separated (and degraded) plutonium product.

The depleted uranium inventory and the separated uranium from reprocessing are in terms of mass, large bodies which have to be stored in the short term; up to now very little attention was given to these U inventories.

Recycle of $U_{\text{Repro}}$ in LWRs is one way to make proper use of an otherwise unused product-stream; it results in a decrease of fresh uranium demand with its environmental impacts during mining and milling. Depleted uranium can only be used to a very limited extent in LWR-MOX fuel. If this product-stream from reprocessing is to be exploited energetically, the use of fast reactors is a necessity otherwise it will have to be stored as a strategic resource or disposed as a waste.

In order to reduce significantly the Pu and MA source term the use of a fast neutron spectrum device is indispensable. MOX fuelled fast reactors or advanced liquid metal cooled reactors (ALMRs) constitute a first choice for the decrease of the Pu-MA source term. However as far as present developments are concerned burnup maxima of 15 at.% and mean values up to 12 at.% have currently been achieved. Consequently a large number of recycle operations have to be scheduled in order to reduce the Pu-MA inventory of LWR-MOX spent fuel to e.g. 10% of the initial Pu-MA content from UO$_2$ LWRs.

Accelerator driven transmutation is a potentially interesting but very complex technology which bears promises to reach much higher burnups than achievable in fast reactors and to accelerate the actinides' transmutation.
The nuclear power production in Western Europe amounts to 120 GWe and in the USA about 100 GWe; the other countries make up a joint capacity of 120 GWe. Throughout the world about equal capacities (~ 130 to 140 GWe) are committed to the fuel cycle with reprocessing and to the direct disposal option.

In order to normalize the data in this paper the quantities will be expressed in appropriate units per tonne of heavy metal (t HM) and summarized per 100 GWe installed capacity with a load factor of 0.8 and a mean fuel burnup of 40 GWd/t. Table I summarized the output of a 100 GWe NPP park.

<table>
<thead>
<tr>
<th>TABLE I. ANNUAL FUEL CYCLE OUTPUT FROM A 100 GWe NPP PARK</th>
</tr>
</thead>
<tbody>
<tr>
<td>Load factor : 0.8</td>
</tr>
<tr>
<td>Electricity production</td>
</tr>
<tr>
<td>Discharged fuel</td>
</tr>
<tr>
<td>Uranium content</td>
</tr>
<tr>
<td>Plutonium content per t HM</td>
</tr>
<tr>
<td>Plutonium content</td>
</tr>
<tr>
<td>Total plutonium inventory</td>
</tr>
<tr>
<td>Total minor actinide (MA) inventory</td>
</tr>
<tr>
<td>HLLW</td>
</tr>
</tbody>
</table>

Table I shows clearly the magnitude of the residual actinide problem (U+Pu+MA) which has to be resolved by an appropriate waste management strategy.

2. ONCE-THROUGH STRATEGY

According to this scenario the entire output of spent fuel is to be stored and eventually disposed of in deep geologic structures. Since a disposal site is limited (in the USA) to 70 000 t HM according to the present US legislation, every 32 years a new disposal site has to be made available. The Pu+MA inventory of such a disposal site would amount to ~800 t HM Pu which is a worrisome prospect. Taking into account that depending on the cooling time 57-66% of the Pu present is fissile material, the loss of resources amounts to 408-473 t Pu fissile and long term criticality issues have to be investigated before adopting such strategy.

3. REPROCESSING AND Pu RECYCLE STRATEGY

The reprocessing strategy with reuse of Pu in LWR-MOX fuel with partial reuse of reprocessed uranium and with vitrification of high level waste is historically the most important venture in Europe.

The quantity of Pu produced by 100 GWe NPP park amounts annually to 22.53 t Pu. This quantity is sufficient to provide 20% of a second core reload i.e. a park loaded with 80% UO₂ and 20% MOX fuel. During this second core irradiation Pu is produced in the UO₂ core and depleted in the MOX fuel. The overall balance of electricity, fissile or fertile products, and waste streams is given in Table II.

The economic benefit results from the surplus electricity production (140 TWh) which has to compensate the costs of reprocessing, transport and MOX fuel fabrication.
If we compare the data of Tables I and II it is obvious that the total Pu inventory after two "once through" cycles would amount to 45 t HM Pu while it is reduced to 33.7 t HM Pu if a combination is made of one UO$_2$ and one UO$_2$-MOX cycle with intermediate reprocessing and partial MOX use in the second cycle. The overall Pu inventory reduction is important (25%) but does not change the long term waste management approach since a substantial fraction, i.e. 46% (or 15.68 t HM Pu) remains occluded in the discharged MOX spent-fuel elements.

### TABLE II. OUTPUT OF A 100 GWe NPP PARK WITH 20% MOX FUEL AT A GLOBAL BURNUP OF 40 GWd/t AFTER 7 YEAR COOLING

<p>| | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Electricity</td>
<td>700 TWh (560 TWh from UO$_2$, 140 TWh from MOX)</td>
</tr>
<tr>
<td>Fresh fuel</td>
<td>2200 t (1760 t HM UO$_2$, 440 t HM MOX)</td>
</tr>
<tr>
<td>Discharged fuel</td>
<td></td>
</tr>
<tr>
<td>1760 t HM (UO$_2$)</td>
<td>2200 t</td>
</tr>
<tr>
<td>440 t HM MOX</td>
<td>1669 t U, 70.4 t FP, 18.02 t Pu, 2.06 t MA</td>
</tr>
<tr>
<td>Totals</td>
<td>2073 t U, 88 t FP, 33.7 t Pu, 3.86 t MA</td>
</tr>
<tr>
<td>Reprocessing yields of UO$_2$ fuel</td>
<td>1660 T U 18.0 T Pu, 250 m$^3$ HLW (70.4 t FP + 2 t MA)</td>
</tr>
<tr>
<td>Spent MOX fuel</td>
<td>440 T HM (with 15.68 t Pu, 1.8 t MA)</td>
</tr>
</tbody>
</table>

### 3.1. Reprocessing of MOX fuel

Reprocessing of a spent MOX fuel load is technically feasible but much more difficult than conventional UO$_2$ spent fuel. Table III shows a comparative spent fuel composition of UO$_2$ and MOX fuel at the same burnup, 7 years after discharge and the resulting compositions 100 and 500 years after discharge if no reprocessing would have been done.

The technological difficulties associated with reprocessing of MOX fuel are the following.

- Industrial reprocessing of large quantities (440 t HM) of MOX fuel requires a very advanced equipment to cope with the increased criticality risk during MOX dissolution. The construction of dedicated head-end facilities has to be envisaged. Small quantities of MOX fuel can be "diluted" with UO$_2$ fuel.

- The high concentration of $\alpha$ emitting nuclides particularly Pu-238 and Cm-244 will seriously increase the solvent degradation. The overall $\alpha$ level is 7 times higher with recycled MOX fuel than with equivalent UO$_2$ fuel of the same burnup. The use of centrifugal extractors in the first extraction cycle might perhaps mitigate the solvent degradation due to Cm-244, but the Pu-238 influence remains until the loaded TBP has been stripped in the BX column.

- The occurrence of (900-1000 g Pu-238/t HM) will complicate the Pu purification, particularly the reextraction/striping steps, due to a higher level of TBP degradation products especially DBP which strongly complexes Pu species.

- The heat emitted by Pu-238 (0.5 W/g) will call for increased precautions during the storage and transportation of Pu containers. This phenomenon will become much more important if multiple MOX recycle would be considered.
TABLE III. EVALUATION OF PU AND MA CONTENT IN UO\textsubscript{2} AND MOX SPENT FUEL OF 40 GWd/t COMPARED TO REPROCESSING RESIDUES OF UO\textsubscript{2} FUEL

<table>
<thead>
<tr>
<th>DECAY TIME</th>
<th>t = 7y</th>
<th>t = 100y</th>
<th>t = 500y</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>UO\textsubscript{2}</td>
<td>MOX</td>
<td>UO\textsubscript{2}</td>
</tr>
<tr>
<td>Pu-238 Ci</td>
<td>3.48 \times 10^3</td>
<td>1.59 \times 10^4</td>
<td>1.67 \times 10^3</td>
</tr>
<tr>
<td>Pu-238 g</td>
<td>203</td>
<td>934</td>
<td>97</td>
</tr>
<tr>
<td>Pu TOT Ci</td>
<td>1.07 \times 10^5</td>
<td>5.20 \times 10^5</td>
<td>3.79 \times 10^5</td>
</tr>
<tr>
<td>Pu TOT g</td>
<td>10,243</td>
<td>35,644</td>
<td>9,140</td>
</tr>
<tr>
<td>Pu TOT Ci</td>
<td>4.67 \times 10^3</td>
<td>1.95 \times 10^4</td>
<td>3.78 \times 10^3</td>
</tr>
<tr>
<td>Np-237 Ci</td>
<td>0.4</td>
<td>0.166</td>
<td>0.49</td>
</tr>
<tr>
<td>Np-237 g</td>
<td>569</td>
<td>237</td>
<td>705</td>
</tr>
<tr>
<td>Am-241 Ci</td>
<td>1.5 \times 10^3</td>
<td>8.2 \times 10^3</td>
<td>4.2 \times 10^3</td>
</tr>
<tr>
<td>Am-241 g</td>
<td>438</td>
<td>2,400</td>
<td>1,230</td>
</tr>
<tr>
<td>Am-243 Ci</td>
<td>25.6</td>
<td>198</td>
<td>25.3</td>
</tr>
<tr>
<td>Am-243 g</td>
<td>128</td>
<td>996</td>
<td>127</td>
</tr>
<tr>
<td>Cm-243 Ci</td>
<td>21.1</td>
<td>152.7</td>
<td>2.2</td>
</tr>
<tr>
<td>Cm-243 g</td>
<td>0.408</td>
<td>2.96</td>
<td>0.042</td>
</tr>
<tr>
<td>Cm-244 Ci</td>
<td>2.75 \times 10^3</td>
<td>3.25 \times 10^4</td>
<td>78.6</td>
</tr>
<tr>
<td>Cm-244 g</td>
<td>34.1</td>
<td>403.2</td>
<td>0.97</td>
</tr>
<tr>
<td>Cm-245 Ci</td>
<td>0.27</td>
<td>14.1</td>
<td>0.268</td>
</tr>
<tr>
<td>Cm-245 g</td>
<td>1.59</td>
<td>46.18</td>
<td>1.58</td>
</tr>
<tr>
<td>Cm-246 Ci</td>
<td>0.56</td>
<td>1.67</td>
<td>0.55</td>
</tr>
<tr>
<td>Cm-246 g</td>
<td>1.83</td>
<td>5.43</td>
<td>1.8</td>
</tr>
</tbody>
</table>

The use of plutonium resulting from MOX recycle will mostly affect the operations in the fuel fabrication plant where increased levels of Pu-238 and Am-241 will boost the neutron and gamma radiation around the glove boxes.

The neutron emission increases with 30% when Pu from spent MOX fuel would be reworked in a fuel fabrication plant. This will mostly affect the powder mixing and grinding steps where large quantities of Pu bearing materials are handled simultaneously. The multiple recycle of MOX fuel as would be required in a P&T scenario will further increase the handling difficulties and call for the development of fully "remote handled" fuel fabrication facilities with remotely operated maintenance.

Delayed reprocessing of spent MOX fuel seems at the present state of the art of the technology as the best alternative to direct disposal. A storage of 50 years, time period also considered for vitrified fission products, is beneficial as it would decrease the Cm-244 content of the spent MOX fuel with a factor of 7 and ease further separations and purification operations. The choice between disposal and reprocessing after that time interval will depend on the availability of fast reactors or accelerator driven transmutation systems for burning massively the Pu and residual MA.
3.2. Minor actinides and partitioning of fission products

Reprocessing is an indispensable step in making Pu available for reuse in LWR-MOX fuel as discussed above but this operation leads to the generation of HLLW which contains MA and fission products. The fission products are a "short term" concern. Each tonne of spent UO$_2$ fuel contains ~5000 Ci $\alpha$ emitting MA. In order of importance Cm-244, Am-241 and far behind Np-237.

Vitrification of fission products is the last step before long term storage and disposal. This implies that the partitioning operation of MA ought to be carried out shortly after reprocessing. But during that time interval (7 to 10 years) Cm-244 is the strongest $\alpha$ emitter which will influence the methods and equipment to be used for these partitioning steps.

Successively the partitioning operation will require the following separations:

- Extraction of MA + rare earths from HLLW.
- Separation of MA from rare earths.
- Conditioning Np, Am and Cm for long term storage.

The strongly radioactive rare earths and the highly-active isotope Cm-244 ($\alpha$ and neutron emitter) interfere with the use of complex and fragile organic molecules which undergo radiation damage. By combining the TRUEX process with an actinide/rare earth separation (CMPA, DTPA) Am and Cm can both be removed together from the HLLW.

The Am-Cm fraction has to be stored indefinitely or transferred to a transmutation scenario.

The rare earths are recombined with the other fission products (Cs-137, Sr-90, Ru-106, etc.) and transferred to the vitrification facility. The vitrified products have to be stored during ~50 years before disposal because of their high heat output.

By delaying the reprocessing step, Am-241 instead of Cm-244, becomes the most important $\alpha$ emitting radionuclide. Some long lived fission products are of importance in a future partition strategy. Table IV contains the most important long lived ($t_{1/2} \geq 30\ y$) fission products. The data for UO$_2$ fuel are very similar and are not treated separately.

As can be deduced from the data of Table IV, Cs-137 and Sr-90 determine the decay pattern of the fission products up to 500 years. Beyond that period some long lived fission products draw the attention: Tc-99 and Sm-151 make up 70% of the total residual FP activity but Tc-99 is the most important one because of its very long half-life.

Though very low in absolute value I-129 plays an important role in the dose to man because of its biologic concentration in the thyroid gland. According to present practice I-129 is discharged into the sea or stored on land as AgI.

According to new trends in the reprocessing technology Tc-99 can be separated from the other fission products by an intermediate extraction process between the HS and BX column of the first PUREX extraction cycle. However this "partitioning" has not been conceived for environmental reasons but because Tc-99 which is present in the FP mixture at a level of ~926 g/t HM interferes with the quantitative Pu (III) stripping from the loaded TBP as it destroys hydrazine, a reductor used as valency stabilizer. The partitioning of Tc-99 is important as this very long lived nuclide could interfere with the long term disposal criteria of HLW in oxidative conditions. In reducing geologic disposal conditions (e.g. clay), Tc-99 is stable as a Tc-TcO$_2$ compound with very low solubility. From environmental point of view only Kr-85 and I-129 are dispersed in the air and in the oceans. Kr-85 with 10.7 y half-life is not...
TABLE IV. CONCENTRATION OF LONG LIVED FISSION PRODUCTS IN Ci/t HM AT DIFFERENT COOLING TIMES FROM MOX FUEL IRRADIATED AT 40 GWD/t

<table>
<thead>
<tr>
<th>NUCLIDE</th>
<th>t½ (y)</th>
<th>7 y</th>
<th>50 y</th>
<th>100 y</th>
<th>500 y</th>
</tr>
</thead>
<tbody>
<tr>
<td>Se-79</td>
<td>10⁴</td>
<td>0.4</td>
<td>0.4</td>
<td>0.4</td>
<td>0.4</td>
</tr>
<tr>
<td>Sr-90</td>
<td>28.5</td>
<td>3.62 10⁴</td>
<td>1.27 10⁴</td>
<td>3.77 10³</td>
<td>0.29</td>
</tr>
<tr>
<td>Y-90</td>
<td>equil.</td>
<td>3.62 10⁴</td>
<td>1.27 10⁴</td>
<td>3.77 10³</td>
<td>0.29</td>
</tr>
<tr>
<td>Tc-99</td>
<td>2.1 10⁵</td>
<td>15.7</td>
<td>15.7</td>
<td>15.7</td>
<td>15.6</td>
</tr>
<tr>
<td>Zr-93</td>
<td>1.5 10⁶</td>
<td>1.5</td>
<td>1.5</td>
<td>1.5</td>
<td>1.5</td>
</tr>
<tr>
<td>Nb-93m</td>
<td>3.6 10⁷</td>
<td>1.42</td>
<td>1.42</td>
<td>1.42</td>
<td>1.42</td>
</tr>
<tr>
<td>Pd-107</td>
<td>6.5 10⁶</td>
<td>0.32</td>
<td>0.32</td>
<td>0.32</td>
<td>0.32</td>
</tr>
<tr>
<td>Sn-121</td>
<td>50</td>
<td>0.41</td>
<td>0.22</td>
<td>0.11</td>
<td>-</td>
</tr>
<tr>
<td>Sb-126</td>
<td>~ 10⁵</td>
<td>1.43</td>
<td>1.43</td>
<td>1.43</td>
<td>1.43</td>
</tr>
<tr>
<td>Sn-126</td>
<td>equil.</td>
<td>1.26</td>
<td>1.26</td>
<td>1.26</td>
<td>1.26</td>
</tr>
<tr>
<td>I-129</td>
<td>1.6 10⁷</td>
<td>0.05</td>
<td>0.05</td>
<td>0.05</td>
<td>0.05</td>
</tr>
<tr>
<td>Cs-135</td>
<td>2 10⁵</td>
<td>0.8</td>
<td>0.8</td>
<td>0.8</td>
<td>0.8</td>
</tr>
<tr>
<td>Cs-137</td>
<td>30.1</td>
<td>1.07 10⁵</td>
<td>3.97 10⁴</td>
<td>1.25 10⁴</td>
<td>1.21</td>
</tr>
<tr>
<td>Ba-137</td>
<td>equil.</td>
<td>1.01 10⁵</td>
<td>3.97 10⁴</td>
<td>1.25 10⁴</td>
<td>1.14</td>
</tr>
<tr>
<td>Sm-151</td>
<td>93</td>
<td>9.08 10²</td>
<td>6.54 10²</td>
<td>4.54 10²</td>
<td>20.45</td>
</tr>
<tr>
<td>TOTAL</td>
<td>4.06 10⁵</td>
<td>1.19 10⁵</td>
<td>3.25 10⁴</td>
<td>46.3</td>
<td></td>
</tr>
</tbody>
</table>

a long term threat but I-129 with its extreme long half-life is a critical nuclide on the very long term. If direct disposal is chosen as fuel cycle strategy the presence of I-129 may become the limiting factor of the repository.

Since Sm-151 is a member of the rare earth family and as its half-life is 93 years, it vanishes within the scheduled period of disposal (i.e. between 1000 and 10 000 years).

The case of C-14 deserves special attention though it is an activation product. During the dissolution of fuel it is discharged with the dissolver off gases but can be trapped if deemed necessary (BNFL). The concentrated $^{14}$CO$_2$ fraction can be stored as BaCO$_3$. The ultimate destination of such C-14 concentrate is the geologic repository.

4. LIMITS OF ACTINIDES' RECYCLE AND IMPACT ON DISPOSAL

Up to very recently the recycle of long lived actinides meant only: the recovery of MA from HLLW, the conditioning of MA for long term storage and the preparation and irradiation of MA targets in nuclear reactors or accelerator-driven transmutations. This way of thinking resulted from the underlying philosophy that after reprocessing the "valuable" elements, U and Pu were quantitatively recycled. In reality neither reprocessed uranium ($U_{REPRO}$) nor Pu are quantitatively recycled.
Whatever optimistic recycle scenario chosen large amounts of unused uranium will have to be stored or disposed of if there is no massive breakthrough of the fast reactor technology.

4.1. Residual uranium characteristics and their consequences

The radiological significance of residual uranium may not be neglected especially because large masses are involved. Table V summarizes the data.

A nuclear capacity of 100 GWe demands a uranium feedstock of 17 000 t/year which is transformed into 2200 t HM U 3.7%. The ore body containing the extracted uranium contains 46,750 Ci a which remains on the mining site in the tail ends. This activity decreases with the half-life of Ra-226 (1640 y).

TABLE V. COMPARATIVE RADIOLOGIC CHARACTERISTICS OF U PRODUCTS IN Ci/t HM

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>t ½(y)</th>
<th>Series</th>
<th>a/At</th>
<th>Nat. U</th>
<th>Depl. U</th>
<th>UO₂ U₉⁰⁰</th>
<th>MOX U₉⁰⁰</th>
</tr>
</thead>
<tbody>
<tr>
<td>U-232</td>
<td>73.6</td>
<td>4 n</td>
<td>7</td>
<td>-</td>
<td>-</td>
<td>0.0475</td>
<td>0.0337</td>
</tr>
<tr>
<td>U-234</td>
<td>2.74 10⁵</td>
<td>4 n+2</td>
<td>7</td>
<td>(0.3131)</td>
<td>-</td>
<td>0.0885</td>
<td>0.6264</td>
</tr>
<tr>
<td>U-235</td>
<td>7.1 10⁸</td>
<td>4 n+3</td>
<td>7</td>
<td>0.0154</td>
<td>0.0035</td>
<td>0.0166</td>
<td>0.0069</td>
</tr>
<tr>
<td>U-236</td>
<td>2.39 10⁷</td>
<td>4 n</td>
<td>8</td>
<td>-</td>
<td>-</td>
<td>0.311</td>
<td>0.0469</td>
</tr>
<tr>
<td>U-237</td>
<td>1.33 10⁻⁵</td>
<td>4 n+1</td>
<td>7</td>
<td>-</td>
<td>-</td>
<td>(2.58)</td>
<td>-</td>
</tr>
<tr>
<td>U-238</td>
<td>4.5 10⁻⁸</td>
<td>4 n+2</td>
<td>8</td>
<td>0.3303</td>
<td>0.3303</td>
<td>0.314</td>
<td>0.3048</td>
</tr>
<tr>
<td>TOTAL</td>
<td>0.3457</td>
<td>0.3356</td>
<td>-</td>
<td>0.77</td>
<td>1.0187</td>
<td>2.75</td>
<td>2.67</td>
</tr>
<tr>
<td>TOTAL (a) AT GEOLOGIC EQUILIBRIUM</td>
<td>2.75</td>
<td>2.67</td>
<td>6.05</td>
<td>7.556</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

About 14 800 t HM depleted U is accumulated per year and will reach its initial activity level over a very extended period of time. This uranium "residue" ought therefore to be considered as a strategic stockpile of uranium for a future fast reactor era.

The spent fuel-uranium mass (2200 t HM/100 GWe year) will contain 3.35 Ci/t HM after reprocessing and this a activity level will increase up to 6.05 Ci/t HM at secular equilibrium. This corresponds to 7370-13 300 Ci a for the total uranium inventory of the discharged spent UO₂ fuel. The a activity of U in UO₂ contained in MOX spent fuel is of the same order of magnitude.

Some minor actinides, e.g. Np-237 which are at 0.16 to 2.28 Ci/t HM spent fuel contribute only with 350 to 5000 Ci to the overall a activity. Such comparisons ought to be further investigated in order to establish the effective contribution of MA to the overall radiotoxicity in order to better understand the potential benefit of a given recycle scenario.

Recycling U from spent UO₂ fuel involves that, apart from reprocessing which is a necessity, a series of additional operations have to be carried out:
- fluorination of UO₂ to UF₄ and UF₆
- enrichment of U-235 from 0.8 to 3.7%
- conversion to U₃O₈ and UO₂.
Enrichment of U\textsubscript{REPRO} has to be carried out in a centrifuge-enrichment-plant in order to avoid permanent build up of Th-228 in the diffusion membranes of a diffusion plant. During enrichment the concentration of lighter isotopes (U-232, U-234) increases in the same proportion as U-235 (a factor of 7) and multiplies with the same factor the $\alpha$ activity of recycled and reenriched uranium. The ingrowth of Th-228 reaches 30% of the equilibrium value after 1 year, 50% after 1.9 year, 84% after 5 years and nearly equilibrium after 10 years.

The utilities should therefore be encouraged to recycle U\textsubscript{REPRO} quickly in order to avoid this buildup. However economic factors have led to the accumulation of large inventories of U\textsubscript{REPRO} in secular equilibrium with the Th-228 decay series.

In the present economic circumstances of very low uranium market prices, recycle of uranium has to be justified by other arguments, e.g. decrease of dose to the workers at the mining and milling facilities, or supported by waste minimization arguments or justified as a strategic resource preservation.

It is therefore not excluded that significant uranium inventories containing radiotoxic impurities will be considered as a waste stream rather than as a resource and will be conditioned for final disposal. In such a scenario the separation and recycle of Np becomes from radiologic point of view unnecessary.

4.2. Recycle and incineration of plutonium versus conventional disposal options

The first result of Pu recycle in LWRs is its "concentration" in a smaller number of fuel elements reducing the MOX elements to 1/5 of the initial UO\textsubscript{2} spent fuel elements. Secondly the total Pu quantity is reduced by 25% in a single recycle.

It is not excluded to consider a second LWR recycle but in that case "early" reprocessing has to be provided for, in order to avoid the loss of important Pu-241 quantities. However as explained above the reprocessing and fuel refabrication operations are in that case complicated by the presence of Cm-244 and Pu-238. Long term storage of spent MOX fuel (~50 years) with reprocessing and recycle of Pu and MA in a fast reactor park is probably the most realistic scenario which transfers the Pu+MA inventory from a storage facility to a reactor which produces at the same time electric power.

However the combustion also called "incineration" of Pu+MA is only a partial process. At the present time the burnup, i.e. the percentage of fission, varies from 12 to 15 at.% which means that at the end of the cycle the fuel still contains 85 to 88% of the initial actinides. A strategy of Pu+MA incineration involves obviously the gradual replacement of the thermal reactor park by fast reactors. Such replacement will take decades to be accomplished.

A typical 1000 MWe fast actinide burner reactor has an initial loading of 6 t Pu+AM and is operated over a 4 year cycle to reach burnups of 12-15%. The initial enrichment is 18 to 25% total Pu depending on the isotopic composition. The annual output coming from the LWRs is 22.5 t Pu which can be reduced to 15.68 t Pu if a single passage to LWR-MOX fuel is inserted.

The fast reactor park necessary to take up the large quantities resulting from a 100 GWe LWR park is of course very impressive. In case of UO\textsubscript{2} fuel cycle 16 GWe FN burner reactors and in the MOX case 12 GWe have to be constructed and operated to "use" the residual Pu+MA as a fuel. Each cycle takes 4 years to be accomplished followed by at least 7 years cooling and two years for reprocessing and fuel-refabrication. At the end of a 13 years cycle of that limited reactor park the initial Pu+MA load would be reduced to about 85% of the initial inventory. From then on a second strategic decision must be taken with
respect to the further elimination of the residual Pu+MA load by fissioning. In order to reduce the annual output to e.g. 10% of the initial LWR output, i.e. 2.5 t Pu+MA per year, an additional fast reactor park almost equal to the initial LWR-NPP park has to be constructed and operated during 14 cycles of each 13 years of the shrinking Pu+MA inventories. The required "incineration" capacity strongly depends on the attainable burnup. Table VI shows some typical values. The residual content of the cores 13-15% of the input have to be further fissioned or disposed of any way in a geologic repository.

### TABLE VI. ACTINIDE REDUCTION YIELDS

<table>
<thead>
<tr>
<th>Burnup%</th>
<th>Cycles</th>
<th>Years</th>
<th>FR capacity GWe</th>
<th>Pu+MA reactor</th>
<th>Reduction factor</th>
</tr>
</thead>
<tbody>
<tr>
<td>15</td>
<td>14</td>
<td>182</td>
<td>72.6</td>
<td>13.6%</td>
<td>7.35</td>
</tr>
<tr>
<td>20</td>
<td>10</td>
<td>130</td>
<td>52</td>
<td>13.7%</td>
<td>7.29</td>
</tr>
<tr>
<td>25</td>
<td>8</td>
<td>104</td>
<td>44</td>
<td>14.5%</td>
<td>6.9</td>
</tr>
<tr>
<td>30</td>
<td>6</td>
<td>78</td>
<td>36</td>
<td>15.8%</td>
<td>6.3</td>
</tr>
</tbody>
</table>

Unless a real fast reactor-breeder economy is going to replace the present LWR-UO\(_2\) era it does not look very attractive to consider such massive investments just to destroy Pu+MA with a factor of 7.

Accelerator driven transmutation is a potentially more interesting technology since it can burn actinides to a much higher burnup than achievable in a fast reactor. The bottle neck of increasing burnup lies with the cladding behaviour but in this field considerable progress has been made with the ferritic steels. If for example the burnup could be doubled to e.g. 25-30% the elimination time will be considerably shortened and the number of recycles significantly reduced to 5 or 6. It may take several decades to develop this technology from the conceptual design at present to a fully industrial plant in the first quarter of the next century.

However for the same throughput the investment is also doubled and the technological complexity of the accelerator-transmuter device may jeopardize the operability of such a plant on a year round, 24 h a day, basis.

### 4.3. Disposal option of MOX fuel

In case of direct disposal 2200 t HM spent LWR-UO\(_2\) fuel has to be disposed of every year. In case of a generalized use of LWR-MOX fuel the Pu inventory would be reduced to 75% and even to 56% in case of two consecutive recycles. But in this case the quantity of MA particularly Am and Cm will become much higher and will dominate the radiological impact during the first 4000 years. A 100 GWe NPP park with partial MOX loading produced as shown above about 440 t HM/year of spent fuel.

The disposal of Pu+MA rich fuel will necessitate a particular conditioning operation by which any danger of criticality is eliminated, e.g. by embedding the fuel elements in refractory type neutron absorbing materials (B\(_4\)C). The other precautions to be taken are nearly the same as for vitrified HLW.

However it must be emphasized that the decision to dispose of Pu+MA is more of an ethical nature rather than of technical or radiological nature. A long term retrievable surface storage of spent LWR-MOX fuel is therefore the most adequate decision to be taken within the current decade.
4.4. Disposal of waste

Conventional reprocessing of spent LWR-\( \text{UO}_2 \) fuel produces apart from U and Pu a HLLW solution which contains all the fission products and the most important MA. Presently this HLLW is stored for a few years and then vitrified. The glass canisters are then stored for \( \sim 50 \) years till the heat dissipation has decreased to levels acceptable for underground disposal.

As shown in Table I the net volume of HLW to be stored amounts to 330 m\(^3\)/year in case of a LWR-\( \text{UO}_2 \) scenario and 250 m\(^3\) in case of a LWR-MOX scenario. Table IV shows the specific activity of each of the important radionuclides, and Table III gives an overview of the MA concentrations in spent fuel. The data shown for the MA are also valid for HLW except the Am-241 levels which are lower in case of reprocessing. The Am-241 levels in HLW after reprocessing of \( \text{UO}_2 \) or MOX fuel are for completeness given below.

<table>
<thead>
<tr>
<th>( \text{t + 7 y} )</th>
<th>( \text{t = 100 y} )</th>
<th>( \text{t = 500 y} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \text{UO}_2 )</td>
<td>( \text{MOX} )</td>
<td>( \text{UO}_2 )</td>
</tr>
<tr>
<td>Am-241 Cl/t HM</td>
<td>1500</td>
<td>1290</td>
</tr>
<tr>
<td>Am-241 g/t HM</td>
<td>438</td>
<td>377</td>
</tr>
</tbody>
</table>

In earlier publications it has clearly been shown that Cs-137, Sr-90, Am-241 and Cm-244 dominate the radiotoxic potential during the first 200 years. Gradually the radiotoxicity of the FP and Cm-244 decreases and beyond 500 years Am-241 takes the lead and continues to be the most important \( \alpha \) nuclide in HLW till its extinction after 4000 years.

On the very long term only Tc-99, Zr-93, Sb-126 and Cs-135 remain. Since Tc-99 is to be separated from other nuclides during reprocessing, transmutation is technically feasible but it is a very slow process due to the tiny absorption cross section. Moreover its impact on the biosphere is very limited if the repository host rock has reducing properties as it is the case for clay deposits and underground aquifers. The partitioning of Zr-93, Sb-126 and Cs-135 is very difficult and not cost effective since isotopic separations ought to be done. Their contribution to the overall radiotoxicity as yet is very low.

Only by separating Am-241-243 from HLW its radiotoxicity can be significantly reduced. But in the most optimistic partitioning-transmutation scenario known up to now this complex process will only partially destroy the Am-241-243 and transform it into Pu-238 and higher Am, Cm isotopes. This P&T scenario will reduce the period during which high level waste is very radiotoxic from 4000 y to about 800 years and this is a time horizon which can easily be overlooked by man and its technology. The cost-benefit analysis of such a proposal has to be thoroughly made on the basis of a systematic investigation of the processes involved and the expected environmental impact reduction.

Alternatively the separated actinides from HLW could be stored till the decision is taken whether or not the MOX fuel which contains the large majority of the actinides produced, is reprocessed. This outcome will primarily depend on the availability of fast reactors or accelerator-driven-transmutation devices during the first decennia of the 21st century. In the mean time vitrification of HLW and eventual disposal in a suitable geologic repository is the most obvious practice to be continued till more R&D results are available on the newly proposed partitioning-transmutation technologies.
5. CONCLUSIONS

P&T is scientifically a very challenging fuel cycle option, but it is not a short term alternative to HLW disposal. In the long term P&T holds promises to reduce, but not to eliminate, the potential hazard associated with all the actinides.

However the concurrent production of additional power from these processes is a condition sine qua non for a favourable outcome. This scenario implies the deployment of a very large fast reactor park or of large accelerator driven transmutation devices.
ACTINIDE RECYCLING IN FAST AND THERMAL REACTORS

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Abstract

The possibilities to reduce the long-term toxicity of nuclear waste from a Light Water Reactor fuel, are evaluated for some options of actinide recycling in fast reactors. A possible first recycling in the light water reactor itself is analyzed with regard to implications on reactor physics and core management. Problems of radioprotection associated to the addition of the various minor actinides at the fabrication of mixed oxide fuel are identified and solutions are considered. Most of the results of this paper were presented at the recent GLOBAL'93 meeting in Seattle [6].

1. INTRODUCTION

The Uranium fuel used for energy production in nuclear reactors is partly transformed into fission products and other actinides. The Plutonium can be reused for further energy production but the minor actinides (M.A.) : Americium, Neptunium, Curium are nowadays accompanying the fission products into waste from reprocessing the irradiated fuel and extracting Plutonium.

One important objective for the future is the reduction of the amount of waste to be stored and of their potential radiotoxicity. In that respect, the recycling of the M.A. in power reactors needs still research and development.

During their irradiation in reactors, the M.A. are partly transmuted into other radioactive products with shorter decay half-lifes and/or less potential radiotoxicity. The general problem has been dealt with in several publications, like Reference 1 ; this paper focuses on feasibility aspects.

The major objective of this paper is to review the differential impact of the M.A. recycling in power reactors according to several options, taking account of the influence of reprocessing parameters. A second objective is to identify the radioprotection problems in a reference mixed oxide (MOX) fuel fabrication plant. Possible limitations on the amount of the actinides to be recycled, from the neutron physic point-of-view, are mentioned.

The main criterion of comparison is based on the evolution of the potential radiotoxicity of waste over several thousands of years.

The potential radiotoxicity of an isotope is defined here as the ratio between its radioactivity and the annual limit on intake (A.L.I.) by ingestion for workers. The ALI values correspond to the latest ICRP recommendations (ICRP 61-1990).

The total radiotoxicity of waste is related to a reference value corresponding to the natural Uranium ore quantity needed to feed the nuclear reactor and the recycle chain.

The time evolution of the radioactivities are obtained here from a detailed simulation of the power production cycles, the cooling periods before reprocessing and fuel refabrication, several recyclings and the long-term storage of waste. The ORIGEN-2 code (Ref. 4) is used with its 1990 libraries of constants. Specific computer codes are used for the radioprotection analysis and the neutron physics study.
2. INITIAL FUEL IRRADIATION IN LIGHT WATER REACTORS

2.1. According to the core management option, the U235 enriched UO2 fuel remains in the reactor core along three or four annual power cycles, during which actinides and fission products are built.

Table I refers to the basic option of a 3.25 % U235 fuel with a 33,000 MWD/TU average burn-up in a 950 MWe PWR; it presents first the evolution of the potential radio-toxicity of actinides produced from 1 ton of enriched Uranium initially loaded in the reactor, and directly stored as waste during one million of years. However, some years (e.g. seven) after the end of irradiation, reprocessing takes place and the actinides are separated from the waste with an efficiency taken as a parameter: for Uranium and Plutonium, 99 % is already obtained today and 99.9 % is a near future objective; for minor actinides, 99.5 % is assumed to be achievable in the future.

### TABLE I: Relative Potential Radiotoxicity of Actinides in Waste from PWR Irradiated Fuel (Reference: 1 for U ore)

<table>
<thead>
<tr>
<th>Time (Years)</th>
<th>10</th>
<th>100</th>
<th>10³</th>
<th>10⁴</th>
<th>10⁵</th>
<th>10⁶</th>
</tr>
</thead>
<tbody>
<tr>
<td>Direct storage</td>
<td>1039</td>
<td>899</td>
<td>257</td>
<td>56</td>
<td>3</td>
<td>0.52</td>
</tr>
<tr>
<td>0.999 U/Pu separated</td>
<td>315</td>
<td>203</td>
<td>50</td>
<td>1,83</td>
<td>0.19</td>
<td>0.15</td>
</tr>
<tr>
<td>0.999 U/Pu + 0.995 Am separated</td>
<td>87</td>
<td>5</td>
<td>1</td>
<td>0.32</td>
<td>0.07</td>
<td>0.08</td>
</tr>
</tbody>
</table>

If U, Pu and Am can be separated with such a good efficiency, the remaining actinide radiotoxicity in waste is strongly reduced; the radiotoxicity reference level (=1) corresponding to the needed Uranium ore is reached after about 1,000 years, while for the directly stored waste, this would be after more than 100,000 years. The separated actinides are assumed to be recycled in mixed oxide fuels of power reactors.

2.2. The analysis of the contributions of each individual actinide element present at the irradiation end, in case of the unreprocessed fuel points out some priorities for reprocessing.

Figure 1 gives the time evolution of each contribution along a million of years. At medium-term (100 to 10,000 years), the most important actinides are Plutonium and Americium. Reprocessing improvements should focus on both. Curium has only a second order importance and Neptunium plays a role at very long term only (as Uranium also). The relative contributions of each actinide family after 1 million years are 44 % for Pu, 27 % for U, 15 % for Np, 13 % for Am, 0.03 % for Cm.

3. ACTINIDE RECYCLING IN FAST REACTORS

The present results refer to a detailed explicit study of successive recyclings in LMFR cores over about one century. Starting from an initial
Relative toxicity

Storage years

**FIG. 1. Contribution to actinide radiotoxicity from 1 t U PWR fuel (33 000 MWd).**

\[ \text{Relative toxicity} \]

<table>
<thead>
<tr>
<th>Relative Toxicity</th>
<th>Storage Years</th>
</tr>
</thead>
<tbody>
<tr>
<td>1000</td>
<td>1</td>
</tr>
<tr>
<td>100</td>
<td>10</td>
</tr>
<tr>
<td>10</td>
<td>100</td>
</tr>
<tr>
<td>1</td>
<td>1000</td>
</tr>
<tr>
<td>0.1</td>
<td>10000</td>
</tr>
<tr>
<td>0.01</td>
<td>100000</td>
</tr>
<tr>
<td>0.001</td>
<td>1000000</td>
</tr>
<tr>
<td>0.0001</td>
<td>10000000</td>
</tr>
</tbody>
</table>

\[ \text{Storage years} \]

UO₂ fuel irradiated in PWR with a 43 GWD/t average burn-up in four years, the Plutonium and M.A. are separated at reprocessing and recycled successively seven times in LMFR cores of an EFR-type reactor (1,450 MWe). Cooling periods between irradiation, reprocessing, refabrication and reloading are taken into account.

Figure 2 shows the evolution of the waste toxicity for different cases:

. Direct storage of irradiated LWR fuel (43 GWD/t) as a reference,
. Successive homogeneous recyclings in LMFR:
  a) 99.5% Pu only;
  b) 99.5% (Pu + M.A.);
  c) 99.9% Pu + 99.5% M.A.

For the comparison of recycling schemes, due consideration is given to the total electrical energy production associated with the recycled actinides, issued from 1 ton of the initial enriched Uranium, extracted from 7.67 tons of U ore.

A previous paper (Ref. 2) presented detailed results of some recycling schemes. As a summary, the major contributors to the medium-term and long-term actinide waste toxicity are Am241 between 100 and 2,000 years, Pu239 and Pu240 between 2,000 and 30,000 years, Np237 (and Th229) beyond 100,000 years. (Fission products dominate first the total waste toxicity, but after 200 years, their contribution becomes inferior to the non-recycled actinide fraction toxicity).
FIG. 2. Radiotoxicity of waste related to uranium ore.

The most efficient recycling (99.9 % Pu + 99.5 % M.A.) leads to a toxicity reduction factor of about 250 to 300 between 1,000 and 100,000 years in comparison with the direct storage of PWR irradiated fuel. The toxicity level of U ore is recovered after about 1,200 years, while in case of recycling only 99.5 % Pu, the time span would be about 50,000 years.

However, if the sequence of recycling were stopped after 100 years, adding the last core inventory to the waste, the reduction of the M.A. amount in waste would not be so significant (for the Np mass, a factor 14 instead of 90 and for the Am241 a factor 3 instead of 60; there would be no reduction for Curium, but this has no important effect).

On the other hand, the plutonium reduction efficiency could be significantly better if the LMFR core operates without blankets as a Pu-burner. The core considered here was supposed to be just self-sustaining in Pu thanks to the presence of blankets.

Moreover, the reduction of M.A. masses could be increased if the mixed oxide fuel were doped with more actinide than it is produced and recycled from the previous core of the sequence. As an example, while the "natural" fraction of Np recycled from PWR to LMFR fuel is about 5 % of the Pu (i.e. 1 % of the total mass of heavy metal (U, Pu) in the mixed oxide), a "doping" up to 3 % Np of the heavy metal loaded in LMFR could be allowed in order to accelerate the Np reduction. The limitation to 3 % is linked to the production of Pu238 in LMFR core, which should be kept below 5 % of the total discharged Pu according to present practices (Ref. 3).
4. ACTINIDE FIRST RECYCLING IN WATER REACTORS

Before recycling in LMF, the M.A. could first be recycled in LWR as it is already done for Plutonium in mixed oxide fuel. Physics studies are in progress to identify problems and come to recommendations. Some first results are presented hereafter for Np237 and Am241 in a PWR with average discharge burn-up of 45,000 MWd/t.

4.1. In case of homogeneous recycling in UO2, all oxide fuel elements would contain a small fraction of M.A. For the MOX core, it is assumed that only 25% of assemblies include Pu and a small amount of M.A. Criticality and burn-up conditions are analyzed using the reactor physics computer code LWR-WIMS with a 69 energy groups library of cross sections. Due to the neutron absorption effect of M.A., the U235 or Pu enrichment must be increased in order to keep the same reactor cycle length (The criterion is to keep the same multiplication factor - k infinite value - at end of cycle as in the standard PWR core without boron).

The enrichment needs in UO2 or MOX fuel are given in the Table II, as well as the burnt fraction of the added M.A. and the specific Pu production or disparition related to the standard core.

<table>
<thead>
<tr>
<th>TABLE II : Features of an Homogeneous Recycling in PWR Fuel</th>
</tr>
</thead>
<tbody>
<tr>
<td>Added M.A. (%)</td>
</tr>
<tr>
<td>----------------</td>
</tr>
<tr>
<td><strong>UO2 fuel</strong></td>
</tr>
<tr>
<td>0 (ref.)</td>
</tr>
<tr>
<td>1 (Np02)</td>
</tr>
<tr>
<td>2 (Np02)</td>
</tr>
<tr>
<td>1 (Am02)</td>
</tr>
<tr>
<td><strong>MOX fuel</strong></td>
</tr>
<tr>
<td>0</td>
</tr>
<tr>
<td>1 (Np02)</td>
</tr>
<tr>
<td>1 (Am02)</td>
</tr>
</tbody>
</table>

The overenrichments are seen as a major penalty.

The initial quantities of Np or Am are strongly reduced, but they produce Pu, mostly Pu238 which is a source of neutrons and residual heat, which could hinder further steps of the recycling scheme.

4.2. Another solution would be the heterogeneous recycling in target rods located in the 24 guide tubes of 25% of standard (17 X 17) assemblies in the PWR core.

The fuel of these rods could be made of 20% NpO2 and 80% Al2O3. The fuel enrichment in the standard rods should not be increased, but the poisoning effect of M.A. on the reactivity evolution implies that either the target rods should be removed before the end-of-cycle or the cycle length should
be reduced (in this case, with 24 target rods in 25% of the core assemblies, the loss of energy production could reach 15%).

The reduction of the Neptunium mass in this recycling mode in UO$_2$ fuel cores is about 45%, a figure very close to those presented for the homogeneous recycling. Due to the conversion into Pu238, only 10% of the initial Np does really disappear from the actinide family into fission products.

The recycling in MOX fuel assemblies is slightly less efficient.

5. IMPACT OF RECYCLING ON FUEL FABRICATION

The impact is analyzed for a MOX fuel factory similar to the BELGONUCLEAIRE one at Dessel (Ref. 5), in case of an homogeneous recycling of minor actinides mixed with the PuO$_2$ primary powders, either in LWR fuel or in LMFR fuel.

The individual effects of either Np or Am or Cm addition to the MOX fuel were evaluated with a three year time interval between re-processing and refabrication.

Problems of handling are created by the emissions of neutrons and photons from the powders to be mixed. With respect to the pure PuO$_2$ reference case:

- Neptunium recycling does not change the neutron source but increases somewhat the gamma source (due to the daughter Pa233); after recycling in a LMFR, a further small contribution to high energy gamma rays is due to the chain of Pu236-U232-Tl208.

- Americium recycling increases both the neutron source and especially the photon source; these effects depend very much on the "age" of the Am-Pu mixture. Besides the Am isotopes themselves, small contributions to both sources come from Np239 and Cm242 produced by the decay of Am242 and Am243.

- Curium recycling would lead to a factor 100 increase of the neutron source; this would require so thick protection layers that it practically excludes the operation in present fuel fabrication plants.

Two steps at the front-end of the fabrication process are particularly critical from the shielding point-of-view; they are:

a) the storage of actinide oxide powders (in steel boxes located in cavities with concrete walls and a shielded door),

b) the glove box where the primary blending with UO$_2$ is achieved.

Total equivalent dose rates have been calculated at a short distance from either the cavity door or the blending glove box, to find out the additional shielding required to obtain the same conditions as in the reference case.

Table III gives results for the storage of oxide powders.

For the Neptunium case, the shielding problems are minor ones at the first fuel refabrication for recycling; after the first recycle in LMFR, the total dose rate increase by a factor 1.25 would require 4 mm of lead (or equivalent) as additional protection.
TABLE III
Impact of Actinide Recycling
at Oxide Powder Storage, Front End
of MOX Fabrication.
(Initial Shield = 6mm. Lead, 110 mm. Polythen)

<table>
<thead>
<tr>
<th>Recycling from PWR to FR</th>
<th>Dose Rate (µSv/h)</th>
<th>Additional Shield Needed (mm.)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Gammas</td>
<td>Neutrons</td>
</tr>
<tr>
<td>0.995 Pu (Ref)</td>
<td>18</td>
<td>13</td>
</tr>
<tr>
<td>Ref + 0.8 Np</td>
<td>26</td>
<td>13</td>
</tr>
<tr>
<td>Ref + 0.8 Am</td>
<td>89</td>
<td>14</td>
</tr>
<tr>
<td>Ref + 0.8 Cm</td>
<td>240</td>
<td>460</td>
</tr>
</tbody>
</table>

For Americium recycling fuel 3 years after reprocessing, the increase of the total dose rate by a factor 3.25 must be compensated by an additional 20 mm lead thickness in the shield of the cavity door; on the primary blending drum, an increased steel protection of about 20 mm should be foreseen also. Such additions would be costly and cumbersome, but they are still feasible.

Complementary protection measures should also be implemented to limit the radiation doses for the personnel; they would imply remote handling and a revised concept of some equipment.

6. CONCLUSIONS

- The studies show that an efficient waste toxicity reduction at long term (1,000 years and over) by a factor higher than 100, can be obtained by the multiple recycling of minor actinides in fast reactors. This effect can be enhanced if the LMFR cores are designed as Plutonium burners. The reduction of the mass of waste due to actinide recycling is also considered as an advantage.

- The radiotoxicity level of the waste can be lowered to that one of the initial Uranium ore after about 1200 years. The prerequisite is a very efficient recovery of Pu and of Am at reprocessing, which should be the target of R & D work.

- Plutonium obviously, Americium in priority and possibly Neptunium are the most important actinides to be recycled; Curium extraction has a low priority because it has only a weak contribution to the long term toxicity and, on the other hand, it would induce difficult handling problems at the fuel fabrication plant.

- The impact of Neptunium addition on fuel fabrication in a MOX fuel factory is small; recycling Americium needs more shielding, but this appears to be still feasible.

- Actinides from an irradiated LWR fuel could be recycled first in LWR itself; later on, both the Pu and minor actinides would be recycled in LMFR. Such a strategy allows to start sooner the process of waste toxicity reduction, but fuel overenrichment in LWR with minor actinides is needed, or the power cycle length could be reduced.
REFERENCES


Abstract

At present, the question to be answered is what to do with the spent nuclear fuel in order to be able to prepare locations for its final disposal in the next century. For this reason, not only the feasibility studies are needed concerning the technical possibilities of destroying what is called now "nuclear wastes". Financial aspects of such programs and their time horizon are to be assessed and confronted with estimates of the uranium market development. Doing this, public opinion and general political considerations should be taken into account.

Historical development of mankind is seen by an expert by his own eyes and through the mediation of them he interprets historical convulsions, changes of systems and frontiers, and general growth of living standard.

Let us try to incorporate the development of technique of transmutation and fission of heavy nuclei into a wider context of the development of nuclear power and let us strive for finding the answer to the question if, and under what conditions, this technique can give already now a concrete recommendation for practical plans.

At present most countries operating nuclear power plants are making decision on the way of processing and disposal of the spent nuclear fuel. Considerable role in this issue is played also by general public who do not want to accept the idea of storage facilities aimed at isolation of the spent fuel for tens of thousands of years. At the same time, also majority of politicians and personnel of central authorities are not aware that the spent nuclear fuel is composed of two physically different components with diametrically differing requirements with regard to their disposal:

(1) elements, originated in fission of uranium and other heavier nuclei;
(2) elements, originating in capture of neutrons on uranium nuclei.

From the point of view of storage, elements of the first group are characterized by decay half-lives under 30 years and, generally, it is possible to say that after ten half-life periods their radioactivity will be practically zero. Horizon of three hundred years is technologically implementable without any difficulties even for general public.

It is evident for experts that the whole second group of elements with extremely long decay half-lives and not too well known chemical effects (virulence of Pu) is in fact a very high-grade nuclear fuel the utilization of which in MOX fuel elements has already started. We regard the specificity of the technique of transmutation in real possibility how to use the transuranium elements entirely, that is, the transuranium would be only supplied into the complex composed by an accelerator and reactor (MOX fuel after burn up still contains significant amounts of plutonium and requires special conditions for further storage and processing).

In this way we are already now getting successively to the formulation of the practical aspect of these new technologies. It is quite obvious for experts in the field of accelerators and nuclear reactors that practical realization of transmutations is without
principal difficulties - it is mainly a matter of financial funds and time. If we know with sufficient credibility the answer to the question "when and for what money", this technology will highly probably manifest itself as competitive in comparison with the final disposal of the spent fuel. At present the governments are making decision on the future strategy and they need deeper information and not only an explanation of the basic physical principle.

Standard technique of management of spent fuel is as follows:

(a) storage for 3-5 years in wet storage facilities in the NPPs;
(b1) subsequent storage in dry storage facilities or containers for approximately 50 years;
(b2) reprocessing and partial utilization;
(c) final disposal or further utilization (or extended medium-time storage of waste).

The knowledge whether the transmutation will be practically applicable on large scale in the way which is competitive to the final disposal or extended interim storage in dry storage facilities has great practical value.

What is necessary to take into consideration in this?

Development of prices of nuclear fuel

It is possible to expect that, at the beginning of the next century, the reserves of military fissile materials will be used and the price of the raw material will rise sharply.

Development of the technology of transmutation

If we have a look back by fifty years, only a limited group of experts knew what is military utilization of fission of nucleus and only a negligible percentage of them dreamed on peaceful nuclear power. A futuristic idea of industrial implementation can be formed on numerous analogies of this development and similar innovative trends in other areas.

Increasing claims to preservation of environment will lead to gradual increase of costs of the final disposal, which on territories of small countries is practically impossible to implement.

Irrespective of the all local conflicts and separation of the world into wealthy and poor ones, present science and technology lack boundaries and world is gradually politically united. This development will certainly lead also to a more uniform atomic legislation in such a way, that nobody feels endangered and everybody can fully utilize the development of science and technology.

We think that it is inevitable in principle to accept the opinion of general public that it is necessary to demonstrate the possibility to resolve the problem of waste of every new technology by present technological means. It appears to us, that the disposal of spent fuel for tens of thousands of years is something, which is very difficult to explain to general public, since application of such horizons take us backward to the stone age - it is a time scale, for which no analogy exists. In the relation to the general public we require as a principal task a change of the notion of spent nuclear fuel as a waste into a notion of the spent fuel as a rough material for the next century. Opinion of the public and higher political spheres will affect the supply of financial means for technological development.

The purpose of this very general analysis is to initiate an activity aimed at expert assessment of development of the technology of transmutation and preparation of a basis for decision on long-term strategy in the field of spent nuclear fuel management. We
believe that elaboration of a study of this type would be supported by the International
Agency for Atomic Energy in Vienna, and Czech experts are prepared to participate in this
activity. We also believe that elaboration of the expert estimation of development of
practical application of transmutations would also in a significant way support this
technology, gain financial means and accelerate the development.

There is a zero-power experimental reactor for investigation of various arrangements
of power reactor cores and also other nuclear fuel systems (e.g., spent nuclear fuel storage
facilities) operated in the Nuclear Research Institute, Rez near Prague, since 1972. In the
first stage up to 1982, the reactor was designed and operated as a heavy water reactor
TR-0 with metallic natural uranium fuel assemblies (functional full-scale model of the A-1
nuclear power plant reactor core). Since 1983, the reactor is operated as a LR-0 reactor
for research of the WWER-440 and WWER-1000 reactor cores (light-water system with
slightly enriched UO\textsubscript{2} fuel assemblies, identical in the radial section with the WWER-type
power reactor fuel assemblies and, in the axial direction, reduced to approximately 1/2 and
1/3 of the real height of the WWER-440 and WWER-1000 reactor fuel assemblies,
respectively).

The experimental reactor is designed as a very flexible facility enabling research on
neutronic characteristics and time behavior of various arrangements of fuel systems and
their following effects as, e.g., determination of neutron spectrum and more accurate
determination of reactor radiation doses on the reactor vessel inner wall (e.g., models of
all layers of structural internals between reactor core and pressure vessel body are
installed). Backward reconstruction into the original heavy water system of the TR-0
reactor or other needed critical or subcritical systems of nuclear fuel and other components
is ensured in principle.

Such measurements on such special experimental devices will be surely necessary
in future to model reactor part of a transmutation facility and check nuclear parameters of
the rare transuranium elements which should be transmuted. Czech specialists and their
small experimental possibilities are ready to take part in a coordinated effort of countries
trying to bring this problem to an industrial realization.
OVERVIEW OF THE FRENCH PROGRAM IN CHEMICAL SEPARATION AND TRANSMUTATION OF MINOR ACTINIDES AND LONG LIVED FISSION PRODUCTS

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Abstract

The separation of long-lived elements and their transmutation is one of the goals stated in the December 30, 1991 law. With this in mind, the CEA has set up the SPIN program (Separation - Incineration). The separation of actinides through diamides is considered. The basic studies have resulted in the choice of dimethyl dibutyl tetradecyl methyl amide (DMDBTDMA) and in the development of the AMIDEX process. Macrocycles are contemplated to separate the other long-lived fission products. The separated actinides will be transmuted from homogeneous fuels or heterogeneous targets in PWR or FR reactors. Data show that, compared to current reprocessing, a transmutation in FR is likely to reduce the radiotoxicity by a factor of about 30, with a set of FBRs representing 20 or 30% of all the reactors depending on whether recycling is of the homogeneous or heterogeneous type. The Superfact experiment has shown good behavior of the actinide-loaded targets. Transmutation of the fission products could be obtained by means of targets placed in an outer area of the FR core with thermalized neutrons. Special attention is devoted to the use of accelerators.

INTRODUCTION

The concern for long-lived element management is taken into account by the December 30, 1991 law on wastes which, among the three points concerning research programs, states "the search for solutions allowing separation and transmutation of long-lived radioactive waste present in these wastes".

The laws stipulates that every year a report on the results obtained and the evolution of the programme must be presented by the French Government to the Parliament. The report will be presented by the "Commission Nationale d'Evaluation" an assembly of will known and independent experts designated by organismes (Academy of Sciences).

To meet these requirements, the CEA has set up a program called SPIN which consists of two aspects: separation-processing and transmutation-incineration.

SPIN includes two sub-programs:

PURETEX, carried out in close cooperation with Cogema, aims at improving the performances of the La Hague plants in order to decrease the volume of liquid waste released and waste produced and to reduce the activity of B wastes to make them surface-disposable insofar as possible.

Table I shows the objectives to be reached, i.e. the limitation of the volume of wastes produced per ton of reprocessed spent fuel to 0.5 m$^3$ instead of the 1.5 today. This is to be compared with 1.7 m$^3$ in geological direct disposal.

ACTINEX, aimed at performing very selective and specific separations of long-lived elements to ensure their destruction by incineration-transmutation and, possibly for some of them, their incorporation into very high performance matrices.
TABLE I. VOLUMES OF WASTES PRODUCED IN REPROCESSING AT UP3 (litre per tonne of heavy metal reprocessed)

<table>
<thead>
<tr>
<th>Embedding Medium</th>
<th>Nominal Value</th>
<th>Obtained in 1991 *</th>
<th>Expected IN 1995</th>
<th>Expected IN 2000</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fission products</td>
<td>glass</td>
<td>130</td>
<td>115</td>
<td>115</td>
</tr>
<tr>
<td>Hulls and dead-ends</td>
<td>grout</td>
<td>600</td>
<td>&lt;600</td>
<td>150</td>
</tr>
<tr>
<td>Sludges</td>
<td>bitumen</td>
<td>630</td>
<td>450</td>
<td>0</td>
</tr>
<tr>
<td>Technological wastes (deep disposal)</td>
<td>cement block CAC</td>
<td>1700</td>
<td>200</td>
<td>200</td>
</tr>
<tr>
<td>Aggregate value</td>
<td></td>
<td>3060</td>
<td>1365</td>
<td>&lt;915</td>
</tr>
<tr>
<td>Technological wastes (shallow land burial)</td>
<td>cement block CBF (fiber reinforced concrete)</td>
<td>3800</td>
<td>1400</td>
<td>1400</td>
</tr>
</tbody>
</table>

*First year of production

PURETEX is a short and medium term program, ACTINEX is a long term program (Fig.1).

**PURETEX**
- ENHANCED Pu SEPARATION
- Np SEPARATION
- REDUCTION IN THE VOLUME AND ACTIVITY OF "B" WASTES EXISTING PLANTS (LA HAGUE)

**ACTINEX**
- ENHANCED SEPARATION:
  - Pu
  - Np, Am, Cm
  - FP WITH LONG HALF-LIFE
- ENHANCED REDUCTION IN THE ACTIVITY OF "B" WASTES NEW FACILITIES
- TRANSMUTATION
  - REACTORS
  - ACCELERATORS

**FIG 1** Strategies for the back-end of the fuel cycle R&D program proposals
THE P&T CONCEPT

Criteria and radionuclides to betaken into account

The goal of the P&T strategy can be expressed in the most general way by a reduction of waste radiotoxicity with time. This reduction may be appreciated according to two fundamental criteria: "potential radiotoxicity" referring to the radioactivity existing globally in the waste disposed at a given time or "residual radiotoxicity" referring to the radionuclides reaching the outlets of a waste disposal site.

These two approaches to the evaluation of the advantages of the P&T strategy contradict each other from the ethical point of view. Indeed, "potential radiotoxicity" is evaluated without taking the geological barrier into account, whereas "residual radiotoxicity" does. Time delays caused by the geological barrier differ widely with the radionuclide concerned and will therefore radically modify their relative importance. Similarly, the absolute values of radiotoxicity reaching the outlets, estimated to be extremely low, will considerably reduce the interest of the P&T strategy.

At the present stage, it is possible to consider that the first approach, based on "potential radiotoxicity" is the only one that would enable building a valid research program. Indeed, the second approach, which is more technical, requires that the performances of the disposal site used be known and, although the results of exercises such as PAGIS have shown great consistency among all the disposal media, these results constitute only a preliminary approach that will have to be completed in coming years. In particular, it will be necessary to consider long term prospects (beyond the 10 000 years for which geologists agree to make accurate predictions) site by site according to the regional locations.

The radionuclides to be taken into account in the "potential radiotoxicity"are mainly the minor actinides.

The radionuclides to take into account in priority for "residual radiotoxicity" are those with the greatest mobility in the disposal medium. This includes the chemical elements that can take on anionic forms or alkaline cationic elements. Depending on the site, the hierarchy will be different for these elements which are essentially long-lived fission products: I-129, Tc-99, Cs-135, Se-79.

When examining the potential toxicity of wastes, i.e. the toxicity that takes into account all the radionuclides contained in the wastes, considered as immediately ingestible, without taking into account the quality of the packaging and what is retained by the storage site, the paramount role of the actinides is seen. Consequently, the actinides must be separated.

Figure 2 shows the evolution in radiotoxicity according to 4 separation scenarios:

- no separation: i.e. disposal of the fuel as is,
- reprocessing with today's separation of the uranium and of 99.7% of the Pu with disposal of glass,
- storage of a glass corresponding to an additional separation of the minor actinides (factor 10),
- storage of a glass corresponding to an additional separation of the minor actinides (factor 100).

Comparison of the curves shows that the preponderant isotopes vary with time: americium-241, from 300 to 1000 years, then residual plutonium (10 000 to 100 000 years) and lastly, neptunium. The fission products present a potential toxicity about 10 000 times lower than that of the actinides.
The level of reduction of radiotoxicity to be obtained is presently subject to discussion.

---

**Strategy**

A purely conceptual study has been launched, under a CEE contract to analyse the potentials of a strategy of radioactive waste management aimed at reducing the quantity of long lived radionuclides by partition and transmutation.

The study is taking into account different scenarios for which the evolution of potential radiotoxicity of stockpile are computed and compared.

Are compared several recycling options either homogeneous or heterogeneous in PWR or/and in FBR with different levels of contents. The mains points are: influence of minor actinides on the physical parameters of the core transmutation yields, expected decrease of minor actinides masses and potential radiotoxicities.

Finally different separations techniques are envisaged, with evaluation of yields of recovery, losses.

**CHEMICAL SEPARATION**

Today's reprocessing plants, UP1 in Marcoule, UP2 800 and UP3 in La Hague, use liquid-liquid processes to recover the plutonium and uranium. Studies on the separation of actinides will therefore rely on such techniques since they will be easy to retrofit into current technologies. Moreover, the knowledge acquired on the behavior of actinides in aqueous mediums from the studies carried out in CEA for the preparation of actinide-based sources, further reinforce this orientation.
Separation of neptunium during reprocessing

In the PUREX process, the solvent selectively extracts, from fuel dissolution solutions, uranium at valence (VI) and plutonium at valence (IV). Practically all the fission products and minor actinides (Am and Cm) as well as a part of the neptunium are left in the mother solution. As a second step, the plutonium is separated from the uranium, after reducing it from the extractible valence (IV) to the non-extractible valence (III).

Neptunium is present under two valencies in the dissolution solutions:

- about 20 to 30% at valence (V), non-extractible, remains in the solution of the fission products to be vitrified,
- about 70-80% at valence (VI) is coextracted with Pu and U. First it follows the uranium and then goes into the raffinates of the 2nd uranium cycle which are currently mixed with the fission products solution.

It can be seen that if this raffinate is no longer sent to glassification, 70 to 80% of the Np is immediately recovered. It is, however, possible to go further by making extractible the 20-30% of the neptunium which is not, namely by oxidizing it at the state of Np (VI).

The whole problem lies in the selection of the oxidant that should oxidize Np (V) and Np (VI) without making Pu (IV) go to the hardly extractible Pu (VI).

At the present time, modification of acidity and temperature may offer the possibility to oxidise a large part of Np V, if not sufficient addition of vanadium salts appear to offer a possibility based on different oxidation kinetics between Np and Pu. This possibility is tricky but it would hardly modify the present facility.

Extraction using fission product solutions

In the present process, most of the minor actinides not extracted are mixed with the fission products in the aqueous solution resulting from the first stage of reprocessing.

These solutions of fission product concentrates are produced taking the example of reprocessing a ton of PWR fuel (initial enrichment 3.5%, 33 GW/t, 3-year cooling) at the rate of 300 litres of concentrated solution, from 5000 litres of raffinate containing 20 g of non-separated plutonium, about 1 kg of minor actinides mixed with 30 kg of FP with an activity of 2400 Ci/l.

While neptunium, uranium and plutonium have several valence states that are relatively stable in a nitric medium, americium and curium are similar to the lanthanides in that their valence (III) is very stable.

The similarity in chemical behavior between americium, curium and lanthanides will make it hard to separate them and all the more so since the amount of lanthanides is ten times higher than that of actinides.

This leads to considering two strategies: either simultaneously separate An (III) and Ln (III) and then subsequently separate the two groups, or look for selective extractants or act on the valencies by a specific extraction of An (Fig.3).

The first method has been most thoroughly studied.
In the USA, this has led, for the least radioactive wastes, to the TRUEX process with coextraction by CMPO (carbonyl methylene phosphine oxide) followed by the separation into groups by the TALSPEAK process based on the property of DTPA (diethylenetriamine pentacetic acid) to form strong complexes with actinides.

From 1978 to 1981, CEA in collaboration with the European Center of ISPRA, studied two simultaneous extraction processes with TBP and HDEHP (di-2-ethylhexyl phosphoric acid) which were stopped on account of the difficulties involved in implementing them and of the production of secondary waste containing phosphates when the used solvents are destroyed.

![Diagram](https://example.com/diagram.png)

**FIG 3. Possible way for minor actinides separation from HLW solutions.**

**DIAMEX PROCESS**

The difficulties mentioned above have resulted in the choice of another method: extraction using diamides, since these extractants:

- are able to extract the actinides and lanthanides at valence (III) in an acidic medium,
- lead to easy re-extraction in a low-acid medium,
- are fairly easy to synthesize, hence inexpensive and available in large quantities,
- are generators of radiolysis products that are relatively inert with respect to An (III) and Ln (III),
- are not generators of secondary wastes since they are totally incinerable.

Among the diamides, studies have been carried out over the past several years in the aim of finding, among other things, a molecule:

- barely soluble in an aqueous medium,
- soluble in an alkane-type diluents (TPH in La Hague),
- radiolysis-resistant.

A reasonable choice, that may be improved upon further, lies in dimethyl dibutyl tetradecyl methyl amide (DMDBTDMA) and has led to the development of a process called DIAMEX.
Basic studies have enabled an understanding of the extraction mechanisms for these molecules. Figure 4 shows, for example, the partitioning coefficients for uranium, plutonium, americium and iron versus acidity.

\[
\begin{align*}
R &= C_4H_9 \\
R' &= C_3H_3 \\
R'' &= C_{14}H_{29}
\end{align*}
\]

N,N-Tetraalkyl 2 propanediamide 1,3

![Graph showing partitioning coefficients versus acidity](image)

**DISTRIBUTION COEFFICIENTS WITH DIFFERENT LINKING GROUPS**

1. \(C_3H_7CH_3NOCH_2CONCH_3C_6H_{17}\)
2. \(C_3H_7CH_3NOCH_2CONH\)
3. \(C_3H_7CH_3NOCH_2CON\)
4. \(C_3H_7CH_3NOCH_2C_6H_{12}\)

**FIG. 4.**
The possibilities of this method have been successfully tested on medium activity liquid waste. Presently, tests on high activity liquid waste are in progress and show minor problems.

In a second stage, it is necessary to separate lanthanides and actinides. The method chosen uses extractants from donors "milder" than oxygen, nitrogen or sulphur for instance.

Interesting preliminary results have been obtained by using a synergetic mixture: THMASCN (trilaurylamide thyocyanate).

Direct extraction

Although this method seems the most accessible at the present time, it does present the serious disadvantage of extracting, in a first step, a mass of lanthanides ten times larger than that of the actinides, hence a facility dimensioned on the quantity of lanthanides and, for the second step consisting of separating the groups, the necessity of still operating at a very high activity level. It would therefore be much more interesting to achieve a selective extraction.

Two possible ways are under consideration:

- selectively oxidizing the actinides at a higher valence state. If valence (VI) seems difficult to stabilize at present, keeping americium at valence (IV) may be possible with polynamine type complexing agents,
- finding selective extractants. Picolinamides offer interesting prospects.

Extracting long-lived radionuclides

If elements such as iodine-129, cesium-135, selenium-79, technetium-99 play a very minor role with respect to actinides when potential toxicity is concerned, this is no longer the case, as seen above, when considered at the outlet.

Technetium is divided into 2 phases: 10 to 30% are left in dissolution residues, 70 to 90% are carried over as TcO4 into the process. In UP3, this is eliminated at the first cycle by an appropriate washing and therefore exists in a specific effluent (representing 96% of the liquid phase).

For all these elements, as well as for the actinides, the possibilities of extraction by macrocycle type molecules such as crown ethers or calixarenes are being contemplated.

The separation of actinides in view of their transmutation has been mentioned above. The latter will be performed on targets for which extractions must be adapted or studied.

TRANS_MUTATION

We will consider here only the transmutation of minors actinides and fission products even in a special program dubbed "CAPRA" has been recently launched to study the feasibility of a Pu-burner fast reactor.

Transmutation may be considered according to two scenarios:

A first scenario for which the nuclear options on the fuel cycle are maintained (reprocessing) and plutonium recycling in PWR-MOX, advanced PWRs and/or FBRs. The minor actinides would then be recycled in these reactor to significantly reduce their quantities.
A second scenario consists in specifying a drastic reduction (factor of 1000) in the plutonium and the minor actinides, possibly by calling on new technologies (accelerators, high flux reactors or dedicated reactors).

Recycling minor actinides

After separation, the recycling of minor actinides in a fission reactor may be considered according to two concepts:

- homogeneous, using standard PWR (UO$_2$ or MOX) or FBR fuel (UPuO$_2$) with small additions of minor actinides so as to modify as little as possible the fabrication, the in-reactor behavior and the core design.

- heterogeneous, in which the actinides targets (using inert matrixes) are introduced in specific core regions.

Homogeneous recycling

In the case of PWRs, the limits of enrichment in minor actinides (Np, Am) have been examined, using standard fuel or MOX fuels; with a necessity of over enrichment of the uranium or plutonium fuel.

The consequences of actinide recycling have been examined on the parameters of the core (void coefficient and its change with burn-up for instance) on the fuel cycle parameters and on the evolution of the source of potential radiotoxicity.

In the case of FRs, studies have been performed for a homogeneous recycling in EFRs (oxide fuel) type of cores. These studies have been extended to other fuels (dense) and to other reactor sizes (to limit any possible negative consequences on the reactivity coefficients and use of potential benefits on the duration of the cycle).

Heterogeneous recycling

In the case of PWRs, assemblies with Np or Am targets (in oxide form in an inert matrix) have been studied, both in the core and in assemblies at the periphery of the core.

In the case of FRs, assemblies with the same type of Np or Am targets have been studied in standard assemblies at the periphery of the reactor (blanket/reflector).

As far as heterogeneous recycling is concerned, the possibility of transmuting the long-lived fission products has also been examined (such as Tc-99), in assemblies including moderator, making use of the experience acquired in the production of Co-60 in assemblies with moderator (calcium hydride) in the radial blanket of PHENIX.

The purpose of the studies was to determine the transmutation rates, the role of the matrix, the possibility of prolonging irradiation (hence transmutation) over very long times to possibly avoid any target reprocessing. Of course, the problems of fluence and damage are studied and so is the feasibility to reprocess targets.

The main results obtained on all the reactor studies and recycling mode have been presented at the GLOBAL’93 (Sept. 1993) Conference.

A typical example of the results obtained is given in Fig. 5; future studies will focus on realistic scenarios, fuels cycle penalties, role of lanthanides, and on realistic strategies to handle Cm (not to be recycled in reactors).
SCENARIOS

A: \[ \text{U-235} \rightarrow \text{PWR (UOX)} \quad \text{WASTES:} \quad 100\% \text{ Pu}, 100\% \text{ MA} \]

B: \[ \text{U-235} \rightarrow \text{PWR (UOX)} \rightarrow \text{Pu on shelf} \quad \text{WASTES:} \quad 0.3\% \text{ Pu (losses)}, 100\% \text{ MA} \]

C: \[ \text{U-235} \rightarrow \text{PWR (UOX)} \rightarrow \text{Pu on shelf} \rightarrow \text{Np, Am on shelf} \rightarrow \text{FR} \quad \text{WASTES:} \quad 0.3\% \text{ Pu, } 10\% \text{ Am, Np (losses)} \]

\[ \text{Cm: temporary storage (left to decay)} \]

FR: \[ 2.5\% \text{ HM of Np (homogeneously in the fuel)} \]
\[ 40\% \text{ HM of Am (targets in 1st row of rad. blkt)} \]

For scenario "C": \[ 87\% \text{ PWRs, 13\% FRs} \]

Potential Radioxicity Reduction

Radiotoxicity (log Sv)

FIG. 5.
**Fuel studies**

Transmutation concepts are defined for Np and Am taking into account fuel cycle strategies.

Destruction of Np does not pose any major problem: it is a by-product of uranium consumption, as Pu and despite of a slight gamma activity due to protactinium-233 it is rather easy to handle. Diluting Np in MOX fuel looks possible as well in the fabrication step as in the reprocessing ones. Consequently we propose to recycle Np in homogeneous mode, inside MOX fuel.

Irradiation may be foreseen in FRs and in PWR either. In pile behaviour of such fuel in PWR conditions will be soon explored in the experimental OSIRIS reactor.

The analysis is more complex for multi-isotope Am; its destruction is difficult because of its gamma radioactivity and chemical properties which complicates fabrication and reprocessing.

Experiments in Phenix reactor and calculation showed that FRs offer a good potential for Am incineration.

Fabrication of Am fuels or targets will arise great difficulties. In order to minimise the flux of materials to be handle we propose to develop the heterogeneous mode.

Am targets (matrices without U) choice are defined in term of:

- safety, considering fuel reaction with cladding and sodium,
- transmutation rate limited by target in pile behaviour,
- reprocessing, checking ability of such targets to be solubilized.

The CEA program includes 2 aspects of fuel studies. For the PWRs, both concepts will be studied (Fig. 6) in OSIRIS, concentrating on neptunium, giving the largest amount of waste without availability or fabrication problems and with the advantage of having been studied for the production of plutonium isotope 238 in the Marcoule Celestin reactors (ACTINEAU program).

\[
\begin{align*}
3. & \quad MgAl_2O_4 + 20\%Np \\
& \quad 400/600J \\
4. & \quad MgAl_2O_4 + 20\%Np \\
& \quad 200J \\
5. & \quad Al_2O_3 + 20\%Np \\
& \quad 400/600J \\
2. & \quad MgAl_2O_4 + 40\%Np \\
& \quad 400/600J \\
6. & \quad Al_2O_3 + 40\%Np \\
& \quad 400/600J \\
1. & \quad UO_2 \\
8. & \quad Al_2O_3 + 40\%Np \\
& \quad 200J \\
7. & \quad UO_2 + 2\%Np \\
& \quad 400/600J
\end{align*}
\]

\[\text{FIG. 6. PWR fuel and target study in OSIRIS pile.}\]
For the FBRs, in cooperation with the transuranium Institute (CEE.JRC-ITU) an experiment called SUPERFACT has been run in PHENIX with the homogeneous concept (Figs 7 and 8) involving elements Am and Np at low (2% weight) and high (40-45%) concentrations. This experiment has concretely demonstrated the feasibility of the design, the fabrication and the irradiation, up to about 6 atoms %, of such fuels and their good behavior with respect to standard pins.

**FIG 7. Design of the PHENIX irradiation homogeneous pins**

<table>
<thead>
<tr>
<th>pin no</th>
<th>fuel</th>
<th>Linear power rating (kW/m)</th>
<th>maximum burnup (at%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>4 - 16</td>
<td>2 % Am</td>
<td>38.0 32.5</td>
<td>6.8</td>
</tr>
<tr>
<td>7 - 13</td>
<td>2 % Np</td>
<td>38.0 32.5</td>
<td>6.8</td>
</tr>
<tr>
<td>6 - 14</td>
<td>20 % Am</td>
<td>17.4 27.3</td>
<td>4.3</td>
</tr>
<tr>
<td></td>
<td>20 % Np</td>
<td></td>
<td></td>
</tr>
<tr>
<td>5 - 15</td>
<td>45 % Np</td>
<td>20.6 28.3</td>
<td>4.6</td>
</tr>
<tr>
<td>8</td>
<td>standard</td>
<td>43.0 37.0</td>
<td>8.5</td>
</tr>
</tbody>
</table>

**FIG 8 Irradiation of actinide pins**
TABLE II. TRANSMUTATION RATES (%) - CONSUMPTION CALCULATION AND EXPERIMENTAL MEASUREMENTS AFTER IRRADIATION

<table>
<thead>
<tr>
<th>Minor actinide</th>
<th>Np %</th>
<th>Am %</th>
</tr>
</thead>
<tbody>
<tr>
<td>Type of fresh fuel</td>
<td>2</td>
<td>20</td>
</tr>
<tr>
<td>Measurement</td>
<td>*</td>
<td>*</td>
</tr>
<tr>
<td>Calculation</td>
<td>24 - 26</td>
<td>27.4</td>
</tr>
</tbody>
</table>

* in progress.

After irradiation a preliminary analyses confirms the good agreement of experimental values with Np and Am 241 nuclear date calculations for transmutation rates.

Microprobe analysis (Fig. 9) show and confirm a radial flat distribution of the actinides in the pellets whatever the actinide nature or concentration (Fig. 9.c).

FIG. 9 Microprobe analysis radial distribution of actinides in pellets.
New studies are directed towards high transmutation efficiencies, i.e. high burn-up (SUPERFACT 2 experiment).

In the future, the irradiation of metallic targets is planned for CRIEPI (Japan), in association with the Karlsruhe ITU.

For the incineration of long-lived fission products, their transmutation in assemblies including moderators on the periphery of a FBR core based on the experiment run in PHENIX for the production of cobalt 60 is considered.

Table III gives encouraging preliminary data.

### Table III

<table>
<thead>
<tr>
<th></th>
<th>Irradiation in:</th>
<th>Half-lifes (years)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Tc-99</td>
</tr>
<tr>
<td>PHENIX</td>
<td>1st layer (moderated assembly)</td>
<td>5</td>
</tr>
<tr>
<td></td>
<td>2nd layer (moderated assembly)</td>
<td>10</td>
</tr>
<tr>
<td>Radial</td>
<td>3rd layer (moderated assembly)</td>
<td>20</td>
</tr>
<tr>
<td></td>
<td>1st layer (standard assembly)</td>
<td>47</td>
</tr>
<tr>
<td>Blanket</td>
<td>PWR</td>
<td>13</td>
</tr>
</tbody>
</table>

Transmutation in advanced systems

Concerning advanced systems, exploratory studies relating to scenario b) above, are also carried out on incineration systems for minor actinides and long-lived fission products, potentially more efficient. Systems have been studied based on proton accelerators to induce spallation reactions, and to produce an intense neutron source that can supply a sub-critical medium containing the waste to be destroyed (transmuted).

The physical analysis indicates that their potential role can be quantified in terms of "excess" neutrons produced, and which are available for transmutation. The present CEA studies (also reported at the GLOBAL'93 Conference) indicate that:

(a) the neutron "excess" produced in accelerator-based systems is not necessary for transuranium elements transmutations, since fast reactors offer comparable performances (if the proton beam current requirement is to be kept within reasonable limits, around 50 mA);
(b) the neutron "excess" can be possibly used to incinerate some long lived fission products (if isotopic separation is also provided).

More detailed studies are in progress.

INTERNATIONAL COLLABORATION

CEA develops these studies in cooperation with different countries: Japan (PNC, JAERI), CEE (JRC, and share cost actions with several European laboratories), CEI (IPC Moscou, Khlopin Institute, Dimitrovgrad) USA (DOE), Switzerland (Paul Scherrer Institute).
CONCLUSION

The SPIN program is aimed at separating and then transmuting long-lived elements. If all problems are far from eliminated, nevertheless the studies are developing favorably. Basic studies must still be continued and processes set up. Moreover, the cost of such operations and the doses received by the operators must be evaluated.

The information included in this document was given to me by Mr. Madic (DCC) and Mssrs Bergeonneau and Zaetta (DRN) whom I wish to thank for their help.
TRANSMUTATION OF ACTINIDES AND FISSION PRODUCTS AND CORRESPONDING FUNDAMENTAL SAFETY ASPECTS

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France

Abstract

Since the Japanese Omega-Programme had been launched in 1987, worldwide efforts in exploring the possibilities of reducing hazardous nuclear waste were started. Not all problems are solved yet, but some clarification now indicates the limits of certain proposals.

1. THE TRANSMUTATION CONCEPT

Besides the widely accepted strategy of direct disposal of nuclear waste into deep geologic formations, alternative solutions are being searched:

1. Reduction of long-term potential hazards from nuclear waste (especially from Np, Pu, Am, Cm and Te$^{99}$, I$^{129}$) via transmuting with neutrons the hazardous nuclei to much shorter lived nuclei. Here one has also to realize the aspect that any reduction of long-term hazards increases the short term hazards considerably.

2. Reduction of the main fissile material Pu with nuclear reactors. Here one has to consider two aspects:

(a) no recycling of Pu;
(b) recycling of Pu and transmutation.

Multiple recycling of Pu in nuclear reactors deteriorates the Pu composition and helps to prohibit proliferation.

These strategies have to be seen before the background that the amounts of actinides and long-lived fission products are increasing with time for the near future, as shown in Figs 1-4 for the French scenario of N4-reactors. This is true also if Pu-recycling is introduced.

The aim of hazard reduction via transmutation also becomes clear from Fig. 5, where various possibilities of handling the nuclear waste from UOX fuelled PWRs is demonstrated together with the potential radiotoxicity reduction over time.

From this discussion the following general strategies of the nuclear waste treatment follow:

1. Direct disposal (accepted strategy);
2. Partial reduction of Pu, minor actinides (MA) and fission products via transmutation, followed by disposal of "rest hazard quantities" into geological formations;
3. Reduction of nuclear waste as much as possible via transmutation in capable machines, e.g. accelerator based transmutation (rigorous strategy).
FIG. 1. Cumulated production of actinides in N4 PWRs.

FIG. 2. Cumulated generation of long lived fission products.
Recycling of Self-Generated Pu
Mox : U = 24 : 76
Burn-up 50 Gwd/t

Pu
Pu-fiss
Am
Np
Cm

Start of Operation of Pu Recycle N4-PWRs

FIG. 3. Production of actinides in recycle N4 PWRs.

Recycling of Self-Generated Pu
Mox : U = 24 : 76
Burn-up 50 Gwd/t

Tc 99
I 129

Start of Operation of Pu Recycle N4-PWRs

FIG. 4. Production of long lived FP in recycle N4 PWRs.
Potential Radiotoxicity Reduction

FIG. 5. Scenarios for waste disposal of French N4 reactors.
2. THE GENERAL SCHEME OF INVESTIGATION FOR HAZARD REDUCTION IN GERMANY AND FRANCE

Table I comprises the investigations for light water reactors (LWRs), fast reactors (FRs) and for accelerator driven spallation sources. In the LWR column, we just want to indicate that Pu-burning is most effective in fuel elements without uranium, i.e. without Pu-build-up via U-238. In the second column, the aspect of introducing moderated subassemblies into the second core zone or into the blanket region of an FBR is of definite interest because of burning fission products in special fast reactor subassemblies.

The last column in Table I states activities in the fuel of accelerator driven spallation sources, especially based on the LANL-ATW proposal.

The items in Table I will now be discussed in more detail, especially for an admixture of hazardous nuclei to the fuel of current reactors.

3. ADMIXTURE OF HAZARDOUS NUCLEI TO THE FUEL OF USUAL POWER REACTORS

One result of our investigations for fast and for thermal reactors was that fission products cannot be burnt effectively in standard power reactors: the neutron flux density would have to be raised appreciably. Full loading of all pins of LWR or a FR with a high amount of waste actinides is not allowed because of safety reasons: The Doppler and Na-void reactivity effect change into an unfavorable direction. In addition, in this case the Pu enrichment would be relatively high to get an acceptable reactor performance together with a major destruction of actinides (especially of plutonium), recriticality problems would have to be considered in case of a core melt-down.

If Pu is not removed from the waste, the longer range reduction with the admixture procedure is only moderate.

In Figures 6-8 the risk reduction by burning waste in thermal and in fast reactors is compared.

As far as the support ratio is concerned, with an admixture of 1.5% of MA (in this case Np or Am) to the fuel of LWRs, the waste of only about 2 LWRs could be cleaned up, an admixture of MA by 3% to the fuel of fast reactors (type EFR), the waste of about 3 LWRs of type N4 can be charged to a burner reactor of type EFR.

Figure 8 shows that after 24, 54 or after 108 years of burner operation the hazard reduction is converging; a reduction of the risk potential of 40% to 50% in the time range of interest can be expected, if Pu is not removed from nuclear waste.

If a normalization to the same power of both reactors is made, this support ratio increases from 3 to about 5.

4. SAFETY ASPECTS

In this chapter we address mainly those safety issues, which may become important. The corresponding investigations were recently started and have not yet been completed. In addition, reliable safety analysis can only be performed if the system of interest is defined completely in all aspects of the design. Thus we give only general remarks what topics shall be investigated in more details.
<table>
<thead>
<tr>
<th>LWRs (Φₜ ≈ 10¹⁴ n/cm² sec)</th>
<th>FBRs (Φₚ ≈ 10¹⁵ n/cm² sec)</th>
<th>Accelerator-Driven Spallation Sources (Φₜ ≈ 10¹⁶ n/cm² sec)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. Actinide burning</td>
<td>1. Actinide burning</td>
<td>1. High proton energy</td>
</tr>
<tr>
<td>ineffective; mainly</td>
<td>possible in oxide and</td>
<td>(1 - 2 GeV)</td>
</tr>
<tr>
<td>build-up of higher actinides</td>
<td>metal fuelled FBRs.</td>
<td></td>
</tr>
<tr>
<td>(σᵣ &gt; σₚ)</td>
<td>Remote handling in</td>
<td></td>
</tr>
<tr>
<td></td>
<td>fabrication necessary</td>
<td></td>
</tr>
<tr>
<td>2. Pu-burning effective in</td>
<td>2. Admixture of MA to</td>
<td>2. Continuous beam (not pulsed) with high</td>
</tr>
<tr>
<td>LWR fuel elements</td>
<td>fast reactor fuel up</td>
<td>current of = 200 mA</td>
</tr>
<tr>
<td>with (Ce/Pu) instead of</td>
<td>to about 5 %; reduction</td>
<td></td>
</tr>
<tr>
<td>(U/Pu) (no build-up of Pu²³⁹</td>
<td>of burn-up reactivity</td>
<td></td>
</tr>
<tr>
<td>via U²³⁸)</td>
<td>losses, but not</td>
<td></td>
</tr>
<tr>
<td></td>
<td>a very effective burning.</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Multiple Recycling</td>
<td></td>
</tr>
<tr>
<td>products</td>
<td>an FBRs: [DC] reduced, Na-</td>
<td></td>
</tr>
<tr>
<td></td>
<td>void increased</td>
<td></td>
</tr>
<tr>
<td>4. Special high flux</td>
<td>4. Positioning of MA-</td>
<td>4. Moderation of the spallation neutrons to Φₜ = 10¹⁶ n/cm²</td>
</tr>
<tr>
<td>reactors with Φₜ ≳ 10¹⁶ n/cm² se</td>
<td>elements in outer core and</td>
<td>sec</td>
</tr>
<tr>
<td></td>
<td>in blanket regions: no</td>
<td></td>
</tr>
<tr>
<td></td>
<td>recycling necessary, if</td>
<td></td>
</tr>
<tr>
<td></td>
<td>clad can withstand an</td>
<td></td>
</tr>
<tr>
<td></td>
<td>irradiation up to</td>
<td></td>
</tr>
<tr>
<td></td>
<td>10 - 20 years and if Φₜ ≳</td>
<td></td>
</tr>
<tr>
<td></td>
<td>10¹⁵ n/cm² sec can be</td>
<td></td>
</tr>
<tr>
<td></td>
<td>reached; build-up of Ra</td>
<td></td>
</tr>
<tr>
<td></td>
<td>after 10⁵ years</td>
<td></td>
</tr>
<tr>
<td>5. Introduction of</td>
<td>5. No FP burning in</td>
<td>5. Low inventory of fissionable nuclei (kₑₚ &lt; 0.8)</td>
</tr>
<tr>
<td>moderating blanket</td>
<td>standard FBR concepts</td>
<td></td>
</tr>
<tr>
<td>assemblies; only effective</td>
<td></td>
<td></td>
</tr>
<tr>
<td>for MA and FP burning if Φₜ</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>≥ 10¹⁵ n/cm² sec</td>
<td></td>
</tr>
<tr>
<td>6. No FP burning in</td>
<td></td>
<td></td>
</tr>
<tr>
<td>standard FBR concepts</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

*) MA: Minor Actinides
FP: Fission Products
FIG. 6. Comparison of FR and PWR burner MA load capabilities.

FIG. 7. Risk reduction by burning in FRs and PWRs.
4.1. In-pile operational safety

In case of admixing MAs to the fuel of LWRs or FRs, following topics have to be studied:

(a) Reactivity behavior of the modified system in dependence on the amount of admixture of MAs to the fuel of the system in question: Doppler and void reactivity effects, the temperature distribution over a pellet (especially for high Pu contents in the fuel), control rod worth, etc.

(b) Pu-distribution in the pellet during burn-up due to migration.

(c) Fuel-clad interaction.

(d) Stability analysis for the new core.

(e) Optimization of loading and reloading schemes for the burner reactors.

(f) Determination of $b_{eff}$ and $l_{eff}$.

4.2. Accident considerations

Besides the investigations for loss of flow (LOF) and loss of coolant accidents (LOCA) recriticality situations of the core will play a major role; especially recriticality, if high Pu-contents are loaded to the core, has to be avoided in any case.

The activity-inventory in all stages of the reactor has to be determined.

An investigation was started recently for fast cores of different size with special emphasis on the late core meltdown phases.
First results show that:

(a) The energetics of recriticalities increase with core size;
(b) The loading of MAs together with high enriched Pu leads to a deterioration of the Doppler effect and the kinetic parameters which gives a significant increase of the energy release during an excursion.
(c) As the cores loaded with MAs will probably have no blankets, a higher potential for early fuel losses exists and the disassembly effect is enhanced. The stronger material dispersion leads to a reduction in accident energetics.

From this first analysis would follow that the amount of admixtures would have to be reduced - or burner reactors should be either of medium size or of a module type.

4.3. Out-of-pile considerations

(a) In a fuel with MA the decay heat increases. The consequences for fuel transport and interim storage conditions have to be analyzed.
(b) A major point of investigations should be the separation of Am and Cm from rare earth isotopes
(c) New reprocessing schemes have to be developed in order to avoid radiolysis
(d) The refabrication techniques have to be extended to work reliably fully remote.

5. EXPERIMENTAL SAFETY STUDIES WITHIN THE FRENCH EFTRA AND CAPRA PROGRAMME

These studies will be performed in:
- HFR - Petten (NL).
- Siloe - France.
- Phenix/Superphenix - France.

The investigations are especially designed to obtain experimental information on Pu and minor actinide burning with high Pu loadings (up to 45%).

The experiments will be executed within a tripartite cooperation of:
- the French CEA at Cadarache (CEA);
- the European Institute of Transuranium Elements in Karlsruhe (TUI);
- the Nuclear Research Center Karlsruhe (KfK);

6. BURNER SYSTEMS

In France and Germany first steps are directed towards an acceptable and licensable change of the fast reactors Superphenix or EFR (European Fast Reactor) with no blankets, and smaller size-reactors with admixtures also of Be-oxide.

7. ACCELERATOR DRIVEN TRANSMUTATION

Both CEA and KfK are participating in international benchmarks organized by NEA-NSC. Three different types of investigations are performed:

(a) Benchmarking of typical spallation problems (1993- in thick targets (Pb or W), bombarded by high energetic protons; the final comparison of contributions is not yet ready.
(b) Following the LANL proposal for a thermal transmutation system, in which the fast spallation neutrons are moderated in a $\text{D}_2\text{O}$ tank. This device should be used preferably for the destruction of long lived fission products as TC$^{99}$, I$^{129}$ and Cs$^{135}$.

(c) Intensive studies have been made for fast transmutation using the BNL proposal (Phoenix).

All systems are based on a subcritical configuration, so that fast transient situations can be avoided. For these systems, in addition, not much concern has to be given to coolant void situations or decay heat removal. But all the machines are very advanced and will take about two decades for development and realization.

Acknowledgements

The discussions with Dr. W. Maschek on the safe of aspects of minor actinide containing fuel in Fast Reactors are gratefully acknowledged.
PARTITIONING OF ACTINIDES FROM ACID WASTE SOLUTIONS OF
THE PUREX PROCESS BY SOLVENT EXTRACTION AND
EXTRACTION CHROMATOGRAPHY

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Abstract

Disposal of nuclear wastes containing long-lived alpha emitters is a matter of great environmental concern. At this Centre, solvent extraction and extraction chromatographic studies are in progress with CMPO as the main extractant for the removal of actinides from Purex high active aqueous raffinate waste (HAW) and high level waste concentrate (HLW) solutions. Batch extraction studies with HAW solutions reveal that after two contacts with 0.2 M CMPO+1.2 M TBP in dodecane, its alpha activity reduces to ~0.2% of initial value. The CMPO loaded actinides are efficiently stripped with suitable reagents. Mixer settler experiments in progress confirm these results. Extraction chromatographic studies with CMPO impregnated Chromosorb-102 (CAC) column show higher uptake of actinides and lanthanides as compared to other metal ions and the capacity in terms of Nd(III) is 18 mg/g of CAC at 3 M HNO3 for less than 1% breakthrough. The results indicate that CMPO based extraction chromatographic techniques have great potential for application in the removal of actinides from Purex waste streams.

INTRODUCTION

Disposal of nuclear wastes containing long-lived alpha emitters is a matter of great environmental concern. The removal of long lived alpha emitting actinides from high level nuclear wastes would greatly reduce the long term radiological hazards of these wastes. Many organic extractants are being tested for this purpose. In this context, Octyl(phenyl)N,N-diisobutylcarbamoylmethyl phosphine oxide (CMPO) is one of the most promising reagents for the extraction and removal of actinides from highly radioactive aqueous waste concentrate solutions [1]. At this Centre solvent extraction and extraction chromatographic studies based on CMPO are in progress for the removal of actinides from Purex high active aqueous raffinate waste (HAW) and high level waste concentrate (HLW) solutions.

Using this reagent, the extraction behavior of actinides and fission products from nitric acid solutions [2], tail-end purification of Am from Pu loading effluents [3] and recovery of Pu from oxalate supernatants [4] have been studied. The studies conducted on partitioning and recovery of actinides from Purex aqueous HAW and HLW using solvent extraction grade CMPO to propose suitable extraction schemes form a part of this paper.

Since extraction chromatography blends the favorable characteristics of solvent extraction and ion exchange, it is finding increasing use in the separation and preconcentration of various metal ions. Extraction chromatographic studies [5,6] using bifunctional organophosphorus compounds have shown high selectivity for the extraction of actinides and lanthanides. Hence extraction chromatographic technique using CMPO adsorbed on chromosorb-102 (styrene divinylbenzene polymer, 100-120 mesh size ) (CAC) was tested as an alternative to the CMPO based extraction process to recover minor actinides from the high level waste solutions of Purex origin.
With phosphonic acids as extractants, a good separation of tetra and trivalent actinides (along with rare earths) can be achieved by careful manipulation of extraction acidity. These reagents also possess a good affinity for U(VI). Hence using an indigenously prepared 2-ethylhexyl hydrogen 2-ethylhexyl phosphonate (KSM-17, equivalent to PC-88A) impregnated on Chromosorb 102 (KSMC), several extraction chromatographic techniques were tried for the following tasks: a) recovery of $^{147}$Pm from Am(III)-rare earth fraction eluted from CAC column b) removal of Pu from Am present in the loading effluents of anion exchange Pu purification and c) sorption of $^{233}$U from $^{233}$U-Al alloy scrap recovery wastes.

**SOLVENT EXTRACTION STUDIES WITH CMPO**

The present day Purex process technology generally yields HLW solution mainly in nitrate form. However, the earlier Purex process utilized ferrous sulphamate as a reducing agent for plutonium partitioning and this resulted in the generation of sulphate bearing HLW with low acidity. The extraction and removal of actinides from such waste pose problems as sulphate ion is a strong complexing agent. In addition to minor actinides and fission products, these wastes invariably contain uranium, S.S. corrosion products and other inert materials.

In the present studies, it was observed that a prior removal of the uranium present in the waste by TBP extraction helps in the subsequent extraction of trivalent actinides. Otherwise, uranium competes for the available CMPO in the organic phase and saturation of CMPO with uranium adversely affects the extraction of trivalent actinide ions.

Hence to assess the extractability of UO$_2^{2+}$ from sulphate medium at about 0.3 M HNO$_3$ by 30% TBP, extraction studies were carried out with synthetic HLW solution containing uranium. Two successive contacts with TBP reduced the raffinate uranium values to less than 2 g/l, even when the feed contained as high as 25 g/l uranium. At this concentration range of uranium, the aqueous feed did not lead to any problems like third phase formation during CMPO extraction.

Three sulphate bearing synthetic waste solutions containing $^{235}$U, $^{239}$Pu and $^{241}$Am tracers were prepared at three acidities: 0.3, 1.0 and 2.0 M HNO$_3$. Two contacts were given with 30% TBP followed by four contacts with 0.2 M CMPO + 1.2 M TBP in dodecane, all at 1:1 volume ratio. At all the three acidities, the final aqueous raffinate was found to contain negligible alpha activity suggesting that even at low acidities of about 0.3 M HNO$_3$ the alpha activity of the sulphate bearing waste solutions can be brought down to considerably low levels by CMPO extraction.

Further experiments were conducted with a typical sulphate bearing HLW of Purex origin. Table 1 gives the level of acidity, total alpha, UO$_2^{2+}$ as well as SO$_4^{2-}$ ion concentration in the waste after extraction with TBP (two contacts) and CMPO + TBP mixture (four contacts). The total alpha activity in the final raffinate is reduced to less than 0.4% of its original value and the activity corresponds to an alpha load of about 6 nCi/ml of the solution. The distribution pattern of fission products during the extraction studies reveals that after four CMPO contacts Ce is completely extracted, Ru is partially extracted, whereas the uptake of Cs is negligible.

The stripping of metal ions from the cumulated loaded organic phase was done with 0.04 M HNO$_3$ (for Am and rare earths), 0.01 M H$_2$C$_2$O$_4$ (for Pu(IV)) and 0.25 M Na$_2$CO$_3$ (for U(VI)) at 1:1 phase ratio. The recovery of total alpha activity as well as uranium was more than 99%. The lean organic phase did not show alpha activity above the background level.

Similar results were obtained during extraction studies using a mixture of 0.2M CMPO+1.4 M TBP with Purex HLW in nitrate form as given in Table 2.
### Table 1

**Extraction and Stripping Data for Sulphate Bearing Purex Waste Solution**

<table>
<thead>
<tr>
<th>Component</th>
<th>Purex HLW Raff. after 2 TBP contacts</th>
<th>HLW Raffinate after 2 TBP &amp; 4 CMPO contacts</th>
<th>Lean Org. after stripping</th>
</tr>
</thead>
<tbody>
<tr>
<td>Acidity, M</td>
<td>0.35</td>
<td>0.35</td>
<td>...</td>
</tr>
<tr>
<td>Alpha Activity (nCi/ml)</td>
<td>1572.97</td>
<td>6.10</td>
<td>ND</td>
</tr>
<tr>
<td>Uranium, g/l</td>
<td>&lt; 2.00</td>
<td>ND</td>
<td>ND</td>
</tr>
<tr>
<td>Beta activity (Ci/l)</td>
<td>4.75</td>
<td>4.11</td>
<td>0.05x10^-3</td>
</tr>
<tr>
<td>Sulphate, g/l</td>
<td>12.02</td>
<td>12.02</td>
<td>...</td>
</tr>
<tr>
<td>Fission Products (mc/l):</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ce</td>
<td>168.0</td>
<td>ND</td>
<td>4.50x10^-3</td>
</tr>
<tr>
<td>Ru</td>
<td>139.5</td>
<td>117.3</td>
<td>0.07</td>
</tr>
<tr>
<td>Cs</td>
<td>2065.4</td>
<td>2064.0</td>
<td>0.002</td>
</tr>
<tr>
<td>Sr</td>
<td>790.0</td>
<td>790.0</td>
<td>...</td>
</tr>
</tbody>
</table>

### Table 2

**Extraction and Stripping Data for High Level Waste (HLW) Solution**

<table>
<thead>
<tr>
<th>Component</th>
<th>Purex HLW Raff. after one TBP contact</th>
<th>HLW Raffinate after one TBP &amp; 4 CMPO contacts</th>
<th>Lean Org. after stripping</th>
</tr>
</thead>
<tbody>
<tr>
<td>Acidity, M</td>
<td>3.05</td>
<td>3.04</td>
<td>...</td>
</tr>
<tr>
<td>Alpha Activity (nCi/ml)</td>
<td>7009.91</td>
<td>2.25</td>
<td>ND</td>
</tr>
<tr>
<td>Uranium, g/l</td>
<td>1.00</td>
<td>ND</td>
<td>ND</td>
</tr>
<tr>
<td>Beta activity (Ci/l)</td>
<td>72.72</td>
<td>45.13</td>
<td>21.63x10^-3</td>
</tr>
<tr>
<td>Fission Products (mc/l):</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ce</td>
<td>13170.0</td>
<td>1.3x10^-2</td>
<td>0.03</td>
</tr>
<tr>
<td>Ru</td>
<td>5700.0</td>
<td>2120.0</td>
<td>32.00</td>
</tr>
<tr>
<td>Cs</td>
<td>20160.0</td>
<td>20090.0</td>
<td>0.03</td>
</tr>
<tr>
<td>Sr</td>
<td>8280.0</td>
<td>8010.0</td>
<td>...</td>
</tr>
</tbody>
</table>
As Purex HAW solutions have low uranium content (<1 g/l), batch extraction studies without any prior extraction with TBP reveal that after two contacts with 0.2 M CMPO + 1.2 M TBP in dodecane, its alpha activity reduces to ~0.2% of initial value.

Thus the results from these studies show that the uranium content of the nitrate as well as sulphate bearing Purex HLW could easily be reduced to low levels by prior TBP contacts and the alpha burden of these solutions could be reduced considerably by extraction with CMPO. It is also observed that the presence of sulphate ion in the feed generally decreases the extraction of metal ions.

Counter current extraction experiments that are in progress [7] with syntheticsulphate bearing waste using low volume mixer settlers (single stage mixer capacity of 30 ml and settler capacity of 130ml) tend to confirm these results. The uranium content in the feed could be brought down to < 0.01 g/l by providing four stages for 30% TBP extraction step at 1:1 volume ratio. This was followed by six stages of extraction for actinide removal with 0.2 M CMPO + 1.2 M TBP mixture in dodecane at 1:1 flow ratio. The scheme under test is given below:

**FEED AQ.**

\[
\begin{align*}
\text{HNO}_3 & = 0.35 \text{ M} \\
\text{Gross } \alpha & = 7232 \text{ Bq/ml} \\
\text{Pu } \alpha & = 5785 \text{ Bq/ml} \\
\text{Am } \alpha & = 1447 \text{ Bq/ml} \\
\text{SO}_4^{2-} & = 14.32 \text{ g/l} \\
\text{Flow} & = 1 \text{ Vol.}
\end{align*}
\]

**FEED ORG.**

\[
\begin{align*}
\text{Pu } \alpha & = 195 \text{nCi/ml} \\
\text{Am } \alpha & = 5785 \text{ Bq/ml} \\
\text{SO}_4^{2-} & = 14.32 \text{ g/l} \\
\text{Flow} & = 1 \text{ Vol.}
\end{align*}
\]

\[\text{CMPO EXTRACTION}\]

\[
\begin{align*}
\text{Gross } \alpha & = \text{Near B.G} \\
\text{HNO}_3 & = 0.27 \text{ M}
\end{align*}
\]

**LOADED SOLVENT**

**AQ. RAFFINATE**

The stage-wise mixer settler sample analysis indicates that almost all the alpha activity was removed in the first four stages. In the first stage of mixer settler itself, the organic phase extracted more than 95% of the total alpha activities and Pu extraction was almost 100%. The final raffinate showed alpha activity near to back-ground. Almost 100% of \(^{144}\text{Ce}\) and about 60% of the \(^{106}\text{Ru}\) also got extracted along with actinide.

During six stages of stripping with 0.04 M HNO\textsubscript{3} for Am and rare earths removal at 1:1 volume ratio, the acidity (0.35 M) of the incoming loaded organic feed from extraction section led to an elevated acid profile in the initial stages of stripping. This in turn resulted in Am and Ce reflux in these stages. Hence, appropriate scrub steps are being evolved to reduce the acidity and \(\beta, \gamma\) activity of the out-going organic from the extraction section. After dilute HNO\textsubscript{3} stripping, the lean CMPO after six stages of 0.01 M H\textsubscript{2}C\textsubscript{2}O\textsubscript{4} stripping was almost free from Pu, Am and Ce. If the CMPO phase contains large amounts of Pu, use of oxalic acid may lead to precipitation problems. Under these conditions, other complexing agents like acetic acid in presence of nitric acid may have to be employed. U was removed by six stages of 0.25 M Na\textsubscript{2}CO\textsubscript{3} stripping. \(^{106}\text{Ru}\) activity followed this stream.
Further studies are still in progress. Initial results indicate the process feasibility of this technique for the removal of actinides from HLW and sulphate bearing low acid wastes. The path of neptunium which generally exists as Np(V) in HLW has not been studied in these experiments. It has to be converted to Np(IV) or Np(VI) for its extraction by CMPO/TBP mixture. Various redox reagents are under test for this purpose.

EXTRACTION CHROMATOGRAPHIC STUDIES WITH CMPO

The extraction chromatographic studies have been conducted on the sorption behavior of U(VI), Pu(IV), Am(III) and several fission products from nitric acid media and from Purex HAW and sulphate bearing low acid waste using CMPO adsorbed on inert support Chromosorb 102 (CAC).

Table 3 gives the D values of all the nuclides studied as a function of HNO\(_3\) concentration. It is seen that the D values at 3.0 M HNO\(_3\) follow the order: Pu(IV) > U(VI) > Am(III) > Eu(III) > Fe(III) > Zr(IV) > Ru(III) > TcO\(_4\). This suggests that the actinides along with lanthanides can be separated from other fission products and the inert materials by CAC extraction chromatography.

Table 4 gives the D values of U(VI) and Pu(IV) at 3 M HNO\(_3\) with varying concentration of U(VI). The D values for both these metal ions decrease with increasing U(VI)
Table 4

Distribution Ratios of Pu(IV) & U(VI) in CAC with varying U(VI) Concentrations at 3 M Nitric acid

<table>
<thead>
<tr>
<th>[UO$_2^{++}$] (g/l)</th>
<th>Distribution ratios</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>UO$_2^{++}$</td>
</tr>
<tr>
<td>0.00 *</td>
<td>5.87x10^4</td>
</tr>
<tr>
<td>0.28</td>
<td>1.34x10^3</td>
</tr>
<tr>
<td>0.57</td>
<td>344.3</td>
</tr>
<tr>
<td>1.13</td>
<td>60.9</td>
</tr>
<tr>
<td>2.27</td>
<td>23.2</td>
</tr>
<tr>
<td>5.67</td>
<td>12.2</td>
</tr>
<tr>
<td>11.34</td>
<td>5.9</td>
</tr>
</tbody>
</table>

* Using U$^{233}$ & Pu tracer alone.

concentration. The D values of Pu(IV) as a function of HNO$_3$ concentration (0.5-7.3 M) at fixed U(VI) concentration (1.13 g/l) and in its absence are given in Table 5. It is seen that in general, the D values in the presence of U(VI) are about two orders of magnitude lower than those in the absence of U(VI) and at lower U(VI) concentrations (< 1 g/l), the D values for both the metal ions are reasonably high. Table 6 gives the D values of Am(III) as a function of Nd(III) and U(VI) concentrations. Here again the D values decrease drastically with increasing Nd(III) and U(VI) concentrations. These batch wise loading experiments in presence of Nd(III) / U(VI) show that at lower concentration of these metal ions, the uptake of Pu(IV), U(VI) and Am(III) are reasonably high.

The loading and elution of Nd(III), U(VI) and Pu(IV) was done individually using a CAC column. In the case of Nd(III) ~ 30 mg of the metal ion (in 3.0 M HNO$_3$) could be loaded on 1.7 g CAC column without leakage. After washing the column, Nd was eluted with 0.04 M HNO$_3$ and the recovery was found to be 99.9%. Using the same column, ~ 90 mg of U(VI) (in 3.0 M HNO$_3$) could be loaded and the same was almost quantitatively eluted with 0.25 M Na$_2$CO$_3$ solution. In the case of Pu(IV), using a smaller column (100 mg CAC), ~ 0.6 mg of the metal ion could be loaded without any leakage, though the capacity of CAC for Pu is much more than this. Pu was quantitatively recovered by eluting with 0.01 M oxalic acid.

The CAC was tested using the actual HAW solution originating from the Purex processing of the thermal reactor fuels to assess its performance under these conditions. When uranium content was in trace level, a large volume of this solution could be passed
Table 5

Influence of U on Pu uptake by CAC at varying nitric acid Concentration

<table>
<thead>
<tr>
<th>HNO3 M</th>
<th>D for Pu(IV) without U(VI)</th>
<th>D for Pu(IV) at 1.13 g/l U(VI)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5</td>
<td>1.69x10^5</td>
<td>2.65x10^2</td>
</tr>
<tr>
<td>1.0</td>
<td>1.79x10^5</td>
<td>4.18x10^2</td>
</tr>
<tr>
<td>2.0</td>
<td>2.54x10^5</td>
<td>6.27x10^2</td>
</tr>
<tr>
<td>3.0</td>
<td>2.99x10^5</td>
<td>7.38x10^2</td>
</tr>
<tr>
<td>4.0</td>
<td>——</td>
<td>7.13x10^2</td>
</tr>
<tr>
<td>5.0</td>
<td>1.57x10^5</td>
<td>6.90x10^2</td>
</tr>
<tr>
<td>6.0</td>
<td>6.90x10^4</td>
<td>6.60x10^2</td>
</tr>
<tr>
<td>7.3</td>
<td>——</td>
<td>5.68x10^2</td>
</tr>
</tbody>
</table>

d by a small column with efficient uptake of the actinides. About 100 ml of this solution with 1.15x10^8 Bq/ml (~865 nCi/ml) could be passed through ~2 g CAC without any leakage of alpha activity. Analyses of the effluents indicate that 137Cs and 90Sr are not taken up in the column and Ru is taken up partially. U, Pu, Am and Ce in the effluent solution from the CAC column was not found above the background level throughout the loading and even in the washing fractions. This shows the efficient loading of the actinides and lanthanides on the 2 g CAC column up to at least 100 ml of the HAW solution. The activities were eluted from the column successively by 0.04 M HNO3, 0.01 M oxalic acid and 0.25 M Na2CO3 solutions. Almost all of the Am activity (along with the rare earths) is eluted with 0.04 M HNO3, which contains a very small fraction (0.1%) of the Pu activity. Efforts to separate Am and rare earths from this fraction are described in the following section. While eluting with 0.01 M oxalic acid almost all Pu activity and the remaining Am activity come down. Finally with 0.25 M Na2CO3 solution, all the U and the remaining other (actinides and rare earths) activities are eluted.
A synthetic sulphate bearing waste solution, with uranium concentration depleted to 0.12 g/l by 30% TBP contact, was passed through an identical CAC column. It was found that 85 ml of the solution with 2.5 ug/ml Pu and 0.0084 ug/ml Am (Table 7) could be passed without any leakage of alpha activity due to Pu and Am. It was also observed that 180 ml of the solution could be passed through the CAC column without any leakage of plutonium (effluent contained only Am). Ce followed the course of americium. Elution was carried out in a similar manner as mentioned before with good recovery of individual actinides.

These experiments indicate that extraction chromatographic technique can be used to remove the residual alpha emitting actinides from Purex high level waste streams using CAC either before or after concentration by evaporation. The CAC has good capacity to remove plutonium and americium from HAW solution that contains very little uranium.

Based on these studies a scheme (Fig. 1) has been proposed for the removal of actinides from high level waste, depleted in uranium by TBP extraction.

EXTRACTION CHROMATOGRAPHIC STUDIES WITH PHOSPHONIC ACID (PC-88A)

2-ethylhexyl hydrogen 2-ethylhexyl phosphonate, KSM-17, (equivalent to PC-88A) was used as the solvent to study the extraction of Pu, Am, U, and FPs. As extraction chromatographic technique appeared more attractive than solvent extraction route, further

| Table 7 |
| Composition of a Typical Uranium Depleted Synthetic Sulphate Bearing Waste Solution After Two contacts with 30% TBP |

| H⁺ | = 0.35 M |
| U | = 0.12 g/l |
| Pu | = 2.502 mg/l |
| Am | = 0.0084 mg/l |
| Gross Beta | = 22.12 mCi/l |
| Fission Products Spiked: |
| 144Ce | = 3.94 mCi/l |
| 106Ru | = 0.96 mCi/l |
| 137Cs | = 2.40 mCi/l |
| 95Zr/95Nb | = ND |

Inert materials:

| Na | = 17.06 g/l |
| Al | = 5.60 g/l |
| Fe | = 2.60 g/l |
| Cr | = 0.64 g/l |
| Ni | = 0.50 g/l |
| Ca | = 0.27 g/l |
| Cs | = 93.2 mg/l |
| Ce | = 0.30 mg/l |
| SO₄²⁻ | = 14.32 g/l |
batch experiments were carried out with KSM-17 impregnated chromosorb-102 (KSMC) to determine D values for Ce(III), Pm(III), Eu(III), Ru(NO)(III), Am(III) and Pu(IV) from 0.01 to 5 M nitric acid. The results are given in Table 8. For all the trivalent elements taken, D values decrease with increase in acidity. In the case of Ru it is negligible throughout the range of acidities studied. D values for Pu(IV) are more or less independent of the concentration of nitric acid in the range. The D values are in the order Pu(IV) > Eu(III) > Pm(III) > Am(III) > Ce(III). Though not reported, the D values for U(VI) remained very high (> $10^3$) throughout the acid range tested. As Pu in contrast to Am has got a high D at high feed acidities and fission products too have low D values at these acidities, the feasibility of using this reagent in actinide and lanthanide separations was investigated.
Table 8
Distribution ratios for metal ions between KSMC and varying concentrations of nitric acid

<table>
<thead>
<tr>
<th>Conc. of HNO₃</th>
<th>Ce(III)</th>
<th>Pm(III)</th>
<th>Eu(III)</th>
<th>Am(III)</th>
<th>Pu(IV)</th>
<th>Pu(IV)*</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.01</td>
<td>5191</td>
<td>8196</td>
<td>9846</td>
<td>6239</td>
<td>-----</td>
<td>-----</td>
</tr>
<tr>
<td>0.05</td>
<td>34.2</td>
<td>414</td>
<td>729</td>
<td>58.2</td>
<td>2331</td>
<td>-----</td>
</tr>
<tr>
<td>0.10</td>
<td>0.8</td>
<td>23.8</td>
<td>56</td>
<td>1.9</td>
<td>2294</td>
<td>-----</td>
</tr>
<tr>
<td>1.00</td>
<td>0.6</td>
<td>0.13</td>
<td>0.49</td>
<td>1.3</td>
<td>2305</td>
<td>2305</td>
</tr>
<tr>
<td>2.00</td>
<td>0.3</td>
<td>0.03</td>
<td>0.01</td>
<td>1.2</td>
<td>2312</td>
<td>2377</td>
</tr>
<tr>
<td>3.00</td>
<td>0.1</td>
<td>0.03</td>
<td>0.01</td>
<td>0.3</td>
<td>2338</td>
<td>2309</td>
</tr>
<tr>
<td>5.00</td>
<td>0.01</td>
<td>0.01</td>
<td>0.01</td>
<td>0.01</td>
<td>2332</td>
<td>2378</td>
</tr>
</tbody>
</table>

* in presence of 50 mg/l Nd(III)

Separation of Am from ¹⁴⁷Pm and other rare earths

As mentioned earlier, experiments using CMPO based extraction chromatographic column and Purex HAW solution generate an Am-rare earth eluate fraction in dilute nitric acid. The Am requires further separation from rare earths. The major radio nuclides present in this 0.04 M HNO₃ eluted fraction of CMPO-Chromosorb column are ¹⁴⁴Ce, ¹⁴⁴Pr, ²⁴¹Am, ¹⁴⁷Pm, ¹⁵⁴Eu and traces of ¹⁰⁶Ru. After adjusting the acidity to 0.01 M, it was passed through a 2.0 gm KSMC column and all the activity except that of ¹⁰⁶Ru got sorbed on the column. ¹⁰⁶Ru was washed off with 0.01N HNO₃. The sorbed band was developed into various fractions with varying concentrations of HNO₃ (0.05 - 0.50 M) and a pure beta emitting ¹⁴⁷Pm fraction could be collected. The elution pattern is shown in Fig. 2. Based on these results,
a scheme shown in Fig. 3 was standardized for the separation of Am and Ce from $^{147}$Pm and other rare earths. $^{242}$Cm, if present, might follow $^{147}$Pm. Subsequently, $^{144}$Ce can be separated from Am(III) after oxidizing Ce to Ce(IV).

**Separation of Pu and Am**

During the purification of aged plutonium by anion exchange technique, the effluents leaving the ion exchange column contain americium with low concentrations of plutonium and uranium. When such solution at 3-4 M nitric acid is passed through KSMC column, plutonium and uranium get sorbed on the column. Americium does not get sorbed on the column above 1 M acid concentration. Hence it is possible to obtain an effluent containing only $^{241}$Am that is free from Pu and U.

**FIG. 3. Schematic diagram for the separation of $^{147}$Pu.**
Recovery of $^{233}$U from recycle wastes

The raffinates generated during the recovery of $^{233}$U from $^{233}$U-Al alloy scrap solutions by TBP extraction contain $^{233}$U in mg range and pose problems of storage and disposal. These solutions are at ~4 M nitric acid. On passing through a column of KSMC, $^{233}$U from the solution gets sorbed on the column leaving Al in the effluent. Uranium sorption capacity is found to be about 60 mg/g KSMC.

CONCLUSIONS

The batch extraction studies with CMPO reveal that a prior TBP extraction step to remove U(VI) followed by four contacts with 0.2 M CMPO+1.2 or 1.4 M TBP in dodecane could reduce the alpha activity of the HLW to less than 2.25 nCi/ml and that of sulphate bearing low acid waste to less than 6.10 nCi/ml. The CMPO loaded actinides could be efficiently stripped with suitable reagents. Mixer settler experiments in progress confirm the results.

The batch wise CAC extraction chromatographic loading experiments have shown that at lower concentrations of Nd(III) (or the total rare earths) and U(VI), the uptake of Pu(IV), U(VI) and Am(III) are reasonably high.

The loading of the metal ions individually on a CAC column has shown that Nd(III) (~30 mg), U(VI) (~90 mg) could easily be loaded on a column containing ~2.0 g CAC, while Pu(IV) (~0.6 mg) could be loaded on a column containing ~100 mg of CAC without any leakage.

About 100 ml of Purex HAW solution could be loaded on a column containing ~2 g of CAC. The effluent did not contain any alpha activity above the background level. After washing, Am (and rare earths) were eluted with 0.04 M HNO$_3$, Pu by 0.01 M oxalic acid and U(VI) by 0.25 M Na$_2$CO$_3$ solution with efficient recoveries.

The extraction chromatographic study with KSM-17 (= PC-88A) adsorbed on Chromosorb 102 column reveal its utility in the following tasks: a) recovery of $^{147}$Pm from Am(III)-rare earth fraction eluted from CMPO column b) removal of Pu from Am present in the loading effluents of anion exchange Pu purification and c) sorption of $^{233}$U from $^{233}$U-Al alloy scrap recovery wastes.

REFERENCES

PARTITIONING AND TRANSMUTATION R&D PROGRAM 'OMEGA' AND PRESENT STATUS OF THE TRANSMUTATION STUDY AT JAERI

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Abstract

The overview of the Japanese partitioning and transmutation R&D program "OMEGA" is described. The R&D areas in the leading organization of OMEGA, namely JAERI, PNC and CRIEPI where the major R&D activities are being carried out are described.

The various activities relating with OMEGA program have been performed at JAERI. Among these activities, the transmutation studies are briefly described. The current status of the conceptual design study of an accelerator-driven transmutation system with a subcritical minor actinide core is discussed. JAERI is proposing the development of a high-intensity proton linear accelerator ETA with a proton energy of 1.5GeV and an average current of 10mA for the engineering test of an accelerator-driven transmutation system. In the course of the accelerator development, the R&D on the BTA which is a lower energy portion of ETA with a proton energy of 10MeV and a current of 10mA is in progress. Some of the achievement in BTA development are presented.

1. BACK GROUND OF OMEGA PROGRAM

In 1973, the Japan Atomic Energy Industry Forum published the report on the waste management of long-lived nuclides after the two years assessment studies by the groups of scientists and engineers in Japan. In that report entitled "A closed system for radioactivity", the importance of research and development for partitioning and transmutation (P-T) of long-lived nuclides was pointed out as long term efforts in developing a complete system for radioactive waste management.

Even after the pessimistic conclusions for P-T of high level radioactive waste(HLW) drawn by Oak Ridge National Laboratory(1980), IAEA(1982), etc., small number of groups in Japan, as well as Europe, the United States etc. continued their studies in P-T as a basic research.

Based on their studies, the interested Japanese organizations proposed to initiate a major R&D program on P-T. The development of P-T technology was deemed to be quite an interesting subject for ongoing investigation from the perspectives of possible long term advances in radioactive waste management and potential utilization of resources.

In 1987, Japan's Atomic Energy Commission(AEC) concluded that the potential benefits from the use of some elements in fission products and from recycling minor actinides for power generation could be achieved provided that a well planned, efficient and effective R&D program could be formulated. The AEC then submitted in October 1988 a report entitled "Long Term Program for Research and Development on Nuclide Partitioning and Transmutation Technology", which plots a course for technological development up to the year 2,000.
The program is called "OMEGA" which is the acronym derived from Options Making Extra Gains from Actinides and fission products.

The R&D programs were jointly stimulated by the collaborative efforts of the Japan Atomic Energy Research Institute (JAERI) and the Power Reactor and Nuclear Fuel Development Corporation (PNC). In the public sector, the Central Research Institute of Electric Power Industry (CRIEPI) also has been carrying out R&D on this subject.

The Japanese government (represented by Science and Technology Agency; STA) proposed an international cooperation for information exchange to cover the areas of nuclear physics, reactor physics, advanced process technologies and physico-chemical characterization relevant to P-T technology under the framework of the OECD Nuclear Energy Agency, in January 1989. The first information exchange meeting on this subject was held in Japan in November 1990. Eleven OECD member countries and two international organizations, namely IAEA and Joint Research Center of CEC, participated in the meeting.

The program is conceived as a research effort to pursue benefits for future generations through the long term basic R&D, and is not to seek a short term alternative for established or planned fuel cycle back-end policies. The program is expected to serve to revitalize nuclear R&D in general, and also to attract capable young researchers dedicated to bringing the nuclear option into the 21st century in a healthy state. In addition, advancement of technologies such as laser and accelerator technology, as advocated in this program, will provide spin-offs for other fields of science and technology.

2. RESEARCH AND DEVELOPMENT AREAS UNDER OMEGA PROGRAM

The program is to be proceeded in two steps: the phase-I and II. The phase-I covers a period up to about 1996, and the phase-II covers a period from about 1997 to about 2000. In general, the basic studies and testing are to be conducted in the phase-I to evaluate various concepts and to develop required technologies. In the phase-II, engineering tests of technologies or demonstration of concepts are planned. After 2000, pilot facilities will be built to demonstrate the P-T technology.

The program covers the following fields;

A. Physical and Chemical Properties of Minor Actinides and Fission Products

Reliable data base of minor actinides and long-lived fission products is indispensable to proceed the program. Underlying studies will improve understanding of the science and technology for separation and recovery of these nuclides from HLW, for fabrication of actinide fuels for recycling to reactors or accelerator driven systems for transmutation, and for utilization of these nuclides. Nuclear data and thermodynamic data of these nuclides are measured, compiled and evaluated for reactor physics and materials development. Following items are to be studied;

- Chemical properties and behavior of actinide species in aqueous and organic solutions
- Analytical techniques and methods
- Physical and chemical properties of actinide compound
- Collection and evaluation of nuclear and thermodynamic data

B. Partitioning

Advanced separation technology of minor actinides and fission products is to be developed on the basis of current trends in the chemical separation processes. Both wet and dry processes are to be developed. This would include application of new extracting solvents, laser induced separation, and sublimation and volatilization processes. Studies for utilization of separated nuclides will also be included. The following items are to be studied;

- Partitioning for HLW
- Partitioning in the reprocessing process
- Recovery of platinum group metals
- Cost estimation of a process.

C. Transmutation

Recently new approaches such as minor actinide transmutation using actinide burner reactors, metal fuel FBRs with dry reprocessing, intense beam proton accelerator as neutron source have been studied. Optimization of minor actinide recycling into Pu-LWR and MOX-FBR is to be studied. Studies of transmutation of Sr-90, Cs-137 with an electron accelerator are also proposed. Fields to be studied are the followings;

- Nuclear data and fuel property data of minor actinides
- Development of computer code
- System design study
- Reactor fuel and accelerator target development
- Fabrication technology of the fuel and target materials
- Development of high power accelerator for transmutation.
- Cost estimation of R&D, construction, and fuel cycle

The areas covered by the program and R&D activities are illustrated in Fig. 1.

3. R&D ACTIVITIES OF JAERI UNDER OMEGA PROGRAM

JAERI has been carrying out studies on the partitioning and transmutation since 1973. A partitioning process has been developed to separate elements in the high level liquid waste (HLW) into four groups; minor actinide element, Sr and Cs, Tc and platinum group elements, and others. A series of laboratory-scaled tests with the actual or synthesized HLW indicated that the proposed partitioning process would be promising. The entire process is to be tested with the actual HLW at the NUCEF(Nuclear Fuel Cycle Safety Engineering Research Facility), of which construction has been completed and the hot operation will be started in spring 1994 at the JAERI Tokai Establishment. The details of partitioning study at JAERI will be presented by Y. Morita at this meeting[1].

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Fig. 1  P-T R/D ACTIVITIES UNDER OMEGA PROGRAM
A special transmutation system with a very hard neutron energy spectrum and high neutron flux would be very efficient and effective for minor actinides (MA) transmutation if it is technically feasible. In this context, JAERI has been developing the concept of MA burner reactors (ABR: Actinide Burner Reactor) and an accelerator-driven MA transmutation system. Once these systems will become available, the troublesome MA from the waste management viewpoint will be confined in a separate fuel cycle from the commercial one.

The concept of the double strata fuel cycle consisting of the commercial fuel cycle and the Partitioning-Transmutation (P-T) cycle is illustrated in Fig. 2. The final HLW from this fuel cycle contains only fission products. The separate treatment of MA from the commercial cycle will be preferable for the conventional fuel cycle because MA is a strong neutron emitter and thus, the MA recycling in the conventional fuel cycle will cause the problem in the radiation shielding of the fuel cycle facilities.

![Diagram of the double strata fuel cycle](image-url)

**Fig. 2** Flow of high-level radioactive waste per year through double strata fuel cycle combined with partitioning and transmutation (MA burner reactor) cycle
A conceptual design study on ABRs and the advantage of the independent transmutation system from the conventional fuel cycle are discussed from both the reactor physics and fuel cycle facilities view points. The details of the study are discussed by T. Mukaiyama at this meeting[2].

A conceptual design study has been carried out for an accelerator-driven MA transmutation system with sodium-cooled subcritical core loaded with actinide alloy fuel and spallation target of solid tungsten. In the system, about 250 kg of actinides are transmuted and 820 MW thermal power is generated by using a 1.5 GeV-39 mA proton beam. The plant produces electric power of 246 MW, and supplies sufficient electricity to its own accelerator. The molten-salt target/core system with a capability of continuous on-line processing is also examined.

In order to conduct various engineering tests for the accelerator driven transmutation system, the high-intensity proton linear accelerator, Engineering Test Accelerator (ETA) with an energy of 1.5 GeV and an average current of 10 mA, has been proposed. In the course of the ETA development, the R&D works of the Basic Technology Accelerator (BTA) with an energy of 10 MeV and a current of 10 mA are in progress.

To support the above-mentioned activities on transmutation systems, several basic researches are in progress or planned. These include the nuclear data measurements of minor actinides and phase diagram, thermodynamic studies on minor actinide compounds that are being carried. The spallation integral experiments at the National Laboratory for High Energy Physics (KEK) are under way. The irradiation experiments of fuel/target of transmutation systems are planned under the collaboration with FFITF or Russia.

3.1 R&D ACTIVITIES ON ACCELERATOR-BASED MA TRANSMUTATION

The R&D includes the conceptual design study of accelerator-based transmutation plant, the development of spallation simulation code system, and the spallation integral experiments.

In the design study of transmutation plant, two types of system concept are being investigated; solid system and molten-salt system. In either system, a MA-loaded subcritical core is driven by a high intensity proton linear accelerator and uses fast neutron to burn MAs efficiently. The design of the solid system is based on the status of sodium-cooled fast breeder reactor technology. Nuclear and thermal-hydraulic performance of the target/core was calculated and the engineering feasibility of the major components was assessed. The molten-salt system with chloride MA fuel is another attractive option for accelerator-based transmutation. Its main advantage is the capability of continuous on-line chemical processing of MAs and reaction products.

As compared with usual nuclear reactors, an accelerator-based system does not require a critical condition, and therefore it has the significant advantage of large criticality safety margin. The thermal power of the subcritical core can be easily controlled by adjusting the power of incident proton beam.

The development of spallation simulation code system and the spallation integral experiments are being conducted to improve the methods and data for accelerator-based transmutation.
1) CONCEPT OF SOLID TARGET/CORE SYSTEM

A conceptual design study has been made on an accelerator-based MA transmutation plant with a sodium-cooled solid target/core[3]. Schematic diagram of the solid system is presented in Fig. 3. A 1.5 GeV proton beam is injected through the beam window into the tungsten target located at the center of the MA-loaded subcritical core. Spallation neutrons from the target induce fission reactions in the MA fuel. Heat generated in the target and fuel is removed by primary sodium, then transported through secondary loop to the power conversion system.

The fuel compositions are Np-15Pu-30Zr and AmCm-35Pu-10Y. These alloy systems are proposed for a sodium-cooled MA burner reactor[4]. Fig. 4 shows the fuel subassembly. To ensure adequate fuel cooling during the out-of-core handling operation, measures are incorporated in the subassembly design.
The target subassembly is depicted in Fig. 5. From the nuclear and thermal-hydraulic points of view, a configuration of multi-disk type was chosen. Thicknesses of the tungsten disks are varied in the axial direction to obtain a uniform distribution of spallation neutron flux. Coolant holes in the disk are arranged to stagger from layer to layer to prevent the penetration of high energy particles through the target.

The fuel and target subassemblies are cooled by forced upward flow of sodium. The whole target/core including reflectors is contained within a steel reactor vessel as shown in Fig. 6. The vertical tube for beam path is inserted into the reactor vessel down to just above the target. The bottom end of the tube forms the beam window. The window is cooled by a sodium flow from the target exit.
Heat transport and power conversion systems are based on the current design practice for a sodium-cooled fast breeder reactor plant, as shown in Fig. 7. A primary reactor auxiliary system with NaK loops is provided as an independent means of removing core decay heat.

With a 1.5 GeV-39 mA proton beam, the target/core having an effective neutron multiplication factor of 0.89 produces 820-MW thermal power. The maximum thermal power of the system is limited by the maximum allowable fuel temperature of 1173 K. The maximum power densities in the fuel and target regions are calculated to be about 930 W/cc and 360 W/cc, respectively. Assuming a load factor of 80%, the MA burnup (fission) is approximately 250 kg/y, or 8%. The result shows that the system can support about 10 units of 1000-MWe light water reactor (LWR). An electric output of 246 MW is obtained at a plant thermal efficiency of 30%. The power required to operate the 1.5 GeV-39 mA proton accelerator is 100 MW, assuming a 40% efficiency. This means that the system is more than self-sufficient in terms of its own energy balance.

Table 1 summarizes design parameters and operating condition of the solid system.

2) CONCEPT OF MOLTEN-SALT TARGET/CORE SYSTEM

A preliminary conceptual design study is being performed on an 800-MWt molten-salt core/target system as an advanced option for an accelerator-based nuclear waste transmutation system[5]. Chloride salt with a composition of 64NaCl-5PuCl_3-31MACl_3 is chosen for the molten-salt system based on the consideration about actinide solubility and nuclear characteristics. The molten salt acts both as fuel and as target material, and at the same time it also serves as coolant in the molten-salt system. This significantly simplifies the core/target system configuration, as schematically shown in Fig. 8.

High energy proton beam at 1.5 GeV is injected into the central core/target region through the beam window. The core/target region is surrounded by an internal reflector. Intermediate heat exchangers and salt pumps are installed in the annular region around the
Table 1  Design Parameters and Operating Condition of Solid Target/Core

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fuel</td>
<td>Metal Alloy</td>
</tr>
<tr>
<td></td>
<td>Np-15Pu-30Zr</td>
</tr>
<tr>
<td></td>
<td>AmCm-35Pu-10Y</td>
</tr>
<tr>
<td>Target</td>
<td>Solid Tungsten</td>
</tr>
<tr>
<td>Primary Coolant</td>
<td>Liquid Sodium</td>
</tr>
<tr>
<td>Actinide Inventory</td>
<td>3160 kg</td>
</tr>
<tr>
<td>Multiplication Factor</td>
<td>0.89</td>
</tr>
<tr>
<td>Spallation Neutrons</td>
<td>40 n/p</td>
</tr>
<tr>
<td>Proton Beam</td>
<td>1.5 GeV-39 mA</td>
</tr>
<tr>
<td>Thermal Power</td>
<td>820 MW</td>
</tr>
<tr>
<td>Burnup</td>
<td>250 kg/y (8.0 %/y)</td>
</tr>
<tr>
<td>Power Density, Max./Avg.</td>
<td>930/400 MW/m³</td>
</tr>
<tr>
<td>Temperature, Core Inlet/Outlet</td>
<td>330/430 °C</td>
</tr>
<tr>
<td>Coolant Maximum Velocity</td>
<td>8 m/s</td>
</tr>
</tbody>
</table>

internal reflector. This in-vessel heat exchanger design minimizes the total MA inventory in the system.

The molten-salt target/core has an effective multiplication factor of 0.92, and produces 800-MW thermal power with a 1.5 GeV-25 mA proton beam. In the molten-salt system, the maximum thermal power is limited not by the maximum fuel temperature or the maximum power density, but heat transfer rate through the heat exchangers. The maximum power density in the target/core region is about 1660 W/cc. Assuming a load factor of 80%, the MA
burnup is approximately 250 kg/y, or 4.6%/y. The molten-salt system can support about 10 units of 1000-MWe LWR. The system operates at molten-salt temperature in the range of 920-1020 K. With such a high operating temperature, a power conversion efficiency around 45% becomes feasible by using a supercritical steam cycle, which can improve the total energy balance of the system.

Table 2 summarizes design parameters and operating condition of the molten-salt system.

<table>
<thead>
<tr>
<th>Table 2 Design Parameters and Operating Condition of Molten-Salt Target/Core</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fuel</td>
</tr>
<tr>
<td></td>
</tr>
<tr>
<td>(MA : Np, Am, Cm)</td>
</tr>
<tr>
<td>Target</td>
</tr>
<tr>
<td>Primary Coolant</td>
</tr>
<tr>
<td>Actinide Inventory</td>
</tr>
<tr>
<td>Multiplication Factor</td>
</tr>
<tr>
<td>Spallation Neutrons</td>
</tr>
<tr>
<td>Proton Beam</td>
</tr>
<tr>
<td>Thermal Power</td>
</tr>
<tr>
<td>Burnup</td>
</tr>
<tr>
<td>Power Density, Max./Avg.</td>
</tr>
<tr>
<td>Temperature, Core Inlet/Outlet</td>
</tr>
<tr>
<td>Coolant Maximum Velocity</td>
</tr>
</tbody>
</table>

Molten state of fuel salt offers several attractive features for the design of the transmutation systems. The main advantage over the solid system is the capability of the continuous on-line separation of fission products and spallation products from the fuel. Furthermore, rather laborious process of actinide fuel fabrication is not required for the molten-salt system. Core melt-down accidents can be impossible as the molten fuel is ready to be dumped from the core in case of emergency, which may add a high degree of safety.

Schematic diagram of continuous on-line processing in the molten-salt system is presented in Fig. 9.

3) CODE DEVELOPMENT AND SPALLATION EXPERIMENTS

A computer code system has been continuously developed for the nuclear design of the accelerator-based transmutation system[6]. In the code system, NMTC/JAERI code[7] simulates the proton-induced nuclear spallation and subsequent internuclear transport process for energies above 15 MeV. It also calculates high energy fission reaction as a competing process with evaporation. Neutronic calculation below 15 MeV is carried out using MORSE-DD and TWOTRAN-2 codes. The time evolution process of transmutation products is calculated by SPCHAIN code in the higher energy range above 15 MeV and by ORIGEN-2 code below 15 MeV. The code system has been upgraded and improved by incorporating current models and methods.
Spallation integral experiments have been carried out to obtain data on nuclide production, to estimate the yields of neutrons and spallation products, and to evaluate the validity of the simulation code system[8]. The 500-MeV booster proton synchrotron facility at the National Laboratory for High Energy Physics (KEK) is used for the experiments. Proton beam was injected into a cylindrical assembly made of lead, with a diameter of 600 mm and a length of 1000 mm. A lead or tungsten target was installed in the center of the lead assembly. The activation samples are Al, Fe, Ni, and Cu cylinders with dimensions of 6 mm in diameter and 4-10 mm in length. These samples encapsulated in 8-mm diameter metallic cans were inserted in the 10-mm diameter holes drilled through the assembly along the beam axis at various radial positions. The numbers of induced reactions in the samples were obtained by measuring γ-rays with a 100 cc Ge-detector. Experimental results were compared with the prediction by NMTC/JAERI code. An example of the comparison between the experimental result and the calculation is shown in Fig. 10. The calculation generally agrees with the measured ones well except for the results on the axis beyond 20 cm in depth.

3.2 HIGH INTENSITY PROTON LINEAR ACCELERATOR

The construction of a high-intensity proton linear accelerator called the Engineering Test Accelerator (ETA, 1.5GeV and 10mA) has been proposed by JAERI. In the first step of the accelerator development, the four main accelerator components, ion source, radio-frequency quadrupole (RFQ), drift-tube linac (DTL) and radio-frequency (RF) source, for the Basic Technology Accelerator (BTA, 10MeV and 10mA), have been developed. The conceptual and optimization studies for the ETA have been simultaneously performed concerning proper choice of operating frequency, high β structure, mechanical engineering consideration and RF source aspect in order to ensure low beam loss, hands-on maintenance and low construction cost.
The conceptual layout of the ETA is shown in Fig. 11. In the case of high intensity accelerator, it is particularly important to obtain well focused beam which minimizes beam losses to avoid damage and activation of the accelerator structures in addition to the requirement for the maximum beam current. The beam quality and maximum current are mainly determined considering the construction of the low energy portion of the accelerator, BTA[9]. The basic specifications of the BTA are given in Table 3. The acceleration frequency of 201.25 MHz was selected both for RFQ and DTL mainly due to the relatively manageable heat removal problem and the availability of RF source. Because of the high beam current and high duty factor, heat removal problem from the accelerator structures is an important issue for the mechanical design.

A. Ion source

Fig. 11 shows a prototype of an ion source[10] which was constructed in collaboration with the group of the NBI (Neutral Beam Injectors) Heating Laboratory for the Fusion Research of JAERI. The ion source consists of a multicusp plasma generator with four tungsten filaments and a two stage extractor. The basic specifications for the source are given in Table 4.
Table 3 The Basic Specifications of the BTA

<table>
<thead>
<tr>
<th>Operation mode</th>
<th>pulse</th>
</tr>
</thead>
<tbody>
<tr>
<td>Duty factor</td>
<td>10%</td>
</tr>
<tr>
<td>Output energy</td>
<td>10MeV</td>
</tr>
<tr>
<td>Average beam current</td>
<td>10mA</td>
</tr>
<tr>
<td>Peak beam current</td>
<td>100mA</td>
</tr>
</tbody>
</table>

Fig. 11 A Conceptual Layout of the ETA, and the BTA (a front end portion of the ETA)

Table 4 The High Brightness Ion Source Parameters

<table>
<thead>
<tr>
<th>Energy</th>
<th>100keV</th>
</tr>
</thead>
<tbody>
<tr>
<td>Current</td>
<td>120mA</td>
</tr>
<tr>
<td>Duty factor</td>
<td>CW</td>
</tr>
<tr>
<td>Emittance</td>
<td>0.5πmm.mrad</td>
</tr>
<tr>
<td></td>
<td>(normalized 100%)</td>
</tr>
<tr>
<td>Proton ratio</td>
<td>&gt;90%</td>
</tr>
<tr>
<td>Impurity</td>
<td>&lt;1%</td>
</tr>
</tbody>
</table>

The high brightness hydrogen ion of 140mA was extracted using the 100kV high voltage power supply. The beam profile was measured with a multi-channel calorimeter and the observed normalized emittance was about 0.45 πmm.mrad(90%). The hydrogen \( \text{H}^+ \) beam current as a function of acceleration voltage (perveance curve) is shown in Fig. 13 as well as the helium beam current. The proton ratio and impurity were 80% and less than 1%, respectively, which were obtained by a Doppler shifted spectroscopy method.

B. RFQ

The design study for the RFQ has been made for a four vane type resonator [11]. The tank was constructed with copper-plated carbon steal. Water flow through each channel is 30l/m and the operating average temperature is 25.5°C. The calculated maximum temperature
Fig. 12 A Cross-Sectional View of the Ion Source for the BTA

Fig. 13 Observed Beam Current as a Function of Acceleration Voltage. The open circles and closed ones show the hydrogen and helium optimum beam current, respectively, which give the minimum beam divergence.
is 39.1°C, and displacement in the transverse and longitudinal directions are 33μm and 99μm, respectively. These deformations are small enough both for the frequency tuning and for the beam dynamics.

The machining of the vanes and RFQ tank have been completed and the RF coupler and vacuum devices were attached to the tank by the Sumitomo Heavy Industries, LTD.. The measured Q value was 8,930 which is 67% of the calculated value with the SUPERFISH code. The electromagnetic field measurements were made by the bead perturbation method resulting in the field balance of ±2.5% and the flatness of ±7.1% in each quadrant. The high-power conditioning is carried out with the 1 MW RF source described below. The RF processing is being performed with the vacuum pressure level below 1x10⁻⁶ Torr at a low duty operation of a few% and about 200 kW RF power. The thermal detuning is compensated with four movable tuners installed in each quadrant. The duty factor will be increased step by step up to 12%.

C. DTL

A resonant frequency, magnetic field strength and heat removal problem were investigated for the DTL under the various mechanical constraints. A hollow conductor type coil with 5x5mm² was chosen for focusing magnet, of which field was 80T/m with 5.5 turns of 780 amperes. Configuration of quadrupole magnet was optimized under the condition on the coolant water (temperature rise 25°C in the coil, pressure drop 5kgf/cm² and velocity 3.4m/s). The DTL parameters are given in Table 5.

The hot test model with 9 cells was fabricated by the Mitsubishi Heavy Industries, LTD.. In the #1 drift tube and the one at the front end plate among the 9 drift tube cells, actual quadrupole magnets were installed. The magnetic field strength and center position, and higher harmonic field components were measured with the conventional rotating search coil technique. The electric field distribution on the beam axis was measured with the bead

<table>
<thead>
<tr>
<th>Table 5 The DTL Parameters</th>
</tr>
</thead>
<tbody>
<tr>
<td>Frequency</td>
</tr>
<tr>
<td>Energy</td>
</tr>
<tr>
<td>Beam current</td>
</tr>
<tr>
<td>Average field</td>
</tr>
<tr>
<td>Tank diameter</td>
</tr>
<tr>
<td>Tank length</td>
</tr>
<tr>
<td>Cell length</td>
</tr>
<tr>
<td>g/L</td>
</tr>
<tr>
<td>DT outer diameter</td>
</tr>
<tr>
<td>DT inner diameter</td>
</tr>
<tr>
<td>Synchronous phase</td>
</tr>
<tr>
<td>DT cell number</td>
</tr>
<tr>
<td>Focus magnetic field</td>
</tr>
<tr>
<td>Q</td>
</tr>
<tr>
<td>Wall loss</td>
</tr>
<tr>
<td>Beam power</td>
</tr>
<tr>
<td>Emittance</td>
</tr>
<tr>
<td>x(rms)</td>
</tr>
<tr>
<td>y(rms)</td>
</tr>
<tr>
<td>z(rms)</td>
</tr>
</tbody>
</table>

90
perturbation method. The observed average accelerating field distribution was in good agreement with the SUPERFISH calculation within an accuracy of ±1.5%. The measured Q value is 42,650 which is 84% of the SUPERFISH calculation.

D. RF source

Three sets of 201.25MHz RF sources with one MW peak class amplifier are required (641kW for RFQ and two 760kW for DTL) for the BTA. The tetrode tube 4CM2500KG (EIMAC), which was originally developed for fusion plasma heating, is used with multistage amplifier configuration. The RF source was designed and one set of the amplifier was manufactured by the Sumitomo Heavy Industries, LTD.. The high power amplifier (HPA) is driven by a 60kW intermediate amplifier (IPA of RS2058CJ) which is fed by a master oscillator and a 3kW solid state drive amplifier. The accelerator voltage and phase control loop with an accuracy of less than 0.1% in amplitude and less than 1° in phase are prepared. The high power RF conditioning for HPA was made achieving the 1MW (peak) output power with a low duty operation (about 1%), and then it was gradually increased up to 12% with 700 kW. The specifications of the RF source are given in Table 6.

Table 6 The Specifications of the RF Source

<p>| | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Frequency</td>
<td>201.25MHz</td>
</tr>
<tr>
<td>Duty factor</td>
<td>12%</td>
</tr>
<tr>
<td>Duration</td>
<td>1.2ms</td>
</tr>
<tr>
<td>Repetition rate</td>
<td>100pps</td>
</tr>
<tr>
<td>Low level amplifier</td>
<td>Solid state (5kW)</td>
</tr>
<tr>
<td>Intermediate power amplifier</td>
<td>Tetrode (60kW)</td>
</tr>
<tr>
<td>High power amplifier</td>
<td>Tetrode (1MW)</td>
</tr>
</tbody>
</table>

E. High β linac and optimization study

The optimization study is important for the actual transmutation plant. The requirement for the accelerator is dependent on the conceptual study of the transmutation target/core. The main plant conditions which will determine the range of the proton beam and current (beam power) are transmutation rate per year and output power density in the target/core. The two major parameters (accelerator length and RF power requirement) will determine the cost of the accelerator construction. The length of the accelerator is simply determined by the accelerating field strength. The required RF power is a function of the field strength, accelerator frequency, shunt impedance of the structure (efficiency) and so on. The trade-off-studies for the various accelerator parameters will be further continued to obtain the optimized system.

4. SUMMARY

The background of OMEGA Program and the research and development areas under OMEGA were overviewed. The program is conceived as a research effort to pursue benefits for future generations through the long term basic R&D, and is not to seek a short term alternative for established or planned fuel cycle back-end policies.

The R&D activities at JAERI under OMEGA include the development of a four group partitioning method, a conceptual design study of MA burner reactors with very hard
neutron spectrum, an accelerator-based transmutation system, the development of a high intensity proton accelerator, and the related nuclear and fuel property data measurements.

A conceptual design study of an accelerator-based MA transmutation plant with a sodium-cooled solid target/core has been carried out. The proposed plant transmutes about 250 kg of MA per year with a 1.5 GeV, 39 mA proton accelerator and generate enough electricity for the accelerator. The molten-salt system of chloride MA fuel has been also examined as an another transmutation option with a capability of continuous on-line chemical processing. Simulation code development and spallation integral experiments have been made to improve the method and data for the design of accelerator-based transmutation systems.

The R&D works with the design and construction of prototype accelerator structures (Ion source, RFQ, DTL and RF source) for high power test (hot model) are in progress. For the high power test, measurements of the electric and magnetic characteristics of the accelerator structure are being made with the single unit of high power RF source. Problems of heat dissipation and heat removal in the structure are studied. The beam test up to 2MeV in combination with the ion source, RFQ and RF source is scheduled in the fiscal 1993. The detailed design works for the BTA construction are followed in the next stage based on the results of the R&D works.

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10. Y. Okumura et al., "Development of A High Brightness Ion Source for the Proton Linear Accelerator (BTA) at JAERI", ibid. p.643-647
RESEARCH AND DEVELOPMENT ON THE PARTITIONING PROCESS AT JAERI

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Abstract

A wet partitioning process has been developed in Japan Atomic Energy Research Institute (JAERI) for separating elements in high-level waste (HLW) into four groups; transuranium elements (TRU), Tc-platinum group metals (PGM), Sr-Cs and the other elements. Extraction with diisodecylphosphoric acid (DIDPA) was studied for the separation of TRU from HLW. Trivalent actinides, Am and Cm, can be extracted with DIDPA from the solution of around 0.5 M nitric acid. In addition, Np that is initially in the pentavalent state can be extracted. Experiments on counter-current continuous extraction of Np from a simulated HLW showed that more than 99.96% of Np was recovered with 14-stages extraction and with the addition of hydrogen peroxide. Since the Np extracted in the presence of hydrogen peroxide is in the tetravalent state, it is easily stripped with oxalic acid. Selective stripping of Am and Cm from DIDPA with diethylenetriaminepentaacetic acid (DTPA) is being studied for their separation from rare earths (RE). Batch experiments showed that the separation factor between Am and RE (ratio of distribution ratios) was more than 10. Continuous experiments are now in progress to find the optimum process condition. These results offer a promising prospect of separating TRU by the DIDPA extraction process that consists of the four steps; simultaneous extraction of TRU with DIDPA, first stripping of Am and Cm with DTPA, second stripping of RE with 4 M nitric acid and third stripping of Np and Pu with oxalic acid. For the separation of Tc-PGM, two methods are studied; one is adsorption with an active carbon and the other is precipitation with reducing the acidity by denitrification. The pretreatment of HLW for the DIDPA extraction is also being studied.

INTRODUCTION

Partitioning of high-level waste (HLW) has been studied in Japan Atomic Energy Research Institute since 1973. A wet partitioning process presently being developed includes the separation of transuranium elements (TRU), Tc, platinum group metals (PGM), Sr and Cs [1]. Separated TRU can be used as a fuel and be transmuted to shorter-lived or stable nuclides. Tc and PGM are valuable elements. By immobilising the Sr and Cs into thermally and chemically stable form, we can use them as heat or radiation sources and reduce the total volume of HLW in solid form.

In 1983, a partitioning process developed for the separation of TRU (except Np) [2], Sr and Cs was tested with an actual HLW (1.2 L, 200 Ci) [3]. The process consists of three steps; extraction of U and Pu with TBP, extraction of Am and Cm with diisodecylphosphoric acid (DIDPA), (In the following step, Am and Cm are separated from rare earths (RE) by cation exchange method.), adsorption of Sr and Cs with titanic acid and zeolite. In the hot test, more than 98% of Pu were extracted with TBP, and more than 99.99% of Am and Cm were extracted with DIDPA [3]. After the hot test the study on an advanced partitioning process was started, which includes the separation of Np, Tc and PGM. The present paper describes the recent studies on this subject.

SEPARATION OF TRANSURANIUM ELEMENTS, ESPECIALLY NEPTUNIUM

Batch experiments showed that Np initially in the pentavalent state was extracted with DIDPA [4] that was used successfully as a extractant for Am and Cm in the partitioning test with an actual HLW. The extraction rate of Np is not fast enough in a practical point of view.
However, it is increased by the addition of hydrogen peroxide [5]. On the basis of these results, experiments on a counter-current continuous extraction using a mixer-settler were carried out to determine the necessary and optimum condition for the Np extraction [6,7].

Figure 1 shows the operating condition of a 16-stages miniature mixer-settler used for the Np extraction. It was operated at 25 or 45°C in a thermostatted air bath under conditions (a), 7-stages extraction, and condition (b), 14-stages extraction.

Table 1 shows the experimental condition of the continuous extraction and the ratio of Np extracted. The ratio was increased, which means the extraction rate became faster, by raising temperature and by adding hydrogen peroxide. The Np extraction from the simulated HLW was further accelerated due to the catalytic effect of some elements in the HLW.

Figure 2 shows concentration profiles of Np in the aqueous phase at extraction section in the Runs No. 14 and 15. The rate of the Np extraction was faster at the first stage, but became slow stage by stage. The decrease of the extraction rate was due to the decomposition of hydrogen peroxide. However, by adding hydrogen peroxide to compensate its decomposition at the stages pointed by downward arrows in Fig. 2, the rate increased again. Finally, 99% of Np was extracted in 7-stages extraction (Run No. 14), and more than 99.96% was recovered in 14-stages extraction (Run No. 15). A consideration of the optimum condition for the Np extraction showed that 9 min of total contacting time of the two phases would give more than 99.96% recovery of Np in 12-stages extraction when hydrogen peroxide was added in every 3-stages at the same concentration as in the experiment.

A cold experiment to examine the behaviors of fission and corrosion products in counter-current continuous extraction with DIDPA and stripping with 4 M nitric acid was carried out at 45°C with addition of hydrogen peroxide to scrub solution [7]. Table 2 shows...
Table 1  Experimental condition of the continuous counter-current extraction of Np and the ratio of Np extracted.

<table>
<thead>
<tr>
<th>Run No.</th>
<th>Temp. (°C)</th>
<th>Feed solution</th>
<th>Extracting stages</th>
<th>H$_2$O$_2$ in scrub (M)</th>
<th>H$_2$O$_2$ Addition Stage</th>
<th>Extracted (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>25</td>
<td>Np alone</td>
<td>7</td>
<td>0</td>
<td>-</td>
<td>24.6</td>
</tr>
<tr>
<td>2</td>
<td>45</td>
<td>Np alone</td>
<td>7</td>
<td>0</td>
<td>-</td>
<td>45.4</td>
</tr>
<tr>
<td>3</td>
<td>25</td>
<td>Np alone</td>
<td>7</td>
<td>0.5</td>
<td>-</td>
<td>78.8</td>
</tr>
<tr>
<td>4</td>
<td>45</td>
<td>Np alone</td>
<td>7</td>
<td>0.5</td>
<td>-</td>
<td>92.7</td>
</tr>
<tr>
<td>9</td>
<td>25</td>
<td>Sim. HLW</td>
<td>7</td>
<td>0.5</td>
<td>-</td>
<td>92.1</td>
</tr>
<tr>
<td>10</td>
<td>45</td>
<td>Sim. HLW</td>
<td>7</td>
<td>0.5</td>
<td>3</td>
<td>95.6</td>
</tr>
<tr>
<td>12</td>
<td>25</td>
<td>Sim. HLW</td>
<td>7</td>
<td>0.5</td>
<td>3</td>
<td>95.5</td>
</tr>
<tr>
<td>13</td>
<td>45</td>
<td>Sim. HLW</td>
<td>7</td>
<td>0.5</td>
<td>3</td>
<td>98.2</td>
</tr>
<tr>
<td>14</td>
<td>45</td>
<td>Sim. HLW</td>
<td>7</td>
<td>1.0</td>
<td>3 &amp; 7</td>
<td>99.0</td>
</tr>
<tr>
<td>15</td>
<td>45</td>
<td>Sim. HLW</td>
<td>14</td>
<td>1.0</td>
<td>3 &amp; 7</td>
<td>&gt;99.96</td>
</tr>
</tbody>
</table>

Fig. 2  Concentration profile of Np in aqueous phase at the extraction section in Runs No.14 and No.15. Downward arrows show the points where H$_2$O$_2$ solutions are added.
Run (14) (7-stages) : 99.0% recovery
Run (15) (14-stages) : >99.96% recovery
Table 2 Behaviors of fission and corrosion products in the continuous counter-current extraction carried out on the same condition as in Run No.10.
- Ratios (%) of each element in the three fractions.

<table>
<thead>
<tr>
<th>Element</th>
<th>Raffinate</th>
<th>4 M HNO₃ (Product I)</th>
<th>Organic Fraction</th>
</tr>
</thead>
<tbody>
<tr>
<td>Na</td>
<td>&gt;99.6</td>
<td>0.31</td>
<td>&lt;0.1</td>
</tr>
<tr>
<td>Cr</td>
<td>99.5</td>
<td>0.38</td>
<td>&lt;0.1</td>
</tr>
<tr>
<td>Fe</td>
<td>0.13</td>
<td>0.1</td>
<td>99.77</td>
</tr>
<tr>
<td>Ni</td>
<td>&gt;99.5</td>
<td>0.44</td>
<td>&lt;0.1</td>
</tr>
<tr>
<td>Rb</td>
<td>&gt;99.97</td>
<td>&lt;0.02</td>
<td>&lt;0.01</td>
</tr>
<tr>
<td>Cs</td>
<td>&gt;99.99</td>
<td>&lt;0.005</td>
<td>&lt;0.002</td>
</tr>
<tr>
<td>Sr</td>
<td>&gt;99.95</td>
<td>0.042</td>
<td>&lt;0.004</td>
</tr>
<tr>
<td>Ba</td>
<td>&gt;99.94</td>
<td>0.027</td>
<td>&lt;0.04</td>
</tr>
<tr>
<td>Ru</td>
<td>93.53</td>
<td>0.98</td>
<td>5.49</td>
</tr>
<tr>
<td>Rh</td>
<td>88.30</td>
<td>1.86</td>
<td>9.84</td>
</tr>
<tr>
<td>Pd</td>
<td>&gt;99.5</td>
<td>0.43</td>
<td>&lt;0.02</td>
</tr>
<tr>
<td>Nd</td>
<td>0.002</td>
<td>&gt;99.99</td>
<td>&lt;0.006</td>
</tr>
</tbody>
</table>

Ratios (%) of each element in the three fractions. It was confirmed more than 99.99% of Nd, which is a representative of Am, Cm and RE, was recovered as Product I. The other element that was quantitatively extracted was Fe. However, since Fe was not stripped with 4 M nitric acid, it can be separated from Nd with a high decontamination factor (about 1000).

About 6% of Ru and 11% of Rh were extracted in this experiment. The other elements including Sr and Cs were not extracted and were transferred to the raffinate with more than 99.5% yield ( >99.95% for Sr and >99.99% for Cs) as shown in Table 2. Decontamination factors of Sr and Cs in Product I (Am, Cm and RE) were 2400 and more than 20000 respectively.

An experiment on counter-current continuous stripping of Np showed that more than 99.9% of Np was recovered with 0.8 M oxalic acid solution from the DIDPA solution in 12 stages [7]. Since Np in the organic phase, extracted in the presence of hydrogen peroxide, is in the tetravalent state and the Np distributed itself between two phases without the change of its oxidation state, the Np behavior in the continuous stripping can be easily explained with distribution ratio of tetravalent Np, obtained through the batch experiment.

Extracted Fe is stripped with the oxalic acid solution together with Np. As for the other two extracted elements, Rh was stripped up to about 98%, but ratio of Ru stripped was only 20%.

The present experiments on the counter-current continuous extraction and stripping offered a promising prospect of separating TRU from HLW by the DIDPA extraction process that consisted of the three steps; the simultaneous extraction of actinides with the DIDPA solvent, the stripping of trivalent actinides, Am and Cm (and RE), with 4 M nitric acid, and the stripping of tetravalent actinides, Np and Pu, with oxalic acid. In this process, an additional step is required to separate Am and Cm from RE.
In the partitioning process developed previously [3], Am and Cm were separated from RE by cation exchange method after the stripping with 4 M nitric acid. In order to develop a more continuous process, selective stripping of Am and Cm from the DIDPA solvent is now being studied using diethylenetriaminepentaacetic acid (DTPA) as a complexing agent.

Figure 3 shows the pH dependence of the distribution ratio of Am and Nd in the stripping from DIDPA with 0.05 M DTPA - 1 M lactic acid solution of which pH was adjusted by ammonium ion. Separation factor, which is the ratio of distribution ratio, was more than 10. Figure 4 shows the comparison of distribution ratio of Am with those of RE. Distribution ratio of Sm was the nearest to that of Am.

One of the important factors in this method is the change of pH in the DTPA solutions, which is caused by the extraction of the ammonium ion. Since the extraction of ammonium ion is determined by its distribution ratio at equilibrium, we can predict the profile of pH in a continuous extractor. In a preliminary experiment on continuous stripping using only Nd at low concentration, the pH profile obtained in the experiment agreed with the profile calculated from the distribution ratio of ammonium ion.
Fig. 4 Comparison of Am Distribution Ratio with Those of Rare Earths.
Org.: 0.5 M DIDPA - 0.1 M TBP in n-dodecane
Aq.: 0.05 M DTPA - 1 M Lactic acid - NH₄OH, pH 3.6 after stripping

From these results we estimated the behavior of each element in continuous stripping. Table 3 shows the decontamination factor, DF, of RE from Am and Cm. The DF of Sm is the lowest but would be able to increase up to 90. Overall DF would be 430, which is comparable to that in cation exchange method. With this DF we could obtain the Am-Cm product of about 90% purity in molar ratio (about 93% in weight ratio). We should examine the actually obtainable DF by performing continuous counter-current stripping, which is now in progress.

Figure 5 shows the flow sheet for TRU separation by extraction with DIDPA followed by selective stripping. After simultaneous extraction of actinides, Am and Cm are stripped with DTPA and then RE are stripped with 4 M nitric acid.

PRETREATMENT OF HLW FOR THE DIDPA EXTRACTION

Before the extraction with DIDPA, nitric acid concentration in HLW should be reduced to about 0.5 M. Denitration with formic acid has been studied for this purpose. One of the advantages of the denitration is that Mo and Zr, which is extracted with DIDPA, are removed as a precipitate from HLW [8]. In the recent study [9], precipitation behavior of TRU (Pu, Np and Am) during the denitration was examined using a simulated HLW. Table 4 shows the dependency of the TRU precipitation on the pH after denitration. The percentages of precipitated TRU increased with decreasing acidity of the denitrated HLW. In the denitration
at $[\text{HCOOH}]/[\text{HNO}_3] = 1.5$, which gives the reduction of the nitric acid concentration to about 0.5 M, the precipitated fractions of Np and Am were only 0.6% and 0.06%, respectively, whereas that of Pu was 93%. Although the mechanism of the Pu precipitation is not clearly known, Pu seems to coprecipitate with Mo and Zr.

Heating without addition of formic acid, which is the simulation of the storage of HLW, also resulted in the formation of a precipitate that included Pu. Therefore, the precipitation of Pu cannot be avoided even if HLW is not denitrated.

Dissolution of the precipitate was also studied using oxalic acid to recover TRU from the precipitate. The precipitate formed in 1 L of the simulated HLW (500 L/1 ton of spent fuel) could be completely dissolved in 0.8 L of 0.5 M oxalic acid solution.

Dilution is an alternative to reduce the nitric acid concentration and was found to cause no additional precipitation. Dilution would be applicable to the pretreatment of HLW for DIDPA extraction. However, it suffers a serious disadvantage. The dilution makes the volume of HLW to be treated in a partitioning process 8 times or more, which increases the cost of partitioning.

Filtration characteristics of the precipitates were also studied using a simulated HLW. It was found that both precipitates formed by heating the HLW and by denitrating the HLW that was previously heated for a long time could be easily filtrated.

Table 3  Separation of Am and Cm from RE. Preliminary Estimation of Decontamination Factor (DF) and Concentration Ratio of RE after Separation.

<table>
<thead>
<tr>
<th>Element</th>
<th>In HLW 1)</th>
<th>Cation Exchange 2)</th>
<th>Extraction</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Conc.(mM)</td>
<td>Ratio 3)</td>
<td>DF</td>
</tr>
<tr>
<td>Am,Cm</td>
<td>3.23</td>
<td>1.00</td>
<td>-</td>
</tr>
<tr>
<td>Y</td>
<td>10.36</td>
<td>3.21</td>
<td>$&gt;10^6$</td>
</tr>
<tr>
<td>La</td>
<td>17.53</td>
<td>5.43</td>
<td>$&gt;10^6$</td>
</tr>
<tr>
<td>Ce</td>
<td>33.80</td>
<td>10.46</td>
<td>$&gt;10^6$</td>
</tr>
<tr>
<td>Pr</td>
<td>15.86</td>
<td>4.91</td>
<td>$&gt;10^6$</td>
</tr>
<tr>
<td>Nd</td>
<td>55.62</td>
<td>17.22</td>
<td>$&gt;10^6$</td>
</tr>
<tr>
<td>Pm</td>
<td>0.52</td>
<td>0.16</td>
<td>$&gt;10^6$</td>
</tr>
<tr>
<td>Sm</td>
<td>11.17</td>
<td>3.46</td>
<td>$&gt;10^6$</td>
</tr>
<tr>
<td>Eu</td>
<td>1.83</td>
<td>0.57</td>
<td>$&gt;10^3$</td>
</tr>
<tr>
<td>Gd-Er</td>
<td>1.28</td>
<td>0.40</td>
<td>3</td>
</tr>
<tr>
<td>Total</td>
<td>148.0</td>
<td>45.82</td>
<td>(350)</td>
</tr>
</tbody>
</table>

1) Calculated on condition that 0.5 m$^3$ of HLW is generated per 1 t of spent fuel which is burned up to 33000 MWD/t and reprocessed after 5-years cooling.
2) The method previously developed.
3) Ratio of RE concentration to the total concentration of Am and Cm.
4) The concentration ratio after separation.
5) DF by the extraction is depend on the process condition. These value are obtained as typical ones by calculation on condition that 99.99% of Am and Cm are recovered.
Fig. 5 Process Flow Sheet for TRU Separation by Extraction with DIDPA Followed by Selective Stripping

SEPARATION OF TECHNETIUM AND PLATINUM GROUP METALS

The separation of Tc and additionally PGM was studied by the adsorption method with active carbon [10, 11]. Figure 6 shows break through curves of Tc and PGM in the absorption from a simulated HLW. Tc and Pd were effectively adsorbed on the column, but Rh was adsorbed only a little. The curve for Ru suggests the presence of several chemical species.

Figure 7 shows the elution curves with an alkaline potassium thiocyanate solution. Complete elution of Tc was achieved but Pd was not eluted quantitatively.
Table 4  pH and Percentage of TRU precipitated during denitrification of simulated HLW

<table>
<thead>
<tr>
<th>[HCOOH]/[HNO₃]</th>
<th>pH</th>
<th>Percentage of TRU precipitated</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Pu</td>
</tr>
<tr>
<td>0.0</td>
<td>-0.4</td>
<td>14</td>
</tr>
<tr>
<td>1.0</td>
<td>0.1</td>
<td>27</td>
</tr>
<tr>
<td>1.2</td>
<td>0.3</td>
<td>31</td>
</tr>
<tr>
<td>1.4</td>
<td>0.4</td>
<td>76</td>
</tr>
<tr>
<td>1.5</td>
<td>0.6</td>
<td>93</td>
</tr>
<tr>
<td>1.6</td>
<td>1</td>
<td>97</td>
</tr>
<tr>
<td>1.8</td>
<td>3</td>
<td>98</td>
</tr>
<tr>
<td>2.0</td>
<td>5</td>
<td>100</td>
</tr>
</tbody>
</table>

Fig. 6  Break Through Curves of Tc and PGM in Adsorption with an Active Carbon Column.

Column: 1 g active carbon, 150 mm long, 7.5 mm diameter
Feed : 0.5 M HNO₃ solution, 5 bed volumes/h

Table 5 shows the summary of the column experiment. More than 98% of Tc was recovered by the present method, adsorption with active carbon and elution with thiocyanate. Pd was also adsorbed quantitatively but only 18% was eluted. Rh could not be separated by this method. Further studies will be required particularly on the elution of Pd. Consequently this adsorption method is effective and applicable for the Tc separation but not for the PGM separation.

Precipitation method was also studied as an alternative for the separation of Tc and PGM. By reducing the acidity to above pH 2 by denitration, more than 95% of Tc, Pd and Ph and about 80% of Ru was recovered as a precipitate from a simulated HLW [1,12,13].
Fig. 7 Elution of Tc and PGM from the Active Carbon Column with an Alkaline KSCN Solution.
Column: Obtained by the adsorption experiments shown in Fig. 6
Eluate : 2 M KSCN - 4 M NaOH solution, 5 bed volumes/h

Table 5 Results of Experiment on Adsorption with Active Carbon and Elution with KSCN.

<table>
<thead>
<tr>
<th>Element</th>
<th>Feed Conc.(M)</th>
<th>Adsorbed(^1) (mM/g)</th>
<th>Ratio in each fraction (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tc</td>
<td>0.0040</td>
<td>0.237</td>
<td>Effluent 1 &gt;98 &lt;0.5</td>
</tr>
<tr>
<td>Ru</td>
<td>0.0085</td>
<td>0.262</td>
<td>Eluate 51 41 8</td>
</tr>
<tr>
<td>Rh</td>
<td>0.0020</td>
<td>0.019</td>
<td>Adsorbent 84 2 14</td>
</tr>
<tr>
<td>Pd</td>
<td>0.0045</td>
<td>0.269</td>
<td>0 18 82</td>
</tr>
<tr>
<td>Others(^3)</td>
<td>0.53</td>
<td>0</td>
<td>100 0 0</td>
</tr>
</tbody>
</table>

1) Amount of the element adsorbed before elution.
2) Ratio of the element remaining in active carbon after elution.
3) Na, Cs, Rb, Sr, Ba, Cr, Fe and Ni.
Tc precipitation, however, depends on the concentrations of Pd and Rh. This method is not applicable for the Tc separation when the total concentration of Pd and Rh is lower than 0.005 M.

CONCLUSION

Through the fundamental studies mentioned above, the partitioning process is made up as shown in Fig. 8. As for the pretreatment of HLW for the DIDPA extraction, denitration is the most applicable method, but further study seems to be required particularly on the relationship to the first cycle raffinate of the Purex process. The extractant DIDPA showed a very good performance on the separation of TRU including Np from HLW. Experiments on continuous stripping of Am with DTPA leaving RE in the DIDPA solvent are now being carried out to examine the actually obtainable decontamination factor of RE and to find the optimum condition for this stripping.

Two methods were studied for the separation of Tc and PGM. Both methods of adsorption and precipitation have their advantages and disadvantages. Although the optimization is still necessary, the separation of these elements is possible.

The partitioning process will be tested with an actual HLW at the NUCEF (Nuclear Fuel Cycle Safety Engineering Research Facility), which is under construction in JAERI Tokai Establishment and will come into hot operation in 1995.

![Flow Scheme of the Partitioning Process in JAERI](image-url)
REFERENCES


MINOR ACTINIDE BURNER REACTOR AND INFLUENCE OF TRANSMUTATION ON FUEL CYCLE FACILITIES

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Japan Atomic Energy Research Institute,
Tokai, Ibaraki, Japan

Abstract

Two concepts of minor actinide burner reactors were previously proposed by the authors, one with metal fuel core and the other with nitride particle fuel core. These reactors have very hard neutron spectrum and very high neutron flux which are effective and efficient for fissioning of minor actinides. In these burner reactors, however, $\beta_{\text{eff}}$ was very small and the sodium void coefficient of a metal fuel reactor was positive and large. In order to improve the safety characteristics, the design of these burner reactors was modified.

As a part of transmutation system evaluation, minor actinide transmutation by actinide burner reactors is compared with that by power reactors from both the reactor physics and fuel cycle view points. The technical feasibility of these burner reactors heavily depends on the technical feasibility of fuel and the fuel cycle process. The study on manufacturing technology of TRU nitride fuel with TiN coating and the TRU alloy fuel studies are discussed.

1. Introduction

Various methods of minor actinide transmutation have been proposed in order to alleviate the burden of the high-level radioactive waste (HLW) depository problem. One of the practical methods is to recycle them in fast reactors since minor actinides such as $^{237}$Np, $^{241}$Am, $^{243}$Am (hereafter referred as MA) are fissionable with fission threshold in about 700 keV range and capture cross sections of these nuclides rapidly decrease with neutron energy higher than this threshold. A special reactor with a very hard neutron energy spectrum and high neutron flux would be very efficient and effective for MA transmutation if it is technically feasible. In this context we have been developing the concept of MA burner reactors (ABR; Actinide Burner Reactor). Once these ABRs will become available, the troublesome MA from the waste management viewpoint will be contained in a separate fuel cycle from the commercial one.

The concept of the double strata fuel cycle consisting of the commercial fuel cycle and the Partitioning-Transmutation (P-T) cycle is illustrated in Fig.1. The final HLW from this fuel cycle contains only fission products. The separate treatment of MA from the commercial cycle will be preferable for the conventional fuel cycle because MA is a strong neutron emitter and the MA recycling in the conventional fuel cycle will result in the requirement of the radiation shielding enforcement of the fuel cycle facilities.

In this report the conceptual design study of ABRs is discussed. Also, the efficiency of MA transmutation is compared between ABRs and power reactors (PWR and FBR). The effect of MA recycling in power reactors is discussed from the fuel cycle facility viewpoint.
Fig. 1   Flow of high-level radioactive waste per year through double strata fuel cycle combined with partioning and transmutation (MA burner reactor) cycle

2. Design Study of ABRs

The guidelines for designing an ABR are as follows;
- MA as the major fuel material,
- very hard core averaged neutron spectrum,
- very high neutron flux,
- pyrochemical reprocessing for compact fuel cycle facilities.

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Two types of ABR design were obtained, namely MA metal fuel with Na-cooling ABR and MA nitride particle fuel with He-cooling ABR. The details of these designing are described elsewhere.1,2,3)

In the original design of ABRs, the effective delayed neutron fraction $\beta_{\text{eff}}$ and Doppler reactivity coefficient are very small and the positive sodium void coefficient is large owing to the lack of U in a core and also to the very hard neutron spectrum.

3. Modification of ABR design

To improve the safety characteristics, the design of the original ABRs was modified, 1) to increase the value of $\beta_{\text{eff}}$, and 2) to avoid the large positive sodium void coefficient. Also, in the new design, MA alloy fuel is replaced with the MA nitride fuel because of relatively low melting point and low thermal conductivity of MA alloy fuel. The other advantage of nitride fuel is that nitride fuel can be processed with the pyrochemical reprocessing and hence, the fuel cycle facilities can be very compact.

To increase $\beta_{\text{eff}}$, uranium is added as a part of fuel material because of its large delayed neutron fraction. $^{238}\text{U}$ has the largest $\beta$ value but is not adequate as fuel material of ABR because the ABR is a system to burn MA and $^{238}\text{U}$ is source nuclide of MA. In the modified ABR, therefore, Pu is replaced with enriched uranium(EU, enrichment; 90%). $\beta_{\text{eff}}$ at the initial cycle of the modified ABR with fuel is 0.36% and at the equilibrium cycle it is 0.25%. These values are significantly larger than 0.16%~0.17% of the original ABRs. The neutron generation time is also increased from $7~11\times10^8$sec of the original ABRs to $1.3~1.5\times10^7$sec of new ABRs.

In ABR, the slight hardening of neutron spectrum introduces large positive reactivity effect because fission chain reaction is maintained by fast fission of MA. It is, therefore, difficult to reduce the large positive value of sodium void reactivity worth of ABR. In the new design, sodium is replaced with liquid lead as a coolant. The lead-cooled fast reactor concept is proposed by Adamov et al4) as an inherently safe fast reactor. In the lead-cooled ABR with MA nitride fuel(L-ABR), coolant density coefficient is always negative.

The fuel concepts of L-ABR and P-ABR are shown in Fig.2. The reactor design parameters of modified ABRs are summarized in Table 1. Comparison of core averaged neutron spectra is shown in Fig.3 In this figure, neutron spectrum of MOX-LMFBR is also shown for comparison. Significantly hard neutron spectra of ABRs are obvious.

As far as the Doppler effect is concerned, no improvement was attained because $^{238}\text{U}$ is not contained in the fuel and the neutron spectrum is very hard.

4. Comparison of MA transmutation in ABRs and in power reactors

In Table 2, the transmutation characteristics are compared between two types of ABRs together with thermal and fast reactors. For the MA transmutation in power reactors, the concentration of MA is assumed as 0.2% and 5% of heavy metal for U-PWR and fast reactors, respectively so that the addition of MA will not affect major reactor design parameters such as enrichment, coolant void coefficient etc.

In the table, the transmutation ratio is defined as the ratio of MA weight at the end of cycle to that of the beginning of cycle. In this definition, any nuclear reaction such as fission,
Table 1 Reactor design parameters of modified Actinide Burner Reactors

<table>
<thead>
<tr>
<th></th>
<th>L-ABR$^{(1)}$</th>
<th>P-ABR$^{(2)}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fuel concept</td>
<td>pin-bundle</td>
<td>coated particle</td>
</tr>
<tr>
<td>material</td>
<td>(64NpAmCm-36U)$^{3}$, $^{90}$N$^{0}$</td>
<td>(65NpAmCm-35U)$^{3}$, $^{90}$N$^{0}$</td>
</tr>
<tr>
<td>HM initial loading, kg</td>
<td>895</td>
<td>2870</td>
</tr>
<tr>
<td>MA/U</td>
<td>573/322</td>
<td>1865/1005</td>
</tr>
<tr>
<td>Reactor power, MWth</td>
<td>180</td>
<td>1200</td>
</tr>
<tr>
<td>Core height (cm)</td>
<td>47</td>
<td>124</td>
</tr>
<tr>
<td>Core diameter (cm)</td>
<td>98</td>
<td>124</td>
</tr>
<tr>
<td>Coolant material</td>
<td>Lead</td>
<td>Helium</td>
</tr>
<tr>
<td>Neutron flux, $10^{15}$/$\text{n/cm}^2$ sec</td>
<td>3.1</td>
<td>5.9</td>
</tr>
<tr>
<td>Core averaged mean neutron energy, keV</td>
<td>720</td>
<td>722</td>
</tr>
<tr>
<td>Reactivity (% $\Delta k/k$)</td>
<td>----------------</td>
<td>--------------------------------</td>
</tr>
<tr>
<td>Coolant-void reactivity/core</td>
<td>-1.3</td>
<td></td>
</tr>
<tr>
<td>Doppler reactivity/core ($\Delta T=300^\circ C$)</td>
<td>-0.01</td>
<td>-0.01</td>
</tr>
<tr>
<td>Kinetic parameters</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\beta_{\text{eff}}$</td>
<td>$2.6 \times 10^{-3}$</td>
<td>$2.6 \times 10^{-3}$</td>
</tr>
<tr>
<td>$L_{\beta}$, sec</td>
<td>$1.3 \times 10^{-7}$</td>
<td>$1.5 \times 10^{-7}$</td>
</tr>
<tr>
<td>Cycle length, full-power days</td>
<td>550</td>
<td>300</td>
</tr>
<tr>
<td>MA burnup, %/cycle</td>
<td>11</td>
<td>13</td>
</tr>
</tbody>
</table>

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

Fig. 3 Comparison of core averaged neutron spectra between minor actinide burner reactors (ABR) and MOX-FBR
Table 2  Comparison of MA transmutation in various reactors

<table>
<thead>
<tr>
<th></th>
<th>MA Burner Reactors</th>
<th>Power Reactors</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>L-ABR</td>
<td>P-ABR</td>
</tr>
<tr>
<td>Output (MWt)</td>
<td>180</td>
<td>1200</td>
</tr>
<tr>
<td>Cycle length (EFPD)</td>
<td>550</td>
<td>300</td>
</tr>
<tr>
<td>Core averaged</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fast neutron flux ( \times 10^{15} \text{n/cm}^2 \cdot \text{s} )</td>
<td>3.1</td>
<td>5.9</td>
</tr>
<tr>
<td>Mean neutron energy (keV)</td>
<td>720</td>
<td>720</td>
</tr>
<tr>
<td>MA loaded (kg)</td>
<td>573</td>
<td>1865</td>
</tr>
<tr>
<td>MA transmutation ratio (%/cycle)</td>
<td>16.0</td>
<td>18.8</td>
</tr>
<tr>
<td>MA burnup ratio (%/cycle)</td>
<td>11</td>
<td>13</td>
</tr>
<tr>
<td>MA transmutation (kg/1GW \cdot year)</td>
<td>278</td>
<td>292</td>
</tr>
<tr>
<td>MA burnup (kg/1GW \cdot year)</td>
<td>191</td>
<td>202</td>
</tr>
</tbody>
</table>

1) Metal fuel FBR
2) Effective fuel irradiation time
3) Concentration of MA in fuel; 0.2% for U-PWR, 5% for MOX-FBR and LMR
4) MA transmutation ratio = \( \frac{\text{MA}(\text{BOC}) - \text{MA}(\text{EOC}) - (\text{MA generated})^*}{\text{MA}(\text{BOC})} \)
5) MA burnup ratio = \( \frac{\text{MA fissioned} - \text{MA generated}}{\text{MA}(\text{BOC})} \)

Values of 4) and 5) are those for the equilibrium cycle in ABRs, and average values of 1 to 10th cycle for power reactors.

*) Values in the parentheses are for the outer core

**) MA generated in the fuel during irradiation

neutron capture, \((n,2n)\), etc. can be used for transmutation and the conversion of \(^{237}\text{Np}\) into \(^{238}\text{Pu}\) is a part of transmutation of Np. The MA burnup ratio is defined as the ratio of MA weight fissioned during the irradiation to that at the beginning of cycle. The latter is the real index of transmutation effectiveness and efficiency because only fission is a real transmutation reaction to solve the problem of long-lived MA.

The transmutation and the burnup ratios of power reactors in Table 2 are the net ones after the MA generation in fuel being deducted. The negative value of burnup ratio of U-PWR implies the more MA generation than MA burnup. The large discrepancy between transmutation ratio and burnup ratio in power reactors indicate the larger conversion of Np into Pu than in ABRs. The net MA burnup per 1GWt a year of ABRs is significantly larger than that of power reactors because in ABRs major fuel material is MA.

5. Impact of MA transmutation in power reactors on fuel cycle facilities

For the MA transmutation using power reactors, not only reactor performance and fuel manufacturing but also the influence of transmutation on the fuel cycle facilities should be taken into account. The large difference between the transmutation ratio and the burnup ratio of power reactors shown in Table 2 indicates the larger conversion of MA to heavier nuclides than fission of MA.
To evaluate the effect of MA addition to the fuel, the analysis was carried out to calculate the increase of decay heat, neutron emission, and \( \gamma \)-ray intensity. In Table 3, the effect of MA addition to the fuels of power reactors, namely, U-PWR, MOX-PWR (Pu usage in PWR) and FBR, is summarized. In this analysis, the effect of MA addition was calculated for both fresh and spent fuel. The amount of MA addition to the fuel is limited so as not to lead to significant change of major reactor parameters which are economically optimized.

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

\( a \): minor actinides (MA) fraction in fuel (HM weight %)  
\( b \): fuel enrichment  
\( c \): fuel burnup (cooling time : 10 years)

The fraction of MA in the fuels of U-PWR, and MOX-PWR are 0.2, 0.5 and 5 weight % of heavy metal, respectively, in this calculation. The increase of decay heat is caused by the generation of \( ^{244}\)Cm and the increase of neutron emission is caused by the generation of higher Cm isotopes and \(^{252}\)Cf.

As a result of increase of these Cm, Cf, for all of the MA transmutation scheme in power reactors shown in Table 3 the radiation shielding design change will be needed not only for the fresh fuel handling (manufacturing and transportation) but also for the spent fuel handling (transportation and reprocessing). This may cause the increase of cost of the of electricity generation. In the case of ABRs, the shielding and the decay heat removal are much severer problem than the MA transmutation in power reactors since the concentration of MA is very high in ABRs. The fuel cycle facilities for ABRs, however, are very compact and the required number of these facilities is small because nitride fuel can be reprocessed by pyrochemical process and the amount of material to be handled is small compared with those of conventional fuel cycle. Therefore, the economy of MA transmutation may be favorable for the ABRs even if the resources required to develop ABRs is larger than that for MA transmutation in power reactors.\(^3\)
Fig. 4 shows the proposed fuel cycle scheme of the TRU nitride. The partitioned MA salts are used in the sol-gel processing of the microspheres of the oxide-carbon mixtures. The microspheres are converted to the nitrides by the carbothermic reduction in the flowing $\text{N}_2+\text{H}_2$ gas. The TiN coatings are applied in a spouted bed. The irradiated spheres are crushed into fragments and charged into the anode basket of a fused salt electrorefiner. The actinide nitrides are anodically dissolved, while the TiN coating remains virtually unattacked. The actinide metals are recovered on the cathode. Liquid Cd cathode may be used for the recovery of heavier actinides. The recovered metals are dissolved into either Cd or Sn, then converted to the nitrides by the reaction with nitrogen. As a liquid medium, Cd is favored for its ease of removal by volatilization, but it has a drawback of lower actinides solubility. Solubility of the actinides in liquid Sn is large, but the removal of Sn is rather difficult. The nitrides are then granulated to microspheres.

Technology base of the chemical vapor deposition (CVD) of high-density TiN coating exists: The CVD TiN is widely used as the coating material on the cutting tools. Besides, spouted-bed coaters for the HTGR particle fuels can be applied with little modification to the TiN coating on the microspheres. However, no serious attempts do not seem to have been made for the CVD of low-density TiN (buffer) coating. The study of the low-density TiN coating fabrication is underway; initial attempts to deposit the low-density TiN on stationary substrates have been successfully made. The temperature during deposition could be kept well below 1200K.
The study on the sol-gel processing of the nitride fuel is also being made. An innovative internal gelation technique where the falling droplets of the nitrate solution containing an ammonia donor are directly heated with microwave, which has been developed in the Paul Sherrrer Institute, is being tried. Emphasis is on the use of a commercially available microwave generator.

Laboratory runs of the fused salt electrolysis of UN have been made. The recovery of uranium metal has been demonstrated. The salt medium was the LiCl-KCl eutectic melt. The anode basket was made of graphite. During the electrolysis, the system was purged with purified helium. Conversion of the metal to the nitride has been readily made in liquid Cd with nitrogen cover gas.

In designing the processes one probably has to keep the temperature as low as possible at all stages, since Am among MA has very high volatility. One of the key stages would be the sintering of the nitride microspheres, where normally a high temperature of 1873-1973K is required.

The knowledge on the actinide alloys is necessary not only in understanding the behavior of nitride fuels but also in designing the TRU alloy fuels which are alternative fuels to the nitride. The alloying behavior of the TRU elements is being systematically studied by Gibson et al. Studies to date concentrated on the Np alloys such as Np-Am, Np-Zr and Np-Fe. Based on those experimental findings, the computer analysis and prediction of the ternary systems, namely, Np-Am-Pu, U-Am-Pu etc., have been made.

7. Summary

The original ABR design was modified to improve safety characteristics. Nitride fuel of MA is used instead of alloy fuel because of relatively low melting point and thermal conductivity of MA alloy. Mixture of enriched uranium in MA fuel increases effective delayed neutron fraction and neutron generation time significantly. Sodium was replaced with liquid lead as coolant to avoid the large positive sodium void effect.

The MA burnup per 1GW per year is 190 to 200kg which is smaller than those of the original ABRs because the contribution of $^{235}$U fission is 30 to 40% of the total fission in the modified ABRs.

From the view points of fuel cycle facilities, MA transmutation in power reactors (LWRs and FBRs) will require the design change of the radiation shielding in the whole fuel cycle facilities because of the increase of strong neutron emitting nuclides as a result of neutron capture of MA even if MA content in fuel is marginally small from reactor physics view points. Cost evaluation is needed to select a cost effective transmutation system based on the reliable data base which is presently not available.

The technical feasibility of these burner reactors heavily depends on the technical feasibility of fuel and the fuel cycle process. The study on manufacturing technology of TRU nitride fuel with TiN coating and the TRU alloy fuel studies are in progress.

The ABR concept will enable the containment of troublesome MA in one closed site. From the economics and safety view point, the containment of MA in one site where the fuel management will be easier may be more desirable than the nationwide spread of MA in power reactors.
REFERENCES


The management of high level radioactive waste (HLW) generated from the reprocessing of spent fuel is essential for the completion of nuclear fuel cycle. Current national policy in Japan is to solidify the HLW into a stable form and to dispose it in a deep geological repository after 30 to 50 years of storage for decay heat cooling. In PNC, many R&D efforts have been devoted to establish technologies for its safety disposal and methodologies for its safety assessment. In parallel with the national project mentioned above, the Japan's Atomic Energy Commission approved the "Long-Term Program for Research and Development on Nuclide Partitioning and Transmutation" in October 1988. The objective of the program is to explore a possibility to utilize HLW as useful resources and to make the geological disposal more efficient. This program is called "OMEGA". The program is composed of two major R&D areas: the nuclide partitioning from HLW based on its half life and its potential value for utilization and the transmutation of minor actinides (MA) and long-lived fission products (FP) to short-lived or stable nuclides. It is important to note that this program has been conducted as a long-term basic research, not as a national project. Based on the "OMEGA" program, partitioning and transmutation (P-T) studies have been conducted in PNC as a part of frontier research. This paper describes PNC's approach, present status and future plans for P-T.

1. Introduction

The management of high level radioactive waste (HLW) generated from the reprocessing of spent fuel is essential for the completion of nuclear fuel cycle. Current national policy in Japan is to solidify the HLW into a stable form and to dispose it in a deep geological repository after 30 to 50 years of storage for decay heat cooling. In PNC, many R&D efforts have been devoted to establish technologies for its safety disposal and methodologies for its safety assessment.

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The outline of the program is shown in Fig.1. The program is composed of two major R&D areas; the nuclide partitioning from HLW based on its half life and its potential value for utilization
and the transmutation of minor actinides (MA) and long-lived fission products (FP) to short-lived or stable nuclides. It is important to note that this program has been conducted as a long-term basic research, not as a national project.

Based on the "OMEGA" program, partitioning and transmutation (P-T) studies have been conducted in PNC as a part of frontier research. This paper describes PNC's approach, present status and future plans for P-T.

2. PNC's Approach to Partitioning and Transmutation

Based on the "OMEGA" program, PNC has been conducting P-T studies as a long-term basic research. In the partitioning study, MA, long-lived FP and noble metals are considered as the candidate nuclides to be separated from the HLW.

PNC's basic approach is to develop partitioning process as a part of advanced reprocessing system, where the improved PUREX process will be combined with MA separation process, as shown in Fig.2. Thus, main efforts have been devoted to separate MA by wet process, such as TRUEX process. In addition, basic research has been conducted to recover noble metals from the HLW or the insoluble residue by wet or dry process.
Table 1 is a list of nuclides, which can be the objects for transmutation study, because of their larger production in LWR and longer half-lives. PNC considers that it would be advantageous to transmute MA in LMFBR, because fission cross sections of most of MA are large enough and also R&D can be minimized by applying existing technologies for LMFBR. Thus, R&D efforts have been concentrated to pursue a possibility to transmute MA in LMFBR. In addition, a concept of Super Long Life Core (SLLC) has been proposed, using MA fuel.

On the other hand, it is usually difficult to transmute most of long-lived FP in fission reactors, because of their smaller neutron cross sections, as shown in Table 1. So, a possibility to transmute long-lived FP by accelerators is also investigated.

P-T studies in PNC have already shown considerable progress as will be shown in the following. However, to establish a concept of P-T system which covers the entire nuclear fuel cycle, long-term R&D efforts will be required to obtain additional data, which are essential to make decision. Thus, PNC will continue R&D efforts in P-T studies and also preliminary system studies to establish a future concept of P-T system.
### Table 1: Main Objective Nuclides for Transmutation Study

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Half Life (year)</th>
<th>Neutron Cross Section*(b)</th>
<th>Production** (Ci/year) (kg/year)</th>
</tr>
</thead>
<tbody>
<tr>
<td>MA</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{237}$Np</td>
<td>$2.1\times 10^6$</td>
<td>181 (1.6)</td>
<td>11</td>
</tr>
<tr>
<td>$^{241}$Am</td>
<td>432</td>
<td>603 (1.3)</td>
<td>$5.0\times 10^3$</td>
</tr>
<tr>
<td>$^{243}$Am</td>
<td>7380</td>
<td>79 (1.1)</td>
<td>601</td>
</tr>
<tr>
<td>$^{243}$Cm</td>
<td>28.5</td>
<td>720 (2.0)</td>
<td>55</td>
</tr>
<tr>
<td>$^{244}$Cm</td>
<td>18</td>
<td>15 (2.0)</td>
<td>$5.8\times 10^4$</td>
</tr>
<tr>
<td>$^{245}$Cm</td>
<td>8500</td>
<td>2347 (1.6)</td>
<td>$4.1\times 10^3$</td>
</tr>
<tr>
<td>FP</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{90}$Sr</td>
<td>29</td>
<td>0.014</td>
<td>$25\times 10^6$</td>
</tr>
<tr>
<td>$^{93}$Zr</td>
<td>$1.5\times 10^6$</td>
<td>2.6</td>
<td>61</td>
</tr>
<tr>
<td>$^{99}$Tc</td>
<td>$2.1\times 10^5$</td>
<td>20</td>
<td>433</td>
</tr>
<tr>
<td>$^{107}$Pd</td>
<td>$6.5\times 10^6$</td>
<td>1.8</td>
<td>3.6</td>
</tr>
<tr>
<td>$^{129}$I</td>
<td>$1.6\times 10^7$</td>
<td>27</td>
<td>1.0</td>
</tr>
<tr>
<td>$^{135}$Cs</td>
<td>$2.3\times 10^6$</td>
<td>8.7</td>
<td>13.5</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>30</td>
<td>0.25</td>
<td>$3.5\times 10^6$</td>
</tr>
<tr>
<td>$^{151}$Sm</td>
<td>90</td>
<td>15000</td>
<td>$1.1\times 10^4$</td>
</tr>
</tbody>
</table>

* For MA, total of $(n,\gamma)$ and $(n, f)$ cross section for thermal neutron, value in parenthesis shows $(n,f)$ cross section for fast neutron (1MeV)
* For FP, $(n,\gamma)$ cross section for thermal neutron
** Annual production in a 1,000MWe LWR

3. Partitioning of Long-Lived Nuclides

PNC has been carrying out basic experiments for the purpose of separating actinides from High Level Liquid Waste (HLLW) since 1990, as a part of an effort to increase the fuel utilization efficiency and to widen the options for waste management in the future. Due to the high compatibility with the present PUREX process, application of the general TRUEX process as a partitioning tool is current PNC's strategy.

3.1 Improvement of PUREX Process

As a concept for advanced reprocessing system illustrated in Fig.2, several improvements are still required on the present PUREX process along with development of the TRUEX process. Namely, for lightening the load on the succeeding TRUEX process, the system which generates smaller amount of HLLW with
simpler compositions preferably should be incorporated into the upper PUREX process. For this purpose, sodium-contained compound should be replaced by another type reagents, and MA, at least Np should be retained as well as U and Pu in the PUREX process.

The significant portion of Np in the dissolver solution was already observed to be extracted in the present PUREX-first cycle without valency adjustment\(^3\). Therefore, quantitative Np extraction can be realized if the treatment that will shift its valency to the higher state in the dissolver solution is added. One idea is based on the fact that high acidification of the feed dissolver solution decreased the less-extractable portion of Np. This method will not produce any secondary waste except for nitric acid. Based on this, a few PUREX flowsheet demonstrations have been carried out in showing the expected Np behavior.

For improving HLLW as well as MLLW quality to reduce sodium contamination, some nitrogen-based salt-free reagents i.e., HAN, hydrazine oxalate and hydrazine carbonate were respectively applied for reductive partitioning and for solvent regeneration, in combination with in situ electrooxidation which acted as an oxidizer for Pu(III) and Np(V) and a decomposer for the waste reagents. Such a process named "Salt-Free PUREX" was successfully demonstrated with various PUREX solutions\(^4\).

Being troublesome characteristics of ionic state of noble metals for the PUREX and TBP-based TRUEX process, these are preferably removed prior to both extraction cycles. This seems to contribute to the simplification of the extraction system. A basic potentiostatic test with diaphragmless electrolysis suggested that Pd, Rh and Ag ions tend to deposit selectively on Ta or Ti electrode surfaces by polarizing the electrode potential to be negative. A very clear potential dependency was observed on Pd deposition rate on Ta electrode in 3M nitric acid\(^5\). A maximum deposition rate was obtained at a little more positive potential than the hydrogen evolution potential. Although an effect of acidity on Pd deposition rate was not so significant, a presence of U(VI) suppressed it greatly. These, however, can be counterbalanced by the long-term electrolysis, because deposition occurred irreversibly. Against Pd, high electrical energy was required to obtain an appropriate deposition for Ru and Rh. Then another interest is the possibility to translate extractable species of Ru or Tc to a less-extractable one by an electrolytic method. PUREX experiments integrated with the electrochemical partitioning using real dissolver solution are being planned in the PNC's hot experiment facility, CPF.
3.2 Application of TRUEX Process for Actinides Partitioning

The TRUEX process is characterized by the use of the bifunctional extractant CMPO, octyl(phenyl)-N,N-diisobutylcalbamoylmethylphosphine oxide, mixed with a PUREX solvent. The mixed solvent was reported to extract well tri-, tetra- and hexavalent actinides from various kinds of solution. The main purpose of our study was to evaluate the applicability of TRUEX process on HLLW-partitioning, then to modify it if necessary. Thus the counter-current tests using real High Active Raffinate (HAR) derived from PUREX-first cycle was conducted in parallel with the basic distribution study.

As a solvent composition, CMPO concentration was fixed to 0.2M, whereas TBP concentration in n-dodecane was changed, from 1.0M to 1.4 M to meet the target waste solutions in the counter-current tests. The HAR was derived from PUREX reprocessing experiments of fast reactor spent fuel burned up to ca 54000 MWD/T and cooled for ca.2 ~ 4 years, and HLLW from LWR reprocessing.

The summary of distribution ratio (D) for major nuclides in the HLLW as a function of nitric acid concentration is shown in Fig.3. It can be seen that D for tri-valent MA and rare earth (RE) agreed well with the past data, but new information was also obtained; namely, Pu(III) obtained by reaction with an excess of HAN shows considerably higher D than expected. This indicates that unlike in the PUREX process, reductive stripping of Pu may not be effective. Np(mix) as prepared by equilibrating Np in nitric acid solution about 24 hours showed a similar distribution tendency with tri-valent MA. A higher acidity will give a higher extractability on Np(mix), then acidification of dissolver solution will be an effective method to cause quantitative extraction. As for Ru, either very high or low nitric acid concentration seems to be preferable to lower its extractability, although not clearly shown herein. The most noticeable behavior of Ru was characterized by the fact that its fractionation behavior in the continuous counter-current tests could not be fully explained by the batchwise distribution data.

In the counter-current tests, the number of mixer-settlers was 19 stages for extraction-scrubbing and 16 to 19 stages for stripping including solvent regeneration. The volume of one stage of the mixer-settler was 23 mL. The HAR was used by not lowering but increasing acidity(ca.7M). Fig. 4 shows the flowsheet modifications for the past tests. To improve the fractionation, management of nitric acid entrainment to the partitioning banks was important, thus dual scrubbing using two different concentrations of nitric acid was adopted. As indicated in the third run, HAN, oxalic acid and sodium carbonate scrubbing stages were prepared in series following to dilute nitric acid stripping, as
a reference to improve appropriate fractionation method of all of actinides in the HLLW. Four runs were carried out up to now.

The counter-current test gave the results that the extraction of all of actinides and tri-valent RE was completed in provided number of stages, and showed a decontamination factor (DF) larger than $5 \times 10^3$ for removal of $^{241}$Am and total-$\alpha$ from the HAR. Although $^{137}$Cs and $^{125}$Sb always showed simple decontamination behaviors, that of $^{106}$Ru was very complicated. Owing to above mentioned improvement, decontaminated portions of $^{106}$Ru became higher from less than 90% to 94% ~ 98%.

The typical partitioning profiles of actinides and FPs are shown in Fig.5. The tri-valent MA, $^{241}$Am, $^{244}$Cm, and RE, $^{144}$Ce, seemed to be stripped well with dilute nitric acid as expected, but recovery rates of these nuclides to aqueous product were still around 50 to
80%. That would be attributed to the small accumulation and/or invisible precipitation in the scrubbing banks. The organic Pu and Ru concentration were almost constant even though contacting with HAN and oxalic acid afterwards dilute nitric acid treatment. However in the sodium carbonate scrubbing banks, their organic concentration became significantly decreased: i.e., a combination of these reagents gave a more than 80% of Pu recovering to the aqueous stream, instead of less than 5% by a simple dilute nitric acid stripping. Np also stripped well by these treatments.

The stagewise distribution ratio of $^{106}$Ru indicated that at least two kinds of complexes were existed in the HAR and a very extractable species having high D value (e.g., $\geq 20$ at 0.01M nitric acid ) was remained in the organic phase. However, considerable effect obtained by a sodium carbonate scrubbing for such retained Ru, might attribute to the same high pH effect observed in the PUREX system. Therefore, the similar scrubbing can be expected for hydrazine carbonate as an alternative.

Through all of counter-current tests for the HAR, the third phase splitting was never found. To avoid third phase for highly concentrated HAR or HLLW, systematic tests on the formation mechanisms were carried out. For this purpose, various involved factors including the branching effect of C-12 diluents were
Fig. 5  Typical Partitioning Profiles of Actinides and FPs in the 3rd Counter-Current Test.
investigated. As a tentative indication, dilution of real HLLW at least 5 times was necessary when the mixed solvent composed of 0.2MCMPO-1.0MTBP in n-dodecane is employing under the room temperature.

The DF values can be improved by sophisticating the flowsheet parameters. The separation factors, however, for RE removal from $^{241}$Am products can not be improved unless another separation techniques are incorporated to the TRUEX process.

3.3 Research for New Extractant

Along with the research on the PUREX and TRUEX process, other extractants were studied. New-type crown ether derivatives having allosteric metal binding site were proposed by Kobuke, et al. That unique concept describes that a host with two metal binding sites, in which pre-binding of M1 to the first site influences the second site electronically and conformationally to convert it from a weak binding site into a potent cooperative binding site for M2. This kind of crown ether seems to be more promising in ability and synthesis than the existing dibenzo-18-crown-6 ether. The PNC/University collaboration work suggested that copper (as M1) - assisted coronands, e.g.,[1,2-Benzene bis(1,4dioxanonyl-6,8-dionato) Copper(II) afforded the most appropriate and selective binding site to alkali and alkaline earth metal ions (as M2). Tentatively, an extraction yield of ca.80% for Sr$^{2+}$ from a LiOH solution with presence of small amount of picric acid was obtained. Crown ethers and calixarenes derivatives seem to have excellent selectivity for the long-lived nuclides in the HLLW.

4. Minor Actinide Transmutation in LMFBR

Some of MA nuclides contained in HLW from reprocessing have extremely long-term radiotoxicity. There are some means of reducing radiotoxicity of the MA nuclides under investigation throughout the world. The MA nuclides produce useful energy when converted into short-lived fission products by neutron bombardment. From this standpoint, a nuclear reactor provides an extremely rational means for transmutation of MA nuclides. Among the various nuclear reactors, the sodium-cooled fast breeder reactor (LMFBR), now under development, can be used for transmutation of MA nuclides, because of the possible nuclear fission generated by high-energy neutrons.

The following studies are implemented to establish MA transmutation technology by LMFBRs in PNC-Japan:

- Feasibility studies of MA transmutation by LMFBR and evaluation of MA material balance,
• Nuclear data evaluation of MA nuclides in sample irradiation experiments.
• Measurement and evaluation of physical and chemical properties of MA compounds,
• Development of fabrication technology of MA fuel,
• Evaluation of fuel behavior by MA pin irradiation experiments.

We introduce the results of the following two feasibility studies of MA transmutation in LMFBR:
(a) Study on a MA burning FBR
   In this study, systematic parameter survey calculations have been performed for a conventional 1000MWe-class LMFBR core. The purpose is to develop LMFBR core concepts loaded with MA fuel which has no serious penalties to core performance in consideration of fuel cycle technology.
(b) Study on an innovative core concepts
   The purpose of this study is to develop an innovative core concept by utilizing the properties of MA. Taking into account the advantage of the MA-loaded core, a Super-Long-Life-Core (SLLC) with no need of fuel exchange during plant life is proposed as an LMFBR core for MA transmutation. The SLLC is an attractive option transmuting MA nuclides since it can transmute them with confining them into the reactor during plant life. A feasibility of the SLLC was studied from the neutronic view point.

4.1 Study on a MA burning FBR

The main results of the study are summarized as follows.
(1) Evaluation of properties of MA containing fuel
   Based on the experimental data on fuel properties of MA fuel published up to now, it was found that MA loading significantly reduces the linear power limit almost proportionally to the MA loading ratio because of degradation of the thermal conductivity and the melting point as shown in Fig.6. For example, the linear power limit for the fuel pin with 50% MA loading reduces about 30% compared with that of no MA-loaded fuel pin. It should be noted that the result includes considerable uncertainty because of insufficient experimental data.
(2) Optimization of loading method of MA
   The MA transmutation in the typical large LMFBR with MOX fuel has no serious penalties from the view point of core performances, provided that the loading method can be employed with small ratio of MA fuel. (~5%MA for homogeneous loading method.)
   The heterogeneous MA-loading method is feasible by optimizing fuel design, loading pattern and coolant flow of MA-loaded fuel subassemblies. The reduction of the fuel pin
diameter and the Pu enrichment is essential to reduce the power of MA-loaded fuel in the heterogeneous MA-loading.

The MA loading in the blanket region has no problems from the viewpoint of core performances. MAs are transmuted at a rate of 6% per cycle in the axial and radial blanket region.

(3) Effect of rare earth (RE) in MA on core characteristics (Table 2)

The homogeneous loading of MA and RE has no serious penalties to the reactor core performance, provided that the amount of MA and RE in the fuel is less than 5 and 10w%, respectively. In case of adding MA and RE in the axial and radial blanket region, it is possible to insert ~5w% of MA and ~20w% of RE in the axial and radial blanket assemblies from the viewpoint of core performance.

(4) Influence of uncertainties of MA nuclear data

The influences of the uncertainties upon nuclear characteristics were evaluated for a large LMFBR core loaded with MA of 5%. Sensitivity analysis on cross sections was carried out and uncertainties of nuclear characteristics were roughly evaluated. Uncertainties of nuclear characteristics are rather large compared with those of conventional cores. Some cross sections of MAs (237Np, 241Am, 238Pu, 243Am and 244Cm) need to be improved.

(5) Influence of MA containing fuel on reactor plant and fuel cycle

Both the decay heat and neutron emission rate of the
Table 2: Core Performance of the MOX Fuel Core with MA and RE

<table>
<thead>
<tr>
<th>Item</th>
<th>Case-1</th>
<th>Case-2</th>
<th>Case-3</th>
</tr>
</thead>
<tbody>
<tr>
<td>RE (wt%)</td>
<td>0</td>
<td>10</td>
<td>30</td>
</tr>
<tr>
<td>MA (wt%)</td>
<td>5</td>
<td>5</td>
<td>5</td>
</tr>
<tr>
<td>Pu Enrichment (wt%)</td>
<td>16.6(Inner)</td>
<td>20.0(Inner)</td>
<td>29.2(Inner)</td>
</tr>
<tr>
<td></td>
<td>20.1(Outer)</td>
<td>24.2(Outer)</td>
<td>35.4(Outer)</td>
</tr>
<tr>
<td>Burnup Reactivity (%Δk/Δk')</td>
<td>2.12</td>
<td>3.71</td>
<td>6.40</td>
</tr>
<tr>
<td>Maximum Linear Heat Rate (W/cm)</td>
<td>407</td>
<td>413</td>
<td>433</td>
</tr>
<tr>
<td>Breeding Ratio</td>
<td>1.07</td>
<td>0.95</td>
<td>0.74</td>
</tr>
<tr>
<td>MA Transmutation Rate (%)</td>
<td>10.9</td>
<td>10.3</td>
<td>9.7</td>
</tr>
</tbody>
</table>

MA(Np, Am and Cm)-loaded fuel are very large in comparison with MOX fuel without MA. The dominant element of these fuel properties is Cm. If it is possible to remove Cm from the MA-loaded fuel, the decay heat value will decrease by one order, and the neutron emission rate by three orders. Since the dominant isotope, $^{244}$Cm, has a relatively short half-life of 18 years, there might be another possibility of the fuel cycle, that is, partitioning of Cm and Am from MA in the reprocess and storing of Cm and Am for a period. Some study will be needed to estimate the trade-off between the plant modification and the reprocessing.

4.2 Study on innovative core concepts

The following results are summarized from the study on the SLLC. 
(1) Study on a 1000 MWe-class SLLC

An LMFBR core loaded with MA has a large potentiality of extending operation cycle length. Making use of the potentiality, a super long life FBR core loaded with MA was studied aiming at continuous operation without refueling during plant life and efficient reduction of MA nuclides. The 1000 MWe SLLC without fuel exchange for 30 years is feasible from neutronic and
thermal hydraulic points of view, because reactivity change and power swing during burnup can be remarkably reduced by MA loading. The MA fuels can be confined and transmuted in the reactor during plant life. The 1000 MWe SLLC can transmute MA of 10 ton which comes from 13 LWRs (1000 MWe).

(2) Study on a 300 MWe-class SLLC

The feasibility study of a 300 MWe SLLC was carried out. As a result, the burnup reactivity change of the optimized core for 34 years is very small as shown in Fig. 7. The power swing is also small, which resulted in satisfaction of the thermal design criteria. The amount of MA transmuted during lifetime is about 5.3 ton, which is equal to that 6 LWRs of 1000 MWe produce during their lifetime. The Doppler coefficient (absolute value) is rather small because of MA loading. Further study is needed on core kinetics from the viewpoint of core safety and control.

![Fig. 7 Reactivity Change due to Burnup (300 MWe SLLC)](image)

4.3 R&D issues for MA transmutation in LMFBR

Shown in Table 3 is the R&D program in PNC for the establishment of MA transmutation technology in LMFBR.

Through design study of LMFBR transmuting MA and evaluation of MA mass balance, it is planned to establish
appropriate core design concepts for a large-size LMFBR and strategy of their introduction. Basic researches including nuclear data evaluation, and fuel property measurement and evaluation are also programmed.

The experimental fast reactor JOYO is planned to be used both for initial transmutation experiments irradiating fuel samples, and for MA-loaded oxide fuel irradiation tests. In addition, basic tests are planned on fuel preparation of MA obtained from the partitioning of nuclides, the results of which will be reflected in fuel fabrication technology.

5. Transmutation by Accelerator

It is usually difficult to transmute long-lived FPs in fission reactors, because neutron cross sections of most of these elements are not sufficiently large. Thus, various methods have been proposed to transmute long-lived FPs using accelerators, such as an electron accelerator, a proton accelerator, an accelerator-driven fission reactor (hybrid system) and so on.

Among these, transmutation by an electron accelerator applies photonuclear reaction, using high energy gamma rays produced by the accelerator. The advantages of this method are smaller production of secondary radioactive wastes and broader base of accelerator technology, compared to that of a proton accelerator.
5.1 Design of high power electron linac

A high power electron accelerator will be required in a future transmutation system. For example, to transmute $^{137}$Cs which is produced in a 1000MWe LWR, a beam current of about 1A will be needed, even if the transmutation efficiency is supposed to be 100%. On the other hand, the energy required will be around 100MeV, because the maximum cross section of $(\gamma, n)$ reaction usually exists between 10 to 20 MeV which is efficiently obtained by the electron beam of 100MeV.

Figure8 shows typical electron linacs in the world. The main effort has been directed to increase the beam energy, to try to find a new phenomenon mainly in the field of elementary particles. Contrary to this, for a future transmutation system, a large current of the order of 1A will be required.

It is technically and also financially quite difficult to achieve the energy of 100MeV and the current of 1A at a step. Thus, the energy of 10MeV and the maximum / average current of 100mA / 20mA were selected as the objective for the first step. The main technical subjects for the high current linac are the suppression of the BBU (Beam Break Up) and the removal of the heat generated in the linac.

The main specification and the basic structure of the linac are shown in Table 4 and Fig. 9, respectively. The beam line consists of an electron gun, a chopper, a pre-buncher, a buncher, accelerator guides and a beam dump. The special feature of the linac is the application of the TWRR (Traveling Wave Resonant Ring), instead of the usual traveling wave or standing wave accelerator guide. The advantage of the TWRR is the better efficiency, because of the reuse of microwave. To prevent the BBU, shorter accelerator guides are used and Q magnets are located between guides to correct the beam. The accelerator guide is designed to remove large amount of heat and to compensate the frequency change due to the temperature rise.

5.2 High power test

High power tests were conducted as a collaboration work between PNC and KEK (National Laboratory for High Energy Physics). The test system is shown in Photo.1. Maximum power of 300kW and 780kW were achieved in CW (continuous wave) mode and in pulse mode, respectively, using trially fabricated klystron. To improve endurance of the klystron window, three types of window were examined; berylia standard type, berylia long type and alumina long type. The temperature increase of the window is shown in Fig.10 as a function of the injected power. Among these, the berylia long type window showed best endurance and it was possible to inject 1.7MW and 4.5MW in CW
Fig.8  Electron Linear Accelerators in the World
Table 4 Main Specifications of PNC Electron Linac

<table>
<thead>
<tr>
<th>Specification</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Max. Beam Energy</td>
<td>10 MeV</td>
</tr>
<tr>
<td>Max. / Ave. Current</td>
<td>100 mA / 20 mA</td>
</tr>
<tr>
<td>Pulse Length</td>
<td>4 ms</td>
</tr>
<tr>
<td>Beam Repitition</td>
<td>50 Hz</td>
</tr>
<tr>
<td>Duty</td>
<td>20%</td>
</tr>
<tr>
<td>Average Beam Power</td>
<td>200 kW</td>
</tr>
<tr>
<td>RF Frequency</td>
<td>1.249135 GHz</td>
</tr>
<tr>
<td>Length of Microwave</td>
<td>24 cm</td>
</tr>
<tr>
<td>Mode of Acceleration</td>
<td>2π / 3</td>
</tr>
<tr>
<td>Klystron Power</td>
<td>1.2 MW</td>
</tr>
<tr>
<td>Total Length of Linac</td>
<td>16 m</td>
</tr>
</tbody>
</table>

Fig. 9 Scheme of CW Electron LINAC in PNC
Fig. 10 Surface Temperature of the Windows

Photo 1 High Power Test System

1 Wave Guide
2 RF Window
3 Directional Coupler
4 Microwave Detector
5 Stub Tuner
6 Coupler
7 Accelerator Guide
8 Coupler
9 Phase Shifter
10 RF Window
11 Microwave Director
12 Dummy Load
mode and in pulse mode, respectively. These exceeded the design values, thus it was shown that the high power klystron would be realized for the planning linac. It was also shown that the RF characteristics could be well estimated by calculations, even though the power was extremely high. It was demonstrated that the trially fabricated accelerator guide could endure the power of 800kW, without any electric discharge or thermal deformation. Based on the design study and the high power test results, the fabrication of the high power electron linac will be started this year and all the components will be installed by 1996.

6. Conclusions

Various R&D activities related to the P-T are in progress in PNC based on the OMEGA program in Japan. Substantial results have been already obtained, as shown in this paper, however, these are not sufficient enough to establish a future P-T system. Additional efforts will be continued in PNC to accumulate basic data and also to examine a future nuclear fuel cycle including the P-T. It should be reminded however that the OMEGA program is not intended to seek short-term alternatives for present back-end policies, but to pursue benefits for future generations through long-term basic R&Ds.

REFERENCES

PYROMETALLURGICAL PARTITIONING OF TRANSURANIUM ELEMENTS AND THE TRANSMUTATION IN METALLIC FUEL FBR

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Abstract

High level radioactive waste (HLW) from reprocessing of LWR spent fuels contains a small amount of long-lived transuranium elements (TRU). A pyrometallurgical process for partitioning of TRU from HLW is being developed for their transmutation in a metallic fuel FBR. This process consists of (1) microwave heating denitration, (2) chlorination, (3) reductive extraction and (4) electrorefining. The wastes salts containing mainly alkali and alkaline metals are electrolyzed to obtain the stable form oxides for the disposal. The TRU recovered are mixed with the metallic fuel of U-Pu-Zr for fissioning in FBR. The characteristics were evaluated for alloys containing minor actinides (MA: Np, Am, Cm). The alloy fuel with MA is scheduled to irradiate in the Phenix Fast Reactor.

1. INTRODUCTION

The partitioning and transmutation of long lived nuclides is one of the innovative technologies for future nuclear fuel cycle. The Central Research Institute of Electric Power Industry (CRIEPI) is now focusing on the research of separation of long lived nuclides, especially transuranium elements (TRU), by pyrometallurgical process from high level liquid waste, scrubbing solution of solvent and undissolved residue coming from Purex type of reprocessing, and on the research of recycling of minor actinides (MA; Np, Am, Cm) as well as uranium and plutonium in a metallic fuel FBR cycle by mixing them in U-Pu-Zr alloys [1-3].

The pyrometallurgical partitioning process is expected to produce a lower amount of secondary radioactive wastes and to favor compact facilities as compared with conventional aqueous processes using solvent extraction, ion exchange, etc., although products with low purity are generally recovered by pyrometallurgical processes [4]. On the other hand, better matching of transmutation of MA separated as metal form through the partitioning process can be attained by use of a metallic fuel FBR. In addition, transmutation of MA in a metallic fuel FBR under development for commercial use has some advantage that can be applied conventional technology with less effort for development compared to other concepts such as transmutation by actinide burning reactor and spallation by high energy proton accelerator. The pyrometallurgical process is also applied for the reprocessing of metallic fuel [5,6]. The other aspect of this concept is the high resistance for nuclear proliferation because of all the actinides including some of rare earths as impurity are recycled instead U and Pu with high purity.

2. DEVELOPMENT OF PYROMETALLURGICAL PARTITIONING

2.1. Pyrometallurgical process for partitioning

Figure 1 shows the flow diagram of the pyrometallurgical process for partitioning of TRU from HLW proposed by CRIEPI [1]. This process consists of (1) microwave heating denitration to make oxides, (2) chlorination to convert oxides to chlorides, (3) reductive extraction to separate TRU from molten chlorides using liquid Cd-Li, (4) electrorefining to purify the TRU recovered and (5) waste salt treatment by electrolysis to convert to the stable form. Both the denitration and chlorination steps are pre-processing for partitioning. In the process, cadmium can be recycled after separating TRU, and chlorine gas and lithium will be also recycled through salt waste treatment in order to minimize the process waste to be disposed.
In the study, we assumed that HLW contains not only 0.5% of U and Pu and all of MA existed in spent fuels but also some contribution of wastes from solvent scrubbing in the reprocessing. This assumption indicates that the level of rare earth elements, which are chemically similar to TRU, is about 10 times as large as the amount of TRU. The objective of the research effort is that more than 99% of TRU in HLW is recovered by pyrometallurgical partitioning, accompanied with the amount of rare earth elements equivalent to the level of MA in the product.
2.2. Denitration

Microwave heating was selected as the preferred method for denitration of HLW. Sample solutions simulating the composition of HLW were heated under suitable conditions of microwave power (frequency : 2,450 MHz). Figure 2 indicates the relationship between denitration rate and heating time. It was shown that denitration time becomes shorter with increasing microwave power. The surface temperature of sample required for almost complete denitration is over 700°C, as suggested from the pyrolysis temperature measurement for major nitrates in HLW [4]. Whereas, electric discharge happened at high microwave power of 6.0 and 7.5 KW.

The denitration test also showed that volatilization rates of alkaline elements increase according to the order of Cs, Rb and Na, whereas Ru added as RuO2 into the HLW-simulated solution was not volatilized. It has been reported that U and TRU nitrates are transformed to stable oxides at 700°C [7], although the denitration test for the sample solution containing those elements was not performed. The design study was carried out for the assumed scale of the denitration step [4].

2.3. Chlorination

Chlorination using chlorine gas together with carbon as the reductant was selected as the preferred method because of recycling of chlorine gas by processing of salt wastes [8]. Chlorination tests were carried out on both the mixed oxide reagents and the denitrated samples obtained by microwave heating, as function of temperature, time, amount of carbon added and flow rate of chlorine gas. And the recovery and volatilization rate for each element and the purity of recovered chlorides were measured. The results showed that nearly 100% chlorination can be attained under suitable conditions.

Figure 3 indicates the effect of temperature on volatilization of Mo, Zr, Fe and U. It is clear that all of the Mo is volatilized at all temperatures of interest, and that other elements...
are increasingly volatilized with increase in temperature. These results were supported by the relation between vapor pressure and temperature for the chlorides [9-11]. Data for the vapor pressure also suggested the possibility of volatilization of Np chloride, because the vapor pressure of Np chloride is similar to that of U chloride. The chlorides volatilized in chlorination step should be recovered by adequate methods, and be supplied to the next partitioning step.

Thus, the chlorination test indicated that the proposed chlorination technology is feasible. Based on the data obtained, a design study was carried out for the assumed scale of the chlorination step [4].

2.4. Pyrometallurgical partitioning

Schematic presentation of pyrometallurgical process is shown in Figure 4. Elements more noble than actinides are reduced to liquid cadmium phase by Li as reductant from salt phase in the reductive extraction step. Afterwards, the actinides are electrowefined in a system of cadmium anode with actinides as well as other elements/eutectic salt of KCl-LiCl/solid cathode. In stead of cadmium anode, the inert anode can be used as well. Figure 5 shows the oxidation/reduction potential of chlorides in a LiCl/KCl salt, which we measured in
Pre-Processing

Chlorides (TRU + FP)

Molten Salt

Liquid Cd

Reductive Extraction

Liquid Cd (TRU + RE)

Cathode

Anode

Recovered TRU

Electrorefining

---

Fig. 4 Schematic presentation of pyrometallurgical process for TRU recovery

A joint study with CRIEPI, Rockwell International Corp., University of Missouri and Kawasaki Heavy Industry, named TRUMP-S Program, though the potential of Cm is assumed from the point of chemical similarity with Am. This figure predicts that some of rare earths are easy to separate with actinides due to the similar potential.

A. Reductive extraction

Prior to the experiment using TRU, the distribution coefficients, defined with the ratio of mole fraction in Cd phase to mole fraction in salt phase, for rare earth elements (La, Ce, Pr, Nd and Y) were determined in a simplified system of molten KCl-LiCl eutectic salt and liquid Cd-Li at 450 to 600°C [12]. Figure 6 indicates the relationship between distribution coefficients of actinides and rare earth against Ce at 450°C. Solid lines in this figure indicate the distribution coefficients calculated by using activity coefficients by electromotive force (EMF) measurements. The electrochemical properties such as activity coefficients for TRU and rare earth elements have been measured in the TRUMP-S Program [13-15]. The distribution coefficients calculated are mostly identical with those obtained experimentally for U and La. The previous data also indicated no discrepancy between measured and calculated values for rare earths, such as La, Ce, Pr and Y. The difference with an order of
Fig. 5  M/MClx potential lines obtained by EMF measurements in the TRUMP–S program

Fig. 6  Distribution coefficients of actinides and rare earths
magnitude was found only for Np. Thus, the distribution coefficients can be explained satisfactorily by using the activity coefficients [2,4,16] of metals and chlorides in salt and Cd phases which were measured electrochemically, which suggests that the calculation using the activity coefficients is useful for the prediction of separation of rare earths and actinides.

B. Electrorefining

Separation efficiency, such as the current efficiency and the effect of current density on separation factor, were evaluated by using rare earths. Figure 7 shows the relationship between the ratio of Gd to Ce deposited in cathode and current density in a system of rare earth-Cd anode/LiCI-KCl/Mo cathode. This figure indicates that the separation factor becomes lower by increasing current density. The actinide chloride is electrolyzed in order to deposit on a tantalum cathode in the salt of LiCl-KCl with some amount of AcCl₃, GdCl₃ and NdCl₃ at 450°C. Figure 8 shows the fraction of Pu removed from salt against current passed (coulomb) obtained in the TRUMP-S program. In addition, the composition of cathode against the coulomb was also determined. The results indicate that actinides can be removed from the salt with high efficiency by allowing the introduction of impurity.

2.5. Salt waste treatment

Pyrometallurgical process produces some amount of alkali, alkaline earth and other chlorides with high radioactivity, which have to be converted to the stable form. Figure 9 shows the flow diagram of salt waste treatment. Lithium is, firstly, selectively recovered into cadmium anode by electrolysis. The Cd-Li alloy and chlorine gases produced are reused in the reductive extraction and chlorination processes, respectively. Afterwards, the remaining salts are electrolyzed by using liquid lead cathode. It was found that all of alkali and alkaline earth metals can be reduced into the lead cathode at 700°C. The cathode added B₂O₃ was oxidized by pouring air at 800°C, which resulted the conversion of nearly 100 % of metals to oxides, forming slag layer. The slag can be vitrified by mixing SiO₂. Based on the experimental results, the mass and heat balances were also elucidated.
Fig. 8 Separation of Pu from LiCl-KCl salt

To reductive extraction process
Salt Waste (LiCl, NaCl, FPClx)

Electrolysis for Li recovery
Salt

Electrolysis for FP/Na Recovery
Pb alloy (Cathode)

Cathode oxidation

To chlorination process

Fig. 9 Flow diagram of salt waste treatment
3. CHARACTERIZATION AND IRRADIATION STUDIES OF METAL FUEL WITH MINOR ACTINIDES

The cooperation study has been carrying out as the joint study with the European Institute for Transuranium Elements to measure metallography and characterization of alloys with minor actinides. In addition, the irradiation program of alloys will be proceeded.

3.1. Metallography and alpha-autoradiography of alloys with minor actinides

The study on metallography of binary alloys of actinides exhibited that actinides are miscible in the molten state, mostly except in the case of U-Am [3]. The multicomponent alloys of $U_{0.03}Zr_{0.15}Y_{0.01}Nd_{0.16}Ce_{0.05}$, $U_{0.45}Pu_{0.19}Zr_{0.11}Np_{0.10}Ce_{0.04}Nd_{0.12}$ and $U_{0.68}Pu_{0.19}Zr_{0.10}Np_{0.012}Ce_{0.008}Nd_{0.014}$ were fabricated by arc-casting in order to measure the miscibility of rare earth in U-Zr or U-Pu-Zr matrix. The metallography indicated that rare earths, which are inevitable to accompany with minor actinides through pyrometallurgical partitioning, hardly appeared to be miscible with U-Zr alloy. The alloys with large amount of rare earths, show that rare earths exist separate from the U-Zr phase. In the case of the alloy with small amount of rare earths, they precipitated uniformly in the U-Pu-Zr matrix. It is also found that yttrium plays a role to pick up oxygen existing as impurity in the matrix.

Afterwards, the multicomponent alloys with americium were fabricated, e.g. U-Pu$_{0.19}$Zr$_{0.10}$ with 2% of minor actinide (Np/Am = 1.5) and 2% of rare earth(Nd/Ce = 2.8), and with 5% of minor actinide and 5% of rare earth by arc-casting. The alloys were annealed at various temperatures. In addition to the alpha-autoradiography, the microstructure of alloys was observed with EPMA analysis. Figure 10 shows the microstructure and EPMA results of alloy with 5% of minor actinide and of rare earth after the annealing at 700°C for 100 hours. It was found that rare earths strongly picked up americium and existed along grain boundaries.

![Composition of the phases (EPMA)](Composition of the phases (EPMA).)

U-Pu-Zr-5MA-5RE (700°C) (wt%)

<table>
<thead>
<tr>
<th></th>
<th>U</th>
<th>Pu</th>
<th>Zr</th>
<th>Np</th>
<th>Am</th>
<th>Nd</th>
<th>Ce</th>
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</thead>
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<tr>
<td>U-rich phase (γ)</td>
<td>66.4</td>
<td>19.9</td>
<td>2.3</td>
<td>3.4</td>
<td>0.6</td>
<td>0.1</td>
<td>0.3</td>
</tr>
<tr>
<td>RE-Am phase</td>
<td>0.1</td>
<td>8.8</td>
<td>0.3</td>
<td>0.1</td>
<td>18.9</td>
<td>54.1</td>
<td>17.7</td>
</tr>
</tbody>
</table>

starting composition

<table>
<thead>
<tr>
<th></th>
<th>U</th>
<th>Pu</th>
<th>Zr</th>
<th>Np</th>
<th>Am</th>
<th>Nd</th>
<th>Ce</th>
</tr>
</thead>
<tbody>
<tr>
<td>U-Pu-Zr-5MA-5RE</td>
<td>61</td>
<td>19</td>
<td>10</td>
<td>3</td>
<td>2</td>
<td>3.7</td>
<td>1.3</td>
</tr>
</tbody>
</table>

U-rich phase (γ)

RE-Am phase 20 μm

Fig. 10 Metallography of U-Pu-Zr-5MA-5RE (after etching)
surrounded by grains consisting of uranium and zirconium co-existing with neptunium and small amount of plutonium. While neptunium exists preferentially in the U-Zr phase, plutonium distributes in both phases of U-Zr and rare earth inclusion. The additional examination specified that minor actinide can be solved in the U-Pu-Zr phase in the case of no existence of rare earths.

3.2. Characterization of U-Pu-Zr with minor actinides

The multicomponent alloys of U-Pu-Zr-5MA-5RE and U-Pu-Zr-2MA-2RE were selected for characterization study based on their metallography. For supply the data on feasibility analysis of reactor irradiation, melting behavior, dilatometry, thermal conductivity, Young's modules and redistribution of components in temperature gradient were measured from the point of fuel characterization, and interaction with stainless steel and compatibility with sodium from the point of the fuel/cladding interaction. Table 1 shows the melting temperature of U-Pu-Zr-5MA-5RE compared with some references. It is obvious that the melting temperature of alloy with minor actinides is not different from that of reference alloy of U-Pu-Zr, which suggests that the apparent temperature doesn't reflect the contribution of rare earth inclusions, but of matrix, i.e. U-Zr-Np with a part of Pu. Similarly, no large difference on thermal conductivity was observed between U-Pu-Zr-5MA-5RE and U-Pu-Zr. In the experiment on redistribution of constituents of alloy with minor actinides and rare earths under temperature gradient, remarkable migration of Am and rare earths was observed over 700°C [18]. In addition, the evaporation and condensation of americium and rare earths occurred at more than 750°C.

The dissolution of Pu, Am and Cm into sodium was determined on the alloy of U-Pu-Zr-MA with Cm-RE by the heat treatment at 600°C for 1 month. It becomes clear that those elements hardly came out into sodium.

<table>
<thead>
<tr>
<th>Specimen</th>
<th>Melting Temperature (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cu</td>
<td>1350 (±10)</td>
</tr>
<tr>
<td>Ni</td>
<td>1720 (±10)</td>
</tr>
<tr>
<td>U61Pu19Zr10Am2Np3RE5</td>
<td>1480 (±10)</td>
</tr>
<tr>
<td>U71Pu19Zr10</td>
<td>1490 (±10)</td>
</tr>
<tr>
<td>[1]</td>
<td></td>
</tr>
<tr>
<td>U Pu19Zr10</td>
<td>1487 (±75) (As liquidus)</td>
</tr>
<tr>
<td></td>
<td>1330 (±75) (As solidus)</td>
</tr>
</tbody>
</table>

3.3. Irradiation study

The fuel alloys of U-Pu-Zr-5MA-5RE and U-Pu-Zr-2MA-2RE together with U-Pu-Zr-5MA are selected as the candidates for irradiation study in the Phenix Fast Reactor, based on the feasibility analysis by CEA, France using the characteristics measured. U-Pu-Zr is also irradiated as reference. Figure 11 shows the irradiation configuration of alloys and arrangement of pins in an irradiation rig. The alloys with minor actinides are sandwiched with U-Pu-Zr on both sides in order to save the MA-materials. The irradiation conditions will be chosen to obtain a maximum fuel center temperature of 750°C and a linear heat power of ca. 400 W/cm. The irradiation continues up to ca. 1.5 at%, 6at% and over 10 at%, which will be initiated in 1994.

4. SUMMARY

The experimental and theoretical approaches indicated that the pyrometallurgical method consisting of reductive extraction and electrorefining devices together with pretreatment step with denitrification and chlorination can be applied for the recovery of transuranium elements from high level liquid waste. The microwave heating method has been verified successfully on denitration of high level liquid waste with some evaporation as oxides. All of the oxides have been converted to chlorides with the aid of carbon as reductant accompanied with some volatilization of a few chlorides. In the reductive extraction step, rare earth chlorides and uranium and neptunium chlorides were reduced into the liquid cadmium phase as function of lithium added as reductant in cadmium. The distribution coefficients between molten chlorides and liquid cadmium determined experimentally showed a good agreement with the theoretical estimation using activity coefficients obtained by EMF measurements except the case of neptunium. The electrorefining technique showed that more than 99% of each minor actinides has been removed from salt phase electrochemically in the simplified system of solid anode/LiCl-KCl with small amount of rare earth and actinide chlorides/solid cathode. The analysis made clear the composition of cathode deposit, i.e. the ratio of minor actinide to rare earths.

---

Fig. 11 Irradiation figure of metal fuels with MA
The metallography showed that rare earths didn't solve in the U-Zr matrix. The rare earths strongly picked up americium and existed along grain boundaries as inclusions. The characteristics, such as melting temperature, dilatometry, thermal conductivity, Young's modules and redistribution of components in temperature gradient, and the interaction with stainless steels, and with sodium were measured on the U-Pu-Zr with some amount of minor actinides and rare earths. The melting temperature and thermal conductivity of the alloys with minor actinides showed apparently the same as those of the U-Pu-Zr by reflecting the contribution of matrix, U-Zr-(Pu) phase. The evaporation and condensation of Am occurred more than 750°C and migration of Am and rare earths became remarkably more than 700°C. It was clear that minor actinides didn't dissolve in sodium even at 600°C.

The alloys of U-Pu-Zr-5MA-5RE and U-Pu-Zr-2MA-2RE together with U-Pu-Zr-5MA are selected as the candidates for irradiation study in the Phenix fast reactor.

REFERENCES

TRANSMUTATION OF LONG-LIVED FISSION PRODUCTS

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Abstract

Arguments for and against transmutation are weighted by three different criteria. For the long-lived fission products Tc-99 and I-129, transmutation would be favored by the criterion, which involves the collective dose due to leakage from repositories. The amount of man Sv avoided might be as high as $10^4$ for each GWe year. It can be shown that especially Tc-99 could be taken care of in several reactor types. There is no incentive to apply accelerator based transmutation to Tc as long as one thinks in terms of continued production of electricity by means of fission reactors. A demonstration in the Petten, High Flux Reactor is described shortly.

1. MOTIVATION FOR TRANSMUTATION OF LONG-LIVED FISSION PRODUCTS

1.1. Arguments for and against transmutation of nuclear waste

Actinides

In literature one finds several arguments in favour of transmutation of nuclear waste [1-4]. Most of these arguments relate to possibilities to reach a more acceptable waste storage strategy, in which the life time of long-lived radioactive components is shortened. One argument in favour of recycling actinides, is related to the wish to utilize the energy content of spent fuel (to reduce the risk and cost related to mining of uranium). Thereby one reduces the long lived actinides in the waste stream as well (at least per unit of electric energy generated). Predictions regarding the integrity of disposal sites might be falsified by human intrusion, which could become attractive especially after the short-lived fission products have decayed and the waste has transformed itself into a more manageable form of partly fissile matter, mixed with only long-lived fission product waste.

Fission products

For fission products the argumentation is quite different. In fact long-lived fission products dominate the risk of stored nuclear waste only at very long storage times. Transmutation of these long-lived fission products is motivated by the wish to reduce possible effects of leakage from geological storage in the far future. Most of the reasoning in favour of the transmutation of fission products is expressed by an "As Low as Reasonably Achievable" (ALARA) principle, and stresses the wish to reduce the radiation dose to generations in a far away future. In this way one might simultaneously increase the public acceptance of geological disposal methods, which has been suffering from distrust in very long-term predictions, due to possibilities of catastrophes or due to accidental mining activities.

Arguments against transmutation are heard as well. Some argue [5] that the recycling process may also dilute the radiotoxicity and could increase risks due to procedures such as machining or due to spilling of solvents. Further it is not at all clear if and how one can compare the radiological risk of partitioning and transmutation with the dose risk reduction for future generations, and in how far one should extend the ALARA principle to these generations. A small additional risk for the present population might be acceptable if it would sufficiently favour future generations. Such favours are however hard to estimate for scenarios, which combine geological disposal with transmutation [5]. Some arguments against transmutation are based on the wish to avoid reprocessing. In the USA for example commercial fuel is not reprocessed at all, in order to restrain the proliferation of the
technology of reprocessing highly radioactive fissionable material, and at the same time to reduce costs. Although safeguarding principles do not concern the fission products directly, it is clear that any partitioning process for fission products also involves the actinides.

**Criteria to weight the arguments** for and against are a key issue in all matters involving transmutation. One criterion might involve the average personal dose in the future due to a possible leakage from repositories. Another possible criterion (especially relevant to intrusion scenarios) involves any incidental dose above average, as measured by the "Annual Limit of Intake" (ALI) for ingestion or for respiration. A possible third criterion regards safeguarding, and it involves the amount of effort needed to produce a weapon from the waste not only now but also in the far future. Transmutation scenarios should be judged by each of these criteria and possibly by other criteria as well. As each scenario also will have a price, it could also be an approach to see this price expressed for example in the amount of ECU's needed to safe a future human life. Costs should compare favourably with investments that society is willing to make for environmental protection of future generations. For oil and coal the loss of human lives to the present generation due to a constant production of one GW(e) lies between 1 and 10 per year. Casualties are almost an order of magnitude lower for nuclear energy [6], and one could argue that also long term risks due to any integrated collective radiation dose, as might be acquired by distant generations, should be lower than long term risks (CO$_2$ and pollutants) due to the use of fossil fuel.

1.2. The situation for fission products in particular

**Amount of fission products**

It is a fact of nature that the use of fission energy will lead to waste due to fission products and actinides. For the actinides the amount of material to be formed is highly dependent on the scenario which one might want to use. For the fission products however the amount is rather independent of the scenario and it is roughly equal to 1000 kg for one GWe year. Almost 90% of this amount is either stable or decaying within a few hundred years to stable products. The long-lived remaining part consists for about 35% of the geochemically mobile fission products Tc-99 and I-129. As it is presupposed that certainly for such a relatively short time the integrity and the safeguarding of any disposal site can be guaranteed, the short lived products are of no concern in the present study. After all in most world scenarios one foresees a few hundred years as a period in which one may safely rely on the knowledge and skills of our descendants to maintain man made barriers.

**Conditioning and storage possibilities**

In current waste management scenarios the fission products are vitrified or conditioned in some other way in order to make them less mobile. In case that waste disposal sites would not be disturbed, and if the waste remains immobilized satisfactorily, the leakage from these sites would hardly have consequences for the radiation dose to mankind. Numerous studies on consequences of nuclear waste stored in salt domes show virtually no effects to mankind. An example of a natural conditioning of radioactive material is given by the thorium in monazite sand [7]. At some beaches the radiotoxicity of the sand may approach that of highly diluted nuclear waste, but the thorium atoms in the sand just happen to be immobilized entirely just like the toxic mercury atoms in our dental fillings. Apparently at least in this case properly immobilized material is not the problem. For nuclear waste the problem rather is to guarantee that all radiotoxic elements should remain immobilized in the long run. Geologists and chemists take great care to think over possibilities to immobilise all waste elements almost forever, but have not yet convinced everyone. This is especially so for Tc-99 and I-129, as these may remain mobile in the environment for many millennia [8]. Mobile elements may have a large impact on the estimated dose risk integrated over the entire population in the long run even if the amount is marginal. A natural example is the highly mobile radon,
a gaseous radiotoxic emanation of radium, the daughter product from the decay of uranium in the soil. This radon dominates the radiation dose to mankind at the moment.

**Dose risk of mobile long-lived fission products technetium and iodine.**

If one would express the dose risk of the mobile long-lived fission products like technetium and iodine in relation to for example the natural dose risk due to radon from the soil, one encounters some difficulties. In particular for the man-made element technetium there are no well-proven models for its transport (acidity and oxidation potential of the soil play a crucial role) and the methods of conditioning have not been tested over more than decades. A pessimist might therefore take an extreme point of view by assuming that all mobile long-lived material will just leak away within a few thousand years. By taking this point of view, the accumulated collective radiation dose from technetium will approach 9,000 man Sv for a GW(e) year of nuclear electricity but for iodine this dose seems much smaller (as can be seen from Table I and Ref. [8]). Although these radio-nuclides live so extremely long the question whether one uses direct storage or conditioned (vitrified) storage, might still be very important, and a more optimistic point of view regarding the mobility of technetium could yield a much lower dose. Table I has been extracted from a UK study [8] on deep reposition in granite, and illustrates long-term risks. This table presents, for a normal evolution scenario (without human intrusion) the collective dose in man Sv, due to 30 t of LWR spent fuel. This amount of fuel would arise from the electricity production of a GW(e) light water reactor, during one year of operation. Relevant nuclides are given for directly stored fuel. In Table I the collective dose is integrated over one million years and over 10^8 years (maxima for individual doses are not considered). From this table it can be concluded that if one does not transmute the waste at all, the integrated collective dose as accumulated over one million year will be less than ten thousand man Sv. If only technetium could be immobilized, one might avoid most of the expected accumulated dose.

**TABLE I. TIME-INTEGRATED LEAKAGE DOSE DUE TO SPENT LWR FUEL**
(direct storage of spent fuel, due to nuclear generation of one GW(e) year)

<table>
<thead>
<tr>
<th>Period</th>
<th>One million years</th>
<th>Hundred million years</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nuclides</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Tc-99</td>
<td>98%</td>
<td>46%</td>
</tr>
<tr>
<td>I-129</td>
<td>2%</td>
<td>1%</td>
</tr>
<tr>
<td>Cs-135</td>
<td>2%</td>
<td>24%</td>
</tr>
<tr>
<td>U-235</td>
<td>6%</td>
<td></td>
</tr>
<tr>
<td>U-238</td>
<td>14%</td>
<td></td>
</tr>
<tr>
<td>Np-237</td>
<td>5%</td>
<td></td>
</tr>
<tr>
<td>Pu-239</td>
<td>4%</td>
<td></td>
</tr>
<tr>
<td>man Sv</td>
<td>9000*</td>
<td>20 000*</td>
</tr>
</tbody>
</table>

*Collective dose for the global population. The average yearly individual dose may be obtained by dividing by the affected number of people and the indicated period.

Although these estimates are based on crude assumptions, it is clear that the postulated leakage of Tc-99 will only lead to very small personal yearly dose rates. In this way one would come to the conclusion that the influence of the long-lived fission products is only marginal. The million year Tc-99 dose should be divided by one million to find an average

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yearly dose integrated over the global population and this small result ($10^{-2}$ Sv) should be divided by the amount of affected people in order to get the expected individual yearly average dose. Some feeling for the relevance of this number is given by comparison with the "natural" individual dose rate from radon (About 60 t of radium in the soil emanates almost 6 litre of radon each day, which builds up an equilibrium value of about 20 litre of radon in the total biosphere. This tiny amount nevertheless gives about 60% of the total radiation dose, which averages to about $10^{-3}$ Sv per person yearly and leads to a collective dose of about $5 \times 10^6$ man Sv/year), which is of the order of $10^{-3}$ Sv per year.

Only sudden releases of radiotoxic material, caused by human intrusion or catastrophes, could change this picture. Such scenarios, though very unlikely, may finally be the main justification for transmutation of the actinides. For long-lived fission products a word of caution should however be given against any argument with respect to the value of a future human life as if it were a parameter in some mathematical expression, which is valid over many ages. A modest investment in decreasing the radiation dose (for example a reduction in the radon dose) could easily be so rewarding that present comparisons become irrelevant. Moreover it has not yet been shown satisfactorily that it is correct to use the linear dose-response rate without any threshold effects, as was done implicitly in the reasoning above. It therefore is certainly is not valid to offer a price for each man Sv avoided, and certainly not at avoidance costs of about 100 kECU per man Sv [1], as spent in radiological work.

It has been noted [4] that results of the UK study [8] rely heavily on assumptions on the effectiveness of vitrification and on geological characteristics of the repository. These may not be quite representative for other repositories, as calculations are based upon site dependent geo-chemical characteristics. If the waste is not in contact with oxygen, the mobility of Tc will be strongly reduced. In clay repositories [10] or in rock salt [11] repositories the I-129 risk could dominate, especially if one would rank the risk not as in Table I (a collective integrated dose), but rather according to the highest possible individual dose. Any such dose could be caused locally and incidentally, and it would be mainly due to uptake of the iodine in the thyroid (the quantity of iodine in this gland is about 10 mg). It is felt, however, that isotopic dilution with the natural iodine could reduce the highest possible individual dose due to the long lived isotope I-129. If one for example dilutes the I-129 with about a factor 500 with natural iodine, the resulting mixture might have a radiological impact less than that of the small amount of the natural isotope K-40 in food.

Although there are different scenarios for deep geological repositories and different definitions of risk, it nevertheless can be concluded that - at very large storage times - fission products will dominate the collective risk in case of direct storage of spent fuel. As was mentioned above, removal of Tc-99 and I-129 from the waste would reduce the collective one million year dose to negligible amounts (the reduction factor would be close to the decontamination factor in the partitioning process). For the actinides, however, Table I would hardly give arguments for transmutation or vitrification, still assuming integrity of the site and trusting assumptions regarding mobility, etc. If one wishes to reduce the integrated actinide leakage dose from undisturbed repositories by means of vitrification or transmutation, this would mainly apply to uranium. It can therefore be concluded that reduction of risk by transmutation of long-lived fission products seems only rewarding if one wishes to reduce the collective dose at extended storage times. Actinide transmutation is justified on different grounds, as it reduces the source term rather than the leakage dose of stored waste, already after relatively short storage times (a few hundred years).

2. PARTITIONING AND TARGETS

Partitioning of fission products

In the presently used PUREX/TRUEX process the spent fuel is removed from the canning and dissolved in nitric acid. The parts which are not solved are called the "fines" and
about one third of the technetium is to be found in these fines. The uranium and the plutonium are extracted from the solution by an organic solvent, tri-n-butyl phosphate (TBP). Later the uranium and the plutonium are again separated by selective reduction of the plutonium (VI) to plutonium (IV), but this has no consequence for the present discussion of the partitioning of the fission products.

Gaseous fission products, such as xenon and krypton and a large part of the iodine are liberated in the process of dissolving the spent fuel, together with the gaseous oxides of nitrogen from the reduction of the acid solvent. Currently noble gases are vented into the air and most of the extracted iodine is gradually diluted into the sea. In order to reduce the dose due to the short lived iodine isotopes, several possibilities exist already to catch the iodine either in alkaline solutions, or in zeolite filters loaded with silver, or in mercury or lead compounds. If one would like to transmute the I-129, these possibilities could be more fully exploited.

The chemistry of technetium in the reprocessing procedures is quite complex. Technetium will be present in 45-70% of the total inventory in the water phase in the form of $\text{TcO}_4^-$, a pertechnate ion. For the PUREX process the concentration is about 40 mg/litre. Some small part of the technetium (about 5% of the inventory) is co-extracted in the organic phase because of the formation of complex compounds between the TBP, the uranium oxide, and the nitric acid. A third fraction (25-50%) of the technetium is present in the fines, which do not easily form a solution in acids. It is technically possible to separate the technetium from the watery phase and from the organic phase, but it is more difficult to extract it from the fines. If at any time the transmutation of technetium should become a current practice, it will be necessary to first solve the problem of the technetium in the fines. Present techniques of solvent extraction might either have to be supplemented with pyro-chemical techniques (which involve liquid salts) or quite different solutions should be searched for. Use of the necessary technology on a large scale will need a major effort in the future. Now up to 90% of the technetium could be recovered on a laboratory scale; the large scale recovery losses should be reduced at least to a few percent in order to prevent too large losses of the technetium, which otherwise would be spilled into the waste.

**Targets for irradiation (homogeneous versus heterogeneous)**

If one wishes to transmute technetium and iodine, the stability of the compounds to be irradiated is of utmost importance. In homogeneous methods one mixes the compounds through the fuel and in heterogeneous methods one physically separates the targets for irradiation. Homogeneous methods are already applied for actinides (MOX) but for fission products it seems more advisable to separate these from the fuel reprocessing cycle. After all, the chemistry of technetium is complex and one does not want to complicate the reprocessing of actinides by admixing substantial amounts of technetium through the fuel. Another possibility would be the application of thin foils of technetium near the cladding of the fuel. At ECN physical and technological aspects of fission product transmutation are studied and demonstrations of irradiations are being prepared for heterogeneous targets [13].

**Research and demonstrations**

For experimental and technological research on transmutation an European network EFFTRA (Experimental Feasibility of Targets for TRAnsmutation) has been founded. In this network ECN participates together with the French organisations CEA and EdF, with the German KfK and the EC-establishment TUI. In the laboratories of TUI, CEA and ECN, targets are prepared, irradiations will be performed in the Petten HFR by ECN (see Fig. 1 and Ref. [13] for a description of the setup), and later also in the French Phénix reactor. After finishing the irradiations, the targets will be examined to study transmutation efficiency and material aspects. During this work the specific competencies of each of the partners can be exploited.
optimally. Most of the EFTTRA programme is focused on the irradiation of the actinides (specifically the americium isotopes), but also the fission products iodine and technetium will be studied. Specific problems to be studied first of all are the chemical and material properties of the candidate target materials. Many aspects play a role such as phase transitions in the materials, sensitivity for radiation damage, and the interaction with the cladding. Transmutation of technetium into stable ruthenium is most suitably performed by irradiating it as a metal or an alloy in a thermal neutron field. Demonstrations are relevant because this element does not occur in nature, and therefore chemical and material aspects of technetium are not yet well known. Therefore, also the interaction with the cladding during irradiation is of relevance. Although iodine is a well-known element, research of iodine compounds is needed as well, mainly because iodine is chemically aggressive. An extra complication in the case of transmutation of iodine is due to the fact that transmutation in a thermal neutron field will lead to (stable) xenon gas, which will cause swelling of all solid compounds which are imaginable. Several compounds (metallic iodides) are being tested, and cerium iodides are considered as most promising for xenon retention and stability. However, very recent results obtained at ECN in Petten, indicate that cerium-iodide reacts with the stainless steel cladding [20]. Therefore, other iodides are investigated at present. Another line of research would be the irradiation of liquids such as eutectic compounds of lead, tin and iodine, while removing the gaseous products either on-line or at intervals.

3. TRANSMUTATION DEVICES

For transmutation of fission products thermal neutrons and/or epi-thermal neutrons are needed, and other sources of neutrons might be relevant than those for transmutation of
actinides (which in some cases may require fast neutrons to avoid capture and to favour fission). As neutron sources one knows reactors and accelerators. Some possibilities will be shortly reviewed with respect to suitability for transmutation of long-lived fission products. Relevant questions related to the neutron source are: is it intense and economical, is its own polluting effect small enough, and what are the safeguarding aspects of these devices? Material irradiation aspects have been mentioned in the chapter about targets. It is felt that these aspects are the same whatever transmutation device one chooses; this assumption should be tested, however, after a specific choice has been made and after analyzing the demonstrations, which have been mentioned in the previous chapter under "Research and demonstrations".

3.1. Transmutation in reactors

Nuclear reactors are the most obvious neutron sources as they are abundant in the world, and they may deliver neutrons during the process of energy production. Reactor physics problems are related to the optimization of the flux of excess neutrons in the reactor, and to the influence of the targets to be irradiated on the safety properties of the reactor.

High flux reactors

From the shielded one-group cross section of the fission products I-129 and Tc-99 (about 12 and 7 b, respectively) in a high flux research reactor such as the Petten HFR, one might conclude that a high transmutation rate of the fission products (50% transmutation within about three years) would be feasible [9]. This 45 MW(th) reactor would however have an amount of excess neutrons which is much too low to load a high amount of fission product waste. A drastic change in reactor parameters and a much higher power would be needed for a rapid transmutation of huge amounts of material in any high-flux reactor. Further it is necessary to study material properties of targets under irradiation.

Light water reactors

If one would accept a lower transmutation rate than in the HFR scenario, reactors with a lower neutron flux could also be used. In a regular LWR power reactor the flux would be an order of magnitude lower than in an HFR, and therefore the effective half life for destruction of the fission products would be at least ten times longer then the fuel cycle time. If one wishes to transmute an amount of technetium of the same order as the Tc production of a same LWR, one would require several tons of technetium in the reactor. Because this technetium has to be present during the full life time of the reactor, it should be reprocessed and/or recanned dozens of times before it is entirely transmuted into ruthenium. This would increase the reprocessing cost of the fuel cycle, but it could also have some impact on the radiation dose of the personnel. Similar considerations apply to the transmutation of I-129.

Other reactor types

From the above reasoning it is clear that the choice of reactor type is dominated by the flux of thermal and/or epi-thermal excess neutrons in the reactor. With fast reactors one might produce a high thermal flux by means of moderating material on the spot of the target. High transmutation rates for such irradiation positions are foreseen in studies made at ECN [9] and CEA, and recently by Westinghouse [18]. Also an EC study has been made by Siemens on large scale transmutation possibilities, in the frame of the CEC strategy studies on transmutation of nuclear waste [21]. These possibilities could be attractive, in particular in combination with nuclear incineration of plutonium and minor actinide waste. A heavy water moderated reactor like the CANDU has some advantages as a transmutation device over other reactor types: it combines a high thermal flux with sufficient excess neutrons and flexible loading during reactor operation [12,14].
Summary

In principle current LWRs could be used for a massive transmutation of Tc and perhaps I, but this would require huge loadings of these materials, additional enrichment and additional reprocessing/recanning efforts. Fast reactors and HWRs have attractive potential with respect to transmutation in moderated assemblies, but more study is required to assess the optimum technology for transmutation of long-lived fission products on a large scale. HWRs like CANDU have easy refuelling possibilities, which may be an additional advantage in the application of heterogeneous transmutation.

3.2. Accelerators as neutron sources

General aspects of accelerator based transmutation

By means of high energy protons (one GeV or more) neutrons are produced through the spallation process. These neutrons (with an energy of the order of 10-20 MeV) can be either used directly to hit actinide nuclei or they can be moderated to transmute actinides and/or fission products. Each high energy proton might liberate dozens of neutrons (see Table II) in a target of heavy elements.

By simple reasoning it is easy to show that most fission-product transmutation will take place by capture of the thermal neutrons rather than by any process in which charged particles are involved (these have smaller cross sections due to the Coulomb repulsion). Because thermal neutrons are most relevant for fission product transmutation, one thinks of neutron boosters. These are sub-critical systems with fissionable material in which each neutron from the accelerator-target system might produce again up to ten new fission neutrons. In fact it has been shown that a sub-critical system will be more economical than a system without a booster. The argument is as follows: One 1.5 GeV neutron produces 30 neutrons in a tungsten target without a booster. Suppose the accelerator efficiency is about 50% (a very optimistic statement). In this case the price of one thermal neutron expressed in terms of electrical energy is about $1500/(30*0.5) = 100$ MeV. As this electrical energy had to made from thermal energy one would have required about three times as much energy. Because it is extremely optimistic and even unrealistic to assume that each neutron will transmute a technetium nucleus, one will need much more than 300 MeV thermal energy for each technetium nucleus to be transmuted. In case that this 300 MeV would have been generated by means of a nuclear reactor, this would mean that more than one fission in the reactor is needed to transmute one technetium nucleus at the accelerator.

Following this reasoning it seems however that direct transmutation of technetium in a reactor is much more economic in case that also one free neutron might become available for each fission in the reactor itself. The reactor method is clearly more direct, and secondly

<table>
<thead>
<tr>
<th>Proton energy GeV</th>
<th>Number of spallations</th>
<th>Number of emitted neutrons per proton in the beam</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Pb</td>
</tr>
<tr>
<td>1</td>
<td>3</td>
<td>17</td>
</tr>
<tr>
<td>1.5</td>
<td>5</td>
<td>20</td>
</tr>
<tr>
<td>2</td>
<td>7</td>
<td>23</td>
</tr>
</tbody>
</table>
the 200 MeV, which is generated by fission, can still be used to generate electricity; in the "pure reactor scenario" this electricity is no longer needed to transmute the technetium nucleus with an accelerator. Any combination of an accelerator and a booster seems to approach the economy of the reactor method more closely if the setup is more close to criticality. It has been shown by arguments of mass and energy flows that hybrid systems are not favored at the moment [12].

Some scenarios

In the USA, the Los Alamos Laboratory proposed a set of special hybrid accelerator-reactor systems, the so called Accelerator Transmutation of Waste (ATW) systems [15]. In these systems a very high powered (100 MWe) high energy proton beam plays a key role. Many possible sub-critical reactor systems have been proposed as booster systems. One of the advantages of the high powered proton beam is that one may use a neutron booster with a relatively low value of $k_{\text{eff}}$. As an actinide transmuter this system is supposed to be equipped with either a molten salt reactor or with a graphite moderated lithium-7 cooled system [16]. Arguments in favour of ATW systems are based on relaxation of safety procedures if $k_{\text{eff}} < 1$ and this would lead to easier licensing possibilities. An other argument used is that the inventory of radiotoxic material is small while the transmutation rate could be the same as in a reactor. Because of the simple and natural laws of nuclear physics this means that one should work with a very high flux and with diluted fission material inventories. As however the liquid reactor concepts ($D_2O$ moderated slurries or molten salts) suffer from criticality problems due to possible loss of homogeneity, the present tendency is for solid (graphite moderated) systems, which could even work on thorium in order to relax the problem of actinide waste at the same time [16]. In order to extract a high power from a small system, one should cool with a liquid metal with a low cross section for neutrons, and one selects Li-7 as a coolant. Time obviously will have to learn whether this system could transmute fission products economically, and be competitive. One of the natural facts for any effective incinerator of fissile material is that a large energy flow from the setup should be dealt with, and that the power density at the target should be high. Economy of incineration might require an approach close to $k_{\text{eff}} = 1$ (a pure reactor) in the end [12].

In the USA (Brookhaven) and in Japan (JAERI), systems have been proposed, which are basically a small (relative to ATW) accelerator with a fast reactor setup as a sub-critical booster [17]. In these PHOENIX and OMEGA proposals one wishes to incinerate the same amount of actinides as in the ATW proposal (the system should be able to compete). Because the accelerator is more modest, the power of the reactor should be high and consequently the value of $k_{\text{eff}}$ should be close to one. In fact this is a fast reactor with possibly some improvement of the safety, which is caused by the fact that it is partly regulated by means of an accelerator beam. Because one may trust in possibilities to switch off the beam, one is in a position that a new and very fast "electronic safety rod" has been introduced this way. Although such fast reactor booster systems might have very attractive features (especially if one wishes to fission the minor actinides and the even plutonium isotopes), these systems apparently suffer from a lack of thermal neutrons and are less suited for the transmutation of fission products. Arguments to be followed in order to reach this conclusion proceed along the same line as for fast reactor with a "mechanical safety rod". Likewise accelerator systems with moderated targets are open to consideration.

4. CONCLUSIONS AND RECOMMENDATIONS

Risk reduction and allowable costs of recycling Tc-99

One of the problems with technetium is that it is an element which does not occur in nature, and that one has little experience how it will behave in the ecosystem. It has been shown above that total transmutation of technetium could save future mankind some risk due
to possible leakage of this radio nuclide at very large storage times. This risk is rather small, but it exceeds the risk of actinides (see Table I). Justification for transmutation of actinides is mainly based on assumptions of human intrusion or on natural catastrophes. The same argument could to a smaller extend be applied to justify transmutation of technetium. It would be important that costs are kept small with respect to the current fuel cycle costs [4]. This seems possible [12], provided that chemical partitioning and reprocessing costs for Tc are low.

Risk reduction and risk of I-129

Two points are in disfavor of transmutation of iodine. First of all the amount of man Sv due to a long term global I-129 contamination would be an order of magnitude lower than that from technetium. Secondly iodine is an element which is rather abundant in nature and the technique of isotopic dilution probably can be applied safely. If one for example dilutes the I-129 with about a factor of 500 with natural iodine, the resulting admixture might have a radiological impact smaller than for example the free amount of potassium in the biological cycle of any living creature. Such a small toxicity has never harmed life as far as we know, and the cost and risk of iodine transmutation should be compared with this marginal risk. As these costs and risks would be much higher for iodine than for technetium, the conclusion could be that isotopic dilution could give a solution if it were to receive public acceptance. Meanwhile both partitioning/transmutation as well as isotopic dilution strategies ought to be further evaluated.

Recommendations

For the time being it is recommended to study technetium, its dose consequences and its ecology, as well as its chemical nature especially in the solid state. The time-accumulated dose related to technetium dominates the leakage doses in most scenarios for unperturbed geological disposal. If human intrusion into geologically stable repositories or other disturbances is taken into account, the actinides determine the maximum value of the expected individual dose rates at shorter storage times. Therefore actinides and safeguarding problems related to the whole Partitioning and Transmutation scheme dominate the discussion on transmutation of nuclear waste. The fact that safeguarding procedures should be accounted for, might lead to considerable complications in scenario's for partitioning the actinides from the fission products.

Issues related to public acceptance

Most discussions on nuclear waste, and also the present discussion are based on average quantities such as given in Table I. Any such probabilistic assignments of risk over extremely long periods appear to create suspicion. It seems that public confidence in predictions that range over more than a few thousand years is diminishing. It should be realized that the problem of public acceptance is too complex to be approached in a purely scientific/technological way. As it nevertheless is necessary to sketch a frame to approach the waste problem, one could start by investigating whether transmutation of fission products is technically feasible or not.

ACKNOWLEDGEMENTS

The CEC is acknowledged for a grant supporting this project, and I wish to thank my ECN colleagues H. Gruppelaar and A.J. Janssen for many critical remarks.
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ANALYSIS OF THE PRELIMINARY RESULTS
OF THE RUSSIAN P&T RESEARCH PROGRAMME

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Abstract

Russia has a large programme on partitioning and transmutation (P&T) of actinides. A lot of research centers directed by Ministry of the Russian Federation for Atomic Energy (Minatom) involved in this activity. The current status of the russian programme on P&T is discussed in this paper.

BACKGROUND

During the session of Minatom Scientific and Technical Council held in October 1993 several papers devoted to the prospects of Partitioning and Transmutation projects were presented. Proposals, being developed by the scientists from the Institute of Physics and Power Engineering (IPPE, Obninsk), Institute of Theoretical and Experimental Physics (ITEPh, Moscow), Scientific and Research Institute of Atomic Reactors (SRIAR, Dimitrovgrad) and some other were discussed among them.

It was accepted, that P&T concept of the long-lived radioactive wastes should be considered as a component for the future large-scale nuclear power industry. That means step-by-step transition towards closed nuclear fuel cycle for all types of spent nuclear fuel and utilization of the recovered fissile materials.

THE PROGRAMME

It was agreed that an expedience of the P&T option needs to be scrutinized. Such factors as arising secondary wastes, additional transportation of highly radioactive materials, possible increase of personnel doses and pollutions are to be taken into account.

Preliminary evaluations show that P&T option is over twice as much expensive as direct disposal of solidified wastes. Further systematic analysis should be carried out, taking into account all the factors involved.

Implementation of the nuclear transmutation of the most dangerous long-lived radioisotopes will be closely connected with:
- Extensive long-term development of the closed nuclear fuel cycle industry;
- Construction of the special or dual-purpose (power and transmutation) nuclear installations;
- Special fuel elements and/or target fabrication;
- Reliable final disposal of all the radioactive wastes arise;
- Acceptable level of the technical and environmental risk for all the additional P&T stages and facilities involved.

It was decided that no reasons exist for the transmutation of the most fission products, except Tc-99 and I-129. Therefore it is necessary to reach maximum possible level of minor actinides (MA), Tc and I extraction from the wastes to be disposed. Separation coefficient for Np and Cm isotopes should be as high as 0.99 and for Pu and Am isotopes - no less than 0.999.

Possibility of I-129 transmutation is quite problematic, on one hand due to the presence of statable iodine isotopes in the mixture and on the other - due to the absence of suitable target form for the element.
The most advanced transmutation method is based on MA burning in the specially designed, or commercial fast neutron reactors. In Russia this method is developed by the scientists from the IPPE (Obninsk) and SRIAR (Dimitrovgrad). Preliminary results show that Na-cooled BN-type reactor could be effectively used as MA and Pu burner. For the nuclear safety, concentration of the MA in the MOX BN fuel have to be less than 5%.

Under those conditions one BN-800 reactor can transmute up to 100 kg of MA per year that is yearly MA amount from 3 units of the WER - 1000 type.

Details of the IPPE project are presented to your attention in a report of Dr. N. Rabotnov. Achievements in radiochemical separation technology are presented in a paper of Dr. V. Romanovskij.

Scientists from the Minatom's Scientific and Research Institute of Atomic Reactors are carrying out experiments to ground the fuel cycle based on "dry" fuel reprocessing method for transmutation of MA in the fast reactors. It is so called "DOVITA" project. The work includes research and feasibility study of the compact pyroelectrochemical technology for production and reprocessing of Np, Am and Cm contained uranium and/or MOX fuel, as well as vibropacking technology for fuel element production.

On the RIAR site there are unique experimental nuclear complex which includes:

- experimental fast reactor BOR-600, high flux reactor SM-3, experimental reactor MIR, RBT-6 and RBT-10;
- facilities of the Test Research Complex (TRC) for production vibropacked MOX fuel elements and assemblies for fast reactors;
- pilot facility for pyrochemical reprocessing of irradiated nuclear fuel;
- other supplementary radiochemical laboratories and facilities.

Despite of apparent advantages of the proposed method, comparison should be done for water-extraction and pyroelectrochemical FBR fuel reprocessing including waste management issues.

Proposed by ITEPh scientists nuclear transmutation facility based on a linear proton accelerator that generates energy in the range 0.8-1.6 GeV. On a heavy element (Pb, W) target those protons are converted to neutrons. Target area is surrounded with heavy water blanket where a high thermal neutron flux is formed. Wastes to be transmuted contain I-129, Tc-99 and MA, and circulate in the form of the solution through the blanket and heat exchanger.

The heavy water solution is simultaneously a heat generator and a coolant. A portion of the flow could be taken for reprocessing continuously or by portions (batch-type reprocessing). Radiochemists from Scientific Research Institute of Inorganic Materials (SRIIM, Moscow) have developed special extraction-membrane method for the transportation of resultant stable isotopes from heavy-water to the light-water medium for further processing.

A lot of complicated and knotty radiochemical and technical problems should be solved for implementation of proposed P&T method. The accelerator based P&T facility construction and operation seems to be unpredictably expensive.

As a result of the meeting mentioned above (in Minatom) it was decided that main directions for the future activities of Minatom's scientific centers should include:

- calculations and analytical research to choose and ground radioecological criteria and requirements to the back end stages of the closed nuclear fuel cycle;
- development of the advanced radiochemical partitioning methods to optimize the separation coefficients of the most important elements and groups of elements;
- experimental measuring and investigation of the necessary nuclear and other constants of the new radioactive materials, involved in the nuclear fuel cycle with P&T;
- development of the new radiochemical and fuel fabrication methods (electrochemical and pyrometallurgical) for MA contained materials management;
- carrying out transmutation experiments with MA and Tc on the nuclear reactors in operation.
- calculations and experiments to optimize reactor core for MA and Tc transmutation.

The goal of the above mentioned programme should be development of the well grounded and ecologically safe waste management concept in the frame of the closed nuclear fuel cycle including definite transmutation method. The work will be carried out as Minatom's Coordinated Experimental and Research Programme in the frame of Russian Federal Waste Management Programme.
FUEL RECYCLING AND RADWASTE TRANSMUTATION

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Abstract

Transmutation of actinides including plutonium is mainly considered because fission products transmutation is practically blocked by low absorption cross section of Sr-90. Tc-99 is the only likely candidate for transmutation among FP. Fast reactors are potentially the main tool of actinide burning. In thermal reactors low fission to absorption ratio for most of the isotopes leads to accumulation of very radiotoxic isotopes and to neutron losses. Radiochemical partitioning should be optimized to minimize irretrievable losses without unnecessary separation of fractions to improve economic efficiency. A calculational example of BN-800 reactor burning VVER-440 actinide discharge is considered. It results in the manifold reduction of the actinide inventory except curium. Some results and programme of transmutation related studies in IPPE are outlined and key issues of further research listed.

Possibility of transmutation does not exclude final geologic disposal. Many questions are still to be answered including the most important one - is the transmutation really necessary, are the costs and problems involved justified? So not only prospects but difficulties as well are to be considered [1,2].

It is also clear that thermal reactors will be the base of nuclear power for 2-3 decades, corresponding research and development taking proper share. A problem of next priority level - fast reactors. The task of more efficient utilization of natural uranium resources is postponed, not canceled. The accumulation of reactor and weapon grade plutonium calls for its use also. Fast reactors provide solution of both problems. Plutonium utilization in fast reactors is naturally connected with burning of the most potentially dangerous component of the radwaste - minor actinide. Plutonium and MA provide additional incentives for the fast reactors development. They are real, industrial scale facilities with about a hundred reactor-years of experience behind them, they belong to the present just like thermal reactors.

But alternatives are also proposed. They have problems still to be overcome and their practical use is far ahead. Electroneutral method belongs to this category. It was proposed some forty years ago but is still to become technical reality. That's a problem of third priority level.

The strategies of managing medium-lived (heat producing) and long-lived nuclides are to be different. Because of the small thermal capture cross section of Sr-90 [3] its transmutation looks impossible and the transmutation of the rest of medium-lived fission products is irrational. On the other hand large heat production and radiation damage to the matrices due to intense heating make immediate final disposal dangerous so it must be preceded by log-term controlled storage, most probably for a few hundred years.

Among long-lived fission products I-129 presents special problem due to high movability in the rocks and volatility in reprocessing. Its efficient transmutation in high flux reactors is in principle possible but solid compound targets may present problems. So I-129 is very important limiting factor in the actinide recycling and burning. Its effective capture and binding in reprocessing is a top priority.

Most probable candidates for the transmutation list are minor actinides and technetium which has large enough thermal capture cross section, capturing up to three neutrons turns into stable isotopes of ruthenium and is a suitable material for reactor targets.
Average fission to capture ratios for threshold nuclides are by an order of magnitude higher in fast reactors than in the thermal ones and that's an important advantage. In high thermal neutron fluxes a considerable part of actinides turns into curium isotopes instead of fissioning. That may not be considered a satisfactory result of transmutation because medium-lived Cm-244 has very high activity and heat production. And it decays to Pu-240. So one gets "squirrel wheel" - spending four neutrons to turn Pu-240 into Cm-244 we return to the start with a very dangerous nuclide as an intermediate stage. And Cm-248 is more radiotoxic than any other actinide in the spent fuel because 8 per cent of its decay is spontaneous fission and total decay period is most inconvenient - 340,000 years.

The accumulation of the threshold nuclides is a decisive disadvantage of the thermal reactors as actinide burners, the total radiotoxicity accumulated in recycling is growing with time. It is not so in fast reactors.

But curium recycling in fast reactors creates some problems due to high activity and heat production. That stimulates looking for some ways of its extracting from the wastes and cooling for a few decades.

Burning rate of 3.5 per cent of actinides added to BN-800 fuel (14 per cent to plutonium) is about 100 kg/year, so support ratio to VVER-1000 is 3. Actinide admixture of a few per cent does not change fast reactor physics much. Higher burning rates may be reached in a specialized BN-800 core with U-238 replaced by inert matrix like zirconium carbide. Fuel composition may be optimized to provide zero sodium void coefficient and acceptable beta-eff. This optimum corresponds to MA/basic fissioning isotope ratio in fresh fuel on the level 1:3. Such a 2000 MWt(t) reactor burns some 350 kg of MA a year (support ratio about 10).

Actinides may be added to reactor fuel in different ways. The addition of preextracted Np and Am is most straightforward. But there are other schemes attracting attention recently with either incomplete or no partitioning of plutonium and neptunium.

Incomplete partitioning of Pu and Am is also to be considered as well as relaxed limits on fission products separation with resulting reduction of reprocessing costs in MA recycling. In particular, Dimitrovgrad Atomic Reactors Research Institute proposes this approach [4].

Fast reactor transmutation meets with rather serious problems. Radioactivity and heating of fresh MA containing fuels is by many orders of magnitude higher than traditional uranium PWR fuel. That complicates all the operations with fresh fuel - making, storage, transportation. Growing variety of fuel compositions is common difficulty of all transmutation schemes. But the factor determining the success or failure of MA burning is the possibility to ensure low enough irretrievable losses of Pu and MA going to the waste. Partitioning of MA one from another as well as from U and Pu is not too important for fast reactors their physics being benign to moderate variations in fuel compositions.

The main problem of MA partitioning and burning in fast reactors seems to be the minimization of irretrievable losses and of the total volume of radioactive waste to ensure an acceptable economy of fuel cycles including transmutation.

Now we consider a computational example - the effect of BN-800 reactor operation reducing the radiotoxicity of Pu and MA accumulated in reprocessing of VVER-440 reactors spent fuel [5]. Evaluated volumes of actinides accumulated to a year 2000 are presented in Table I.
TABLE I. ACTINIDE ACCUMULATION IN VVER-440 REPROCESSED SPENT FUEL BY THE YEAR 2000

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Mass, kg</th>
<th>Nuclide</th>
<th>Mass, kg</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu-238</td>
<td>165</td>
<td>Am-241</td>
<td>420</td>
</tr>
<tr>
<td>Pu-239</td>
<td>11,564</td>
<td>Am-243</td>
<td>146</td>
</tr>
<tr>
<td>Pu-240</td>
<td>4,173</td>
<td>Np-237</td>
<td>550</td>
</tr>
<tr>
<td>Pu-241</td>
<td>2,350</td>
<td>Cm-244</td>
<td>41</td>
</tr>
<tr>
<td>Pu-242</td>
<td>779</td>
<td>Cm-245</td>
<td>2</td>
</tr>
</tbody>
</table>

Three situations are considered:

1. BN-800 is not put into operation, the actinides are stored to 2060.
2. BN-800 reactor is put into operation at 2000 and burns traditional MOX-fuel without MA.
3. BN-800 is put into operation at 2000 and uses MA containing MOX-fuel.

Irretrievable actinide losses are taken fixed and equal to 2 per cent, but the results are also presented for a case when this key parameter is improved to 0.2 per cent for Pu and Am.

The resulting data on the radionuclides inventory and their total radiotoxicity at 2060 for the three scenarios considered are presented in Tables II and III. It is obvious that fast reactor operation reduces significantly the volume and toxicity of the actinides going to the waste.

TABLE II. THE MASS OF RADIONUCLIDES ON THE SITE

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Mass at year 2000, kg</th>
<th>Radionuclide mass at year 2060, kg, including the mass gone to waste (in brackets)</th>
<th>Scenario number</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>1</td>
</tr>
<tr>
<td>Np-237</td>
<td>550</td>
<td>720</td>
<td>785(785)</td>
</tr>
<tr>
<td>Am-241</td>
<td>420</td>
<td>2450</td>
<td>2040(2040)</td>
</tr>
<tr>
<td>Am-243</td>
<td>150</td>
<td>150</td>
<td>630(630)</td>
</tr>
<tr>
<td>Cm-244</td>
<td>40</td>
<td>5</td>
<td>25(25)</td>
</tr>
<tr>
<td>Pu-238</td>
<td>165</td>
<td>110</td>
<td>40(20)</td>
</tr>
<tr>
<td>Pu-239</td>
<td>17,600</td>
<td>17,600</td>
<td>17,230(1750)</td>
</tr>
<tr>
<td>Pu-240</td>
<td>4,370</td>
<td>4,400</td>
<td>3,810(840)</td>
</tr>
<tr>
<td>Pu-241</td>
<td>2,350</td>
<td>165</td>
<td>420(90)</td>
</tr>
</tbody>
</table>

*At losses = 2% for Np and Cm, losses = 0.2% for Pu and Am.
TABLE III. TOTAL RADIOTOXICITY INDICES OF THE ACTINIDES AFTER 1000 YEARS STORAGE IN DIFFERENT SCENARIOS (the unity is $10^7$ m$^3$ of water needed to dilute the mass of a nuclide to drinking standards)

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Scenario</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1</td>
</tr>
<tr>
<td>Np</td>
<td>0.03</td>
</tr>
<tr>
<td>Pu</td>
<td>100</td>
</tr>
<tr>
<td>Am</td>
<td>170</td>
</tr>
<tr>
<td>Cm</td>
<td>0.07</td>
</tr>
<tr>
<td>Total</td>
<td>270</td>
</tr>
</tbody>
</table>

* At losses = 2% for Np and Cm, losses = 0.2% for Pu and Am.

So the long term positive ecological impact of transmutation is convincing enough to justify research and development in this direction [6]. Our Institute of Physics and Power Engineering assigns high priority to transmutation programs. The latest of them are listed below.

1. Following nuclear data are measured: fission cross section of odd Cm isotopes (243, 245, 247), Pu-238 and Am-242m; fission neutrons spectra and fission fragments yields for Np-237; capture cross sections for some fission products. The experimental devices are prepared for the measurements of inelastic scattering cross sections and delayed neutrons yields for Np-237. Besides theoretical models of neutron interaction with fission fragments nuclei were developed and tested, complete files of neutron cross sections up to 20 MeV for Am-241 and Am-243 were analyzed and evaluated. Compilation and analysis of data on neutron, proton and gamma-rays interactions with nuclei for the energies up to 1 GeV is in progress and computer formats for the data are being developed. The accuracy of neutron data for minor actinides in reactor energy range must be improved in many cases.

2. Basic instruments are prepared for experiments on critical assemblies, first of all fission chambers with the layers of Np-237, Pu(238-242), Am-241, Am-243, Cm-244, then track detectors (glasses) for the same isotopes. The chambers were calibrated together with French ones in the experiments on the critical assembly "Harmony" (Cadarache) with ultimately hard spectrum. Those chambers and track detectors were used in critical assemblies experiments aimed at improving accuracy of MA neutron cross sections for various reactor spectra.

3. Chamber experiments are accompanied by reactivity perturbation measurements with small specimens of MA (dozens of grams). The aim of the experiments is to improve accuracy of fissioning and capture cross sections by measuring central reactivity coefficients. Up to now the measurements were made for Np-237, Pu-240, Am-241 for a series of assemblies (about 15) with different spectra. The experiments of the same type were done in Japan. Considerable discrepancy for Pu-240 was observed and the reasons are now investigated.

4. Most detailed information including not only fission and capture cross section but inelastic scattering as well will be obtained in macroexperiments on critical assemblies with considerable amounts of Np-237. Besides improvement in neutron cross sections such experiments will provide direct information on real reactor parameters (sodium,...
void coefficients, power density, beta-eff, control rods efficiency, reactivity swing per cycle etc.) both for homogeneous and heterogeneous burner-reactors. From 50 to 100 kg of neptunium dioxide are needed for full scale experiments of this type. The Institute now has about 10 kg allowing to begin the first stage of the experiments on BFS assembly.

5. Along with critical assemblies measurements the experiments with capillary specimens of MA are under way on BN-350 reactor. Their aim is the improvement of cross section accuracy tracing changes of isotope compositions. Specimens of Np-237, Pu-238,240 and Am-241 were already irradiated and are now to be transported to IPPE for analyses. Some are still being irradiated. BN-350 irradiations will enable not only physical but radiochemical investigations as well.

These and other works now under way allow to hope that IPPE experimental base may be successfully used for programs of development of ecologically acceptable nuclear cycle on both national and international levels.

By the authors' view top priority research directions in actinide burning for the period to 2010 are:
- conceptual calculational and analytical investigations aimed at selection and substantiation of requirements for physic-chemical parameters of nuclear fuel cycle back end;
- improvement of traditional (aqueous) methods of SNF radiochemical partitioning aimed at costs optimization and minimization of irretrievable actinides losses in LWR fuel recycling;
- investigations of radically new methods of SNF processing (pyrometalurgical and electrochemical);
- development of the methods of making reactor fuels using secondary raw materials of various compositions (oxide, metallic etc.);
- measurement and evaluation of nuclear, thermal and technological constants of new radioactive materials entering into nuclear fuel cycle on larger scale due to transmutation;
- calculations and experimental research of MA burning in operating reactors and critical assemblies including optimization of core compositions and designs for fast burner-reactors;
- feasibility study for electronuclear transmutation;
- development of comprehensive methods of estimates of radiational hazards connected with geological disposal of radwastes for different sites and different vitrification methods, including the definitions of radiotoxicity indices first of all for the case of geotoxicity.

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Work on Partitioning of Long-Lived Radionuclides in the Russian Federation

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Abstract

The results of research on HLW partitioning performed in Russia are presented. To substantiate the feasibility of HLW partitioning process, the basics of concept on management of long-lived radionuclides are considered. This concept envisages the separation of HLW into individual fractions, which will make it possible in the future to implement, along with geological disposal, the transmutation process as a radical method for management of the most hazardous nuclides. Such approach requires radio ecological substantiation for recovery degree of radionuclides to be transmuted on the basis of the only criterion, i.e. reasonable risk with allowance for natural and artificial barriers to confine radionuclides within a rock massif block. The report summarizes the data on the Russian studies of partitioning of long-lived radionuclides. It includes the description of modified Purex-process which enables to localize Np, Tc and Zr in I extraction cycle. As to reprocessing of high-level raffinate of I cycle, the achieved indices on recovery of Cs, Sr, U, Np, Pu, TPE and RE are under consideration with the use of chlorinated cobalt dicarbollyde, phosphine oxides, crown-ethers, bidentate extractants and ferrocyanide sorbents. It is shown that the testing of some efficient processes for separation of long-lived radionuclides has progressed in Russia to a pilot industrial scale. Further efforts of investigators and technologists are aimed at higher recovery degree of HLW components.

1. INTRODUCTION

In our country a concept of closed fuel cycle is accepted for spent fuel of WWER-reactors which will provide the basis for the development of nuclear power in the next decades. For this purpose an infrastructure of nuclear branch has been established in Russia, including the capacities for spent fuel reprocessing as follows:

- operating plant RT-1 in Chelyabinsk intended for reprocessing of WWER-440 spent fuel;
- plant RT-2 now under construction in Krasnoyarsk for reprocessing of WWER-1000 spent fuel.

The key problem of safety in closed fuel cycle is connected with the management of long-lived radionuclides contained in spent fuel, because the closed cycle enables to use, along with geological disposal, such a radical method for isolation of long-lived radionuclides from the biosphere as transmutation.

Realization of this method requires, in its turn, deep recovery of long-lived radionuclides with subsequent separation into individual fractions. It should be emphasized that the transmutation process could not completely relieve of having to dispose radioactive wastes into geological formations, as any feasible recovery degree would anyway exceed the permissible concentrations required for near-surface storage.

The proposed concept for management of long-lived radionuclides stems from the fact that the portion of radionuclide to be disposed off is determined by the following:

- ecological requirements on the basis of a single criterion, i.e. reasonable risk;
- account for natural factors affecting the retention of radionuclides in rock massif;
- requirements to artificial barriers and matrices of solidified wastes.

Attainment of such recovery degree of radionuclides from HLW is a very complicated technical task which needs up-to-date separation technologies. Starting from the above notions, below are presented the general principles of the concept for management of long-lived radionuclides.
2. CONCEPT FOR MANAGEMENT OF LONG-LIVED RADIONUCLIDES

2.1. Approach to the classification of radioactive wastes, containing long-lived radionuclides according to reasonable risk as a single criterion

The presently accepted classification of radioactive wastes is based on the principles of total $\alpha$- and $\beta$-y-activity. Such classification is convenient for making the decisions on the management of radioactive wastes over short periods of time, but it is unsuitable when it comes to the problem of isolation of long-lived radionuclides from biosphere for extended periods of hundred thousands and more years. In this case of prime importance are the problems of migration of radionuclides in the environment, transfer through biological chains, and formation of doses specific to each radionuclide.

From the above it follows that the methodology for elaborating the classification of radioactive wastes should be oriented to account for effect of individual radionuclides. The evaluation of radiation hazard for each long-lived radionuclide contained in spent fuel is a complicated and not sufficiently advanced problem. To accomplish this task, a series of issues should be resolved:

- analysis of radiation characteristics of each long-lived radionuclide;
- choice of the most ecologically important radionuclides with allowance for potential hazard;
- determination of dose factors for each nuclide as the values of effective equivalent doses per activity unit of nuclide entered the human organism by inhalation or per oral, with regard to physic-chemical forms of nuclide;
- calculation of individual and collective effective equivalent irradiation doses;
- evaluation of possible effects on ecosystems. The resolution of the above issues would make it possible to perform a correct radio ecological substantiation of permissible levels for release of each radionuclide from geological disposal sites into biosphere.

2.2. List of factors determining the confinement of long-lived radionuclides within the limited block of rock massif

Along with the radio ecologically substantiated standards for release of long-lived radionuclides into biosphere, their permissible content in geological disposal sites should be also determined by artificial and natural barriers confining the radionuclides in rock massif.

Among many factors determining the confinement of radionuclides, the deciding ones are as follows:

- form of solidified product, its resistance of underground waters;
- strength of packing and hydroinsulating materials to corrosion attack by underground waters;
- confining ability of hydroinsulating and sealing materials;
- predictable hydrodynamic regime in waste storing zone in rock excavation;
- generation of gas in storage zone;
- mineral-petrographie composition of rocks;
- confining ability of minerals relative to radionuclides;
- mechanism and rate of isotopic and isomorphous exchange reactions between radionuclides and chemical analogues of minerals;
- diffusional intrusion of long-lived radionuclides into mineral block and their retention in crystal lattice of minerals.
With the use of the mentioned and some other factors concerning the confinement of radionuclides in a limited block of rock massif, consideration should be given to scenarios of events and processes resulting in migration of nuclides from their storing sites. Physic-mathematical description of events and processes serves as a basis of elaborating a model for forecast of migration of long-lived radionuclides.

The model would allow evaluation of suitability of proposed rock massif to disposal of certain long-lived isotope on different amounts.

The results of evaluation should:
- determine the quantitative limits for disposal of a particular nuclide into a rock-massif block;
- or evidence the necessity of search for a more suitable geological formation or its block;
- or provide the basis for studying some alternative methods of determining the migration or localization of a certain radionuclide in particular, transmutation).

So, the proposed concept involves differentiated approach to each long-lived radionuclide. Such approach permits to use an alternative method or a combination of methods. In any case the necessary reprocessing stage of wastes containing long-lived radionuclides includes their separation into individual fractions depending on the chosen method for subsequent management of these wastes.

3. PRESENT STATE OF THE ART OF PARTITIONING IN RUSSIA

3.1. Partitioning at the Plant RT-1

During the last 15 years at the Plant RT-1 in Chelyabinsk there are conducted regular tests of HLW partitioning process developed by Radium Institute, Institute of Chemical Technology, Institute of Physical Chemistry, Institute of Geochemistry and Analytical Chemistry. Selective recovery of cesium, strontium, rare-earth and transplutonium elements, residues of uranium, neptunium and plutonium should be considered as key operation for HLLW partitioning. In world practice different techniques for recovery of above HLW components have been proven to a greater or lesser extent, including precipitation, sorption, extraction and chromatographic methods.

One of the promising flowsheets for recovery of cesium, strontium, RE and TPE fractions from HLW has been elaborated at the Radium Institute (St.Petersburg) in collaboration with Institute of Nuclear Research (Prague). This flowsheet is based on using of chlorinated cobalt dicarbollyde (ChCoDiC) in polar diluent as extractant. In the course of semi-industrial trials of the flowsheet at the radiochemical plant, hundreds of cubic meters of HLLW arising from reprocessing of different spent fuels were reprocessed. The technology has afforded the recovery degree of cesium, strontium, TPE and RE in excess of 99%. Semi-industrial operations have made it possible to recover Mega-Curies' quantities of strontium-90 and cesium-137,134 and to produce a concentrate of TPE containing 240 g of americium-241 and 21 g of curium-244. Further efforts are directed towards more efficient recovery of components and management of secondary wastes.

Deep recovery of total actinides from HLLW may be also achieved by means of monodentate neutral organo-phosphorus extractants, and namely different-radical phosphine oxide (POR) - isoamyl dialkylphosphine oxide, the synthesis of which, as well as the synthesis of ChCoD, is performed on commercial scale in our country. This extractant was also tested on real HLW, affording high recovery degree of actinides (up to 99,9%). Solid extractants on POR basis are also very promising. These sorption materials consist of macroporous polystyrol divinyl benzene matrix containing up to 50% mass of extractant.

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Trials of this sorption variant with the use of POR were conducted on an industrial column of 60 l - volume. The obtained results have confirmed not only high recovery degree of actinides, but also efficient concentrating of them. In these experiments the concentration of americium in desorbates was as high as 0.8 g/l. A distinguishing feature of POR-based reactants is the possibility for recovery of technetium from HLW.

In modern world practice there is wide use of TRUEX-process for recovery of actinides from HLW which is based on bifunctional neutral organophosphorus compounds. An interesting modification of TRUEX-process was also tested in test facility of centrifugal extractors with the use of diphenyl dibutyl carbamoyl phosphate oxide in polar organo-fluorine diluent as extractant. This system has provided the recovery of TPE and RE by 99.5% without any solubilizator which is commonly used in TRUEX-systems in the form of tributyl phosphate additions. As in the case of POR, the solid extractants on the basis of bifunctional compounds impregnated into styrol divinyl benzene matrix have been obtained and tested. These solid extractants afford recovery of americium and curium up to 99.95% from HLLW with acidity from 0.5 to 8.0 M.

Extremely high selectivity of macrocyclic polyethers (crownethers) attracted particular interest to their use for recovery of radiostrontium from HLW. In particular, the study of dicyclohexyl-18-crown-6 (DCH-6) was brought to the level of semi-industrial trials. About 90 m³ of HLLW were reprocessed and, as a result, 5.4 . 10⁶ Ci of strontium were recovered with averaged six-fold concentrating. Recovery degree of strontium averaged approximately 96%. The process of strontium recovery by DCH-6 seems to be attractive due to using distilled water as reextracting solution. This crown-ether ranks below such extractants as ChCoDiC in versatility, however at present the Russian authors of this technology are pursuing investigations into the systems on the basis of crown-ethers which have the capability to recover selectively not only strontium, but also cesium. As an example for successful use of sorption processes in HLW partitioning, we refer to methods for selective sorption of cesium by means of inorganic ferrocyanide-containing sorbents (zinc-potassium, nickel-potassium and copper-potassium forms) which are produced on, pilot-industrial scale. The development of this sorption method for cesium recovery was also brought up to the level of semi-industrial trials. For this purpose a column with a volume of 120 litres was used. The recovery degree of cesium at sorption stage exceeded 98%, the degree of cesium concentrating attained 100. In the course of 10-15 cycles of sorption-desorption-regeneration processes no changes in sorption and operating characteristics of FS-10 sorbent (copper-potassium sorbent) were observed. Mega-curies’ quantities of cesium-137 were produced.

To summarize briefly the present state of work on partitioning of long-lived radionuclides at the Plant RT-1, it should be emphasized that some technologies developed at the Russian institutes have been already now brought to the level of semi-industrial trials. Further elaborations of HLW partitioning at the Plant RT-1 are connected with more efficient recovery of long-lived radionuclides in accordance with their radio-ecological hazard. Work on increased degree of nuclides’ recovery is underway along with the development of radio-ecological criteria limiting the amounts and concentrations of radionuclides to be disposed off into geological formations.

3.2. Partitioning in the flowsheet of the Plant RT-2, now under construction

The advanced level of technologies for partitioning of long-lived radionuclides which have been developed by the Russian institute and brought up to the stage of semi-industrial trials at radiochemical plant makes it possible to include the partitioning concept into the flowsheet of the new large reprocessing plant now under construction in Krasnoyarsk (Plant RT-2).
Initial operations of partitioning are involved in the RT-2 flowsheet within the framework of WWER-1000 spent fuel reprocessing scheme, thereby changing this reprocessing into a modification of traditional Purex-process.

The recovery of some long-lived radionuclides begins with the operation of fuel dissolution when carbon-14, iodine-129 and krypton-85 pass into gas phase. For localization of carbon-14 and iodine-129, a process of their absorption in columns sprayed with alkaline solution is developed.

Taking into account the global hazard of krypton-85 discharge into atmosphere, provision is made for off-gas cleaning from this element. With this aim, the methods are now tested for krypton adsorption on activated coal, absorption by freons and cryogenic distillation.

Further modification of Purex-process is aimed at localization of neptunium, technetium and zirconium within the framework of extraction reprocessing of spent fuel solutions. In the case of neptunium, the recovery degree attains now over 95%; further investigations should increase this value over 99%.

Recent advances in technetium chemistry have shown the feasibility of control for its behaviour within the I reprocessing cycle. One of such control techniques is based on quantitative extraction of technetium into combined reextract of uranium and plutonium with subsequent withdrawal of technetium into raffinate at the stage of plutonium purification. After sorption processing of this product the irretrievable losses of technetium do not exceed 0.5%.

Further development of HLW partitioning process results in a method for localization of zirconium also on the I extraction cycle. By adjustment of acidity in extraction cascade for recovery of uranium and plutonium, it has been confirmed that more than 95% of zirconium could be extracted into an individual product.

Processing of the I cycle raffinates is preceded by an operation for their concentrating on evaporation. To avoid precipitation during this operation, some variants of preliminary recovery of molybdenum from raffinate have been tested. The recovery degree was as high as 99%.

Next operations for HLW partitioning involve processing of evaporated raffinates with the aim of selective recovery of cesium, strontium, residues of uranium, neptunium, plutonium, as well as RE and TPE. Raffinate partitioning is based on the processes developed at the Radium Institute and tested at Chelyabinsk Radiochemical Plant. These processes described in above section in combination with modified Purex-process form technology basis for the new reprocessing plant RT-2 in Krasnoyarsk.

4. CONCLUSION

The concept of closed fuel cycle accepted in Russia for spent fuel of basic WWER-1000 reactors envisages, along with geological disposal of long-lived radionuclides, the possibility of burning these nuclides by means of transmutation.

The use of transmutation does not relieve of having to dispose a non-recovered portion of radionuclides into geological formations. For determining the permissible non-recovered portion of each long-lived nuclides, the radiologic requirements on the basis of reasonable risk criterion are being elaborated. Besides, to assess a rock massif block, the natural factors affecting the confinement of radionuclides are under study, and the essential features required of man-made barriers and matrices are defined.
To obtain such recovery degree of radionuclides from HLW, the separation technologies are now under development in Russia. On spent fuel reprocessing the initial operations of partitioning are involved within the so-called modified Purex-process which provides the localization of iodine, neptunium, technetium and zirconium.

For HLLW partitioning (I cycle raffinates) there are elaborated in Russia extraction and sorption technologies which afford the selective recovery of cesium, strontium, RE, TPE and residues of uranium, neptunium and plutonium. One of the combined flowsheets on the basis of ChCoDiC and POR has been tested on pilot-industrial scale at Chelyabinsk RT-1 plant.

In combination with modified Purex-process, this flowsheet forms the basis of technology for the new reprocessing plant now under construction in Krasnoyarsk. This technology will offer in the future safe management of long-lived radionuclides as a compromise between disposal into deep geological formations and transmutation.

REFERENCES

Abstract

The UK has no major programme on partitioning and transmutation (P&T) of long-lived fission products but has a commitment to keep this, and other, waste management options, under review. The benefits of P&T in terms of reduction of the radiation doses to mankind arising from a waste repository will be vanishingly small. In contrast, the costs are likely to be considerable. A comparative study has been begun of the performance of alternative partitioning schemes described in the literature and their overall plant requirements. An important aspect which will be considered is the arisings and treatment of secondary wastes, both from the conventional PUREX process and from partitioning processes themselves.

Introduction

The United Kingdom view on the concept of partitioning and transmutation (P&T) of actinides and long-lived radionuclides has been presented at a previous IAEA meeting\(^{(1)}\). Based on earlier international studies which are still considered valid, the UK Nuclear Utilities Chairmen's Group summed up their views in the report\(^{(2)}\) of their Topic Group on Decommissioning and Waste Management in 1992:

"International reviews of the potential of PTA\(^*\) came to the conclusion that there will be no significant reduction in potential long-term radioactive risk. These are dominated by isotopes unaffected by PTA and the wastes would therefore still require geological disposal. It was also concluded that the resources needed for the work would be extremely large."

In accordance with this view, there is no major programme of research and development on P&T in the UK. There is, however, a commitment to evaluate long term waste management options including P/T and to keep the topic under review.

Benefits of Waste Partitioning

Any assessment of the value of a waste management option such as P/T must be based on an evaluation of the benefits to mankind that are expected to be achieved and of the costs of achieving these benefits.

The benefits of P/T must be seen primarily in terms of potential reduction in the exposure of mankind to radiotoxic material. Any savings in the cost of final disposal are likely to be minor since the difficulties of complete separation will tend to mean there will still be a need for a geological repository of some kind. The main benefit that has been argued is that there will be a reduction in the overall toxicity of the waste that has to be consigned to a repository.

However, reduction of the toxicity of HLW confined in a repository may not in itself result in any reduction in the radiological dose to mankind. This dose is determined rather by the radionuclides that are released from the repository into the biosphere. Such releases are determined by the

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*PTA - partitioning and transmutation of actinides*
solubility of radionuclides in ground-water under repository conditions and by the rate at which they are able to migrate through the back-fill and the geological strata. The higher actinides, because of well known features of their chemistry, have low solubilities under repository conditions and are also efficiently absorbed by many geological media. Other more mobile isotopes dominate the dose pattern.

It has been shown\(^3\) that doses to mankind arising from releases from HLW expected to arise in the UK by the year 2030, in a repository, are very low. An annual effective individual dose of about $10^{-4}\text{mSv}$ (0.01% of the natural background) will occur between $10^5$ and $2 \times 10^6$ years after disposal, owing to release of technetium-99. Subsequently (after $10^7$ years) there will be a smaller dose from release of caesium-135. These doses are in fact lower than those resulting from disposal of UK ILW arisings over the same period. This is because ILW contains chlorine-36 and iodine-129 which lead to a dose of about $0.01\text{mSv}$ after $10^5$ years. After $2 \times 10^6$ years, the main isotope responsible for dose rates in the biosphere from ILW is radium-226 (a daughter of uranium-238).

In the light of these results, any benefit arising from the separation of radionuclides from high-level waste (HLW) before disposal is likely to be very small. Moreover greater reductions in doses to mankind will be obtained from the removal of technetium-99 and caesium-135 than from that of the minor actinides. It could also be argued that it would be more beneficial to focus attention on the chlorine, iodine and uranium content of ILW, though this would clearly be a very difficult technological problem and the benefits would still be very small.

\textbf{Costs and Technical Performance of Partition Processes}

Against the putative benefits of P/T schemes in comparison with direct disposal of HLW, there must be set the additional costs of the P/T fuel cycle over those of a reference reprocessing fuel cycle. Such cost considerations need to be made in the context of an overall systems assessment. The following steps and their costs must be considered:

1. partition of HLW to yield product streams containing the radionuclides to be separated;
2. conversion of the product streams (often aqueous solutions) into compounds (such as oxides) suitable for fabrication into fuel elements or targets for irradiation;
3. fabrication into fuel elements or targets;
4. irradiation in reactors or accelerators to bring about transmutation of long-lived radionuclides into shorter-lived products;
5. removal and further reprocessing to recover material that has not been transmuted, if required by the characteristics of the irradiation target or fuel, followed by refabrication and irradiation.

All these steps (particularly 1, 2, 3 and 5) will generate secondary waste streams, containing the nuclides being separated for transmutation. Such wastes will either require separate treatment and disposal, or will have to be recycled back into the process after suitable treatment. The degree to which such secondary wastes are generated, and the completeness of the separation of the desired nuclides, are therefore important aspects of the performance of P/T schemes, which need to be addressed in a systems study.

Another aspect that needs to be included is the dosage to process operators that occur in steps 1 to 5. These doses should be set against reductions in the dose to the wider population from material released from a repository in the distant future.

The objective of desk studies being undertaken in AEA Technology is to move towards such a systems study. Most attention is being focused on the performance of processes for HLW partition
and the secondary wastes that arise from them. It is hoped to participate in the CEC R&D programme in this area.

The UK study is in its early stages. A preliminary analysis has been prepared of the relative plant requirements and separation performances of three candidate HLW partition processes. It is planned to extend this study to include other candidate processes, and also to identify and quantify the secondary waste streams that arise from the partition processes, and methods for their treatment.

Secondary Wastes

Table 1 lists the major waste streams that arise from a typical PUREX-type reprocessing flowsheet. The major waste stream that might be passed to a partitioning process is of course the high-level waste stream containing most of the fission products and minor actinides. However other waste streams (see Table) can contain substantial amounts of long-lived nuclides that are the subject of P/T strategies. The first step in a study of secondary wastes is to consider how these other streams should be dealt with.

From the table, two important waste streams can be identified that contain both actinides and fission products viz the cycle 1 solvent wash aqueous raffinate (CISWAR) and the analytical wastes from cycle 1. Because of the spectrum of radionuclides these wastes could perhaps be treated (after evaporation) by the same processes as are envisaged for HLW partition. However, if the usual alkaline solvent wash treatment is used, the CISWAR will contain sodium nitrate. Analytical residues (which can be a significant waste stream) can contain various reagents such as fluoride or EDTA. These contaminants could create difficulties in processing.

A further group of wastes (C2(Pu)AR, C2(Pu)SWAR, Pu evaporator overheads, C2(Pu) analytical wastes and raffinates from MOX finishing and fabrication plants) can be identified in which plutonium is the principle contaminant. Some of these will contain low concentrations only. The cycle 2 (Pu) aqueous raffinate will be capable of evaporation, and the resulting high nitrate Pu solution could be decontaminated by ion exchange. However, there would then be a requirement to manage the loaded ion exchanger. Alternatively, the concentrate might be recycled.

Several other waste streams may need to be considered. Those in which uranium is the main contaminant may be relatively easy to deal with. However, the management of waste solvent, dissolver insolubles and other solid wastes containing actinides, may present difficulties in a complete P/T strategy.

Wastes from HLW partition processes

Work on quantifying secondary wastes from suggested partition processes is at an early stage. It is planned to make as detailed solvent extraction calculations as possible, using published flowsheets and distribution data. Estimates will also be made of solid waste arisings.
## TABLE 1. SECONDARY WASTE ARISINGS FROM THE PUREX PROCESS

<table>
<thead>
<tr>
<th>Stream</th>
<th>U</th>
<th>Pu</th>
<th>Am / Cm</th>
<th>Np</th>
<th>(^{99})Tc</th>
<th>(^{109})Pd</th>
<th>(^{125})I</th>
<th>(^{90})Se</th>
<th>(^{91})Zr</th>
<th>(^{126})Sn</th>
<th>(^{135})Cs</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dissolver Scrubber</td>
<td>*</td>
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<td>HLLW</td>
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<td>C1 SWAR</td>
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<td>Pu Evap Overheads</td>
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<td>U Evap Overheads</td>
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<td>Aq Raff ex UO(_2) **</td>
<td>*</td>
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<tr>
<td>FP Insolubles</td>
<td>*</td>
<td>*</td>
<td>?</td>
<td>?</td>
<td>*</td>
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<td>*</td>
<td>?</td>
<td>*</td>
<td>?</td>
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<tr>
<td>Waste Solvent</td>
<td>*</td>
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<td>*</td>
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<tr>
<td>Analytical Wastes</td>
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<td>ex C1</td>
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</tr>
</tbody>
</table>

**Notes:**  
* - significant concentrations, + - low concentrations, ? - possible contamination  
AR - aqueous raffinate, SWAR - solvent wash aqueous raffinate, FP - fission product  
** - wastes from mixed oxide or uranium finishing processes
References


STRATEGY STUDY ON PARTITIONING AND TRANSMUTATION OF NUCLEAR WASTE

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Abstract

Partitioning and transmutation (P&T) of long-lived radionuclides is studied by some countries as an alternative option for radioactive waste management. To assess the benefits of this option, the Commission of the European Communities has placed research contracts on a strategy study of P&T with three European research institutions at the end of 1991 for a 2 year duration. CEA in France is investigating the potentialities and costs of P&T of long-lived radionuclides. Siemens in Germany is analysing the possibilities of advanced converters for transmutation and their safety behaviour. ECN Petten in The Netherlands is preparing and assessing nuclear data libraries for transmutation studies. The objective of the study and the main results obtained at the end of 1993 are presented for each of the three research contracts. Some of the results presented are still preliminary, as the contracts have been extended until the end of 1994 with an additional working programme.

POTENTIALITIES AND COSTS OF PARTITION AND TRANSMUTATION OF LONG-LIVED RADIONUCLIDES

1. Objectives

This study is carried out by CEA in France. 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 scenarios without and with conventional reprocessing and scenarios using P&T are compared to assess the potentialities of P&T.

Radiotoxicity has been chosen as a measure of the potential detriment of the waste resulting from the different scenarios analysed. This approach does not take into account the various barriers, natural and engineered, which prevent or delay the movement 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.

2. Reference scenario

The three reference scenarios 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.

The potential radiotoxocities are assessed for ingestion and mainly deal with all heavy radionuclides and three long-lived fission products (\(^{99m}\)Tc, \(^{129}\)I, \(^{135}\)Cs). They are calculated for
cooling times between 10 and 10⁷ 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).

Figure 1 shows the time evolution of the potential radiotoxicity computed for the "waste" inventory in the case of the R2 and R3 scenarios normalised with respect to R1. The radiotoxicity is decreased more for the R3 scenario than for the R2 scenario. The reduction factor is larger than 5 for the R2 scenario between 10⁴ and 10⁵ years. In the short term (10⁻¹⁰⁻¹⁰⁵ years), the radiotoxicity is mainly due to ²⁴⁴Cm and ²⁴¹Am. In the long term (10⁵⁻¹⁰⁵ years), there is a paramount contribution of ²⁴³Am, its daughter ²³⁹Pu and ²⁴⁰Pu. In the very long term (>10⁵ years), the radiotoxicity is dominated by ²³⁷Np.

The time variation of the potential radiotoxicity of the "cycle" inventory is presented in Fig. 2. It has been computed in the case of the R2 and R3 scenarios normalised with respect to R1. In the short and long terms, the radiotoxicity is dominated by the same radioisotopes as in the "waste" inventory. In the very long term, it is mainly due to ²³⁷Np and ²⁴²Pu. Beyond 5.10⁵ years, the contribution of uranium and its daughters becomes increasingly more important.

3. RP1 scenario

Two scenarios are considered for partitioning, RP1 with available technologies and RP2 with futuristic technologies. As far as the scenario RP1 is concerned, the transmutation of (minor and major) actinides starts from 2010 in PWRs (scenario RP1-1) or in FRs after 2020 (scenario RP1-2).

At present, the study of the RP1-1 scenario is completed and the main results obtained are summarised in this paper. The data available for the RP1-2 scenario are given also. As work is still in progress for the RP2 scenario, nothing will be reported here concerning this topic.

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*FIG. 1. Radiotoxicity (ingestion) – waste inventory evolution from 2100.*
The general diagram of the RP1 process for partitioning is shown in Fig. 3. The different steps of this process are the following:

- Np is extracted with U by a slight modification of the PUREX process (oxidation of Np[V] into Np[VI] by the vanadile ion VO\(^{2+}\));
- the solution containing the fission products and the actinides is treated by a diamide solvent to co-extract Am, Cm, the rare earths and the traces of Pu, U and Np; Am, Cm and the rare earths are then back-extracted with nitric acid;
- the slightly acidic solution containing Am, Cm and the rare earths is treated by a mixture of DIDPA and TBP; the partitioning between actinides and rare earths is carried out by a process such as "reverse TALSPEAK": Am and Cm are back-extracted from the solvent by a DTPA aqueous solution, while the rare earths stay in the solvent;
- partitioning between Am and Cm is done by selective extraction of Am[VI] by HD(DiBM)P or DIDPA in centrifugal extractors, after electrochemical oxidation of Am[III] into Am[VI]; Am is recovered as americium oxide and Cm is sent to vitrification, since it is not envisaged to transmute Cm in the RP1 scenario.

The partitioning of neptunium and americium starts in 2010 with an efficiency of 95%.

The inventory of the RP1 scenario shows that the plutonium flow is four times larger than in the reprocessing plant UP3 in La Hague. This is due to Pu multi-recycling in MOX reprocessing. The relative average concentration of \(^{239}\)Pu is about 8.4%, which is larger than the generally accepted limit of 5%, above which \(\alpha\)-radiation may degrade the solvent.

3a. Incineration in PWRs (RP1-1 scenario)

This scenario is compared to the R2 scenario. In the RP1-1 scenario, neptunium and americium are incinerated in PWRs either in homogeneous or in heterogeneous mode.
In homogeneous mode, a quantity of neptunium oxide or americium oxide representing 1% of the total mass of heavy isotopes is mixed to the UOX fuel. 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, specific targets are placed in guide tubes of a UOX assembly; the pins are made of either 100% neptunium oxide or 30% americium oxide with 70% alumina. These targets are irradiated during 5 years and are not reprocessed.

Physical properties of the fuel

The consequences of americium and neptunium recycling in PWRs in homogeneous mode on the physical properties of the fuel have been computed using the neutron code APOLLO 1 and the cycle code KAFKA. A larger enrichment of the fuel in $^{235}\text{U}$ (about 5% instead of 4%) is required to keep the same average burn-up of 47.5 GWd/t of metal. When recycling Np, the resulting spent fuel has 1.5 more activity, a slightly smaller neutron source and about twice more residual power than the standard UOX spent fuel after 5 year cooling time. This is due to the larger production of $^{238}\text{Pu}$, which is an $\alpha$-emitter. In the case of Am recycling, the activity is increased by a factor of about 2, the neutron source by a factor of about 30 and the residual power by a factor of around 3 compared to those of standard UOX spent fuel after 5 year cooling time. This is caused by the formation of $^{244}\text{Cm}$, which is a neutron and $\alpha$-emitter.

Radionuclide inventory

The strategy code COSI allows to compute the radionuclide and radiotoxicity inventories. It simulates the working of a reactor park and the associated fuel cycle facilities.
In homogeneous mode, the fraction of the reactor park necessary to recycle Np in PWRs varies from 5% at the beginning of the RP1 scenario to 9%, when the park has reached a steady state. Correspondingly, 9 to 33% of the park is necessary to burn Am in PWRs.

In heterogeneous mode, 5% of the reactor park is enough to incinerate Np, but the park is not sufficient to recycle all Am coming from reprocessing of UOX and MOX fuel.

Burning Np and Am in PWRs in homogeneous mode results in the production of about 8% more depleted uranium for the homogeneous RP1-1 scenario than for the R2 scenario in 2100. This increase is due to the larger enrichment of the fuel in $^{235}$U required by the presence of Np or Am.

In 2100, 130 tons of Np and 370 tons of Am have been recycled in homogeneous mode. These figures are 90 tons and 215 tons respectively in heterogeneous mode. The differences between the two modes of recycling are due to the facts that in heterogeneous mode the targets are not reprocessed and Am is not completely recycled under the present assumptions (in 2100, there is about 125 tons of Am in storage, which represents about 30 % of the recycled Am).

The stock of irradiated targets in heterogeneous mode reaches 300 tons in 2100, 30% of them being Np targets and 70 % Am targets.

The cumulated actinide losses coming from reprocessing until 2100 are compared in Table I for the R2, homogeneous RP1-1 and heterogeneous RP1-1 scenarii. These losses will be disposed of. There is also 125 tons of Am in storage in the heterogeneous RP1-1 scenario. The larger production of Cm in the homogeneous case is due to Am recycling. In the heterogeneous mode, the amount of Cm produced reaches the same level, when taking into account the Cm contained in the irradiated Am targets also.

Potential radiotoxicity inventory

Figures 4 and 5 show the time evolution of the potential radiotoxicity computed from 2100 for the homogeneous RP1-1, heterogeneous RP1-1 and R2 scenarii normalised with respect to the R1 scenario for the "waste" and "cycle" inventories respectively (see Section 2).

<table>
<thead>
<tr>
<th>scenario</th>
<th>actinide</th>
<th>R2</th>
<th>RP1-1 homogeneous</th>
<th>RP1-2 heterogeneous</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>U</td>
<td>470</td>
<td>470</td>
<td>470</td>
</tr>
<tr>
<td></td>
<td>Pu</td>
<td>22</td>
<td>22</td>
<td>22</td>
</tr>
<tr>
<td></td>
<td>Np</td>
<td>100</td>
<td>12</td>
<td>10</td>
</tr>
<tr>
<td></td>
<td>Am</td>
<td>420</td>
<td>27</td>
<td>25</td>
</tr>
<tr>
<td></td>
<td>Cm</td>
<td>65</td>
<td>108</td>
<td>51</td>
</tr>
</tbody>
</table>

TABLE I. CUMULATED ACTINIDE LOSSES (IN TONS) FROM REPROCESSING IN 2100 FOR DIFFERENT SCENARIO. THESE LOSSES ARE INTENDED FOR DISPOSAL

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FIG. 4. Radiotoxicity (ingestion) – waste inventory evolution from 2100.

FIG. 5. Radiotoxicity (ingestion) – cycle inventory evolution from 2100.
Concerning the "waste" inventory, the reductions in radiotoxicity with respect to the R2 scenario are nearly the same for the two RP1-1 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 $5 \times 10^5$ and $5 \times 10^6$ year cooling time due to the smaller content in Np. For the other time periods, the gains with respect to the R2 scenario are offset by the increased production of Cm.

In the "cycle" inventory, the part due to the "waste" inventory represents less than 20% of the total radiotoxicity beyond 100 year cooling time. The main part of the radiotoxicity comes from the irradiated fuels and targets (in storages or in reactors) up to $10^6$ years and from depleted uranium beyond. The gains with respect to the R2 scenario are not significant; the radiotoxicity is even increased up to 100 year cooling time because of the production of Cm. The storage of irradiated targets degrades the heterogeneous mode scenario with respect to the homogeneous one.

Cost assessment

Up to now, the progress of this study enables to compare the global cost of the homogeneous RP1-1 scenario to that of the R2 scenario. The costs are assumed to be the same for both scenarii for the following fuel cycle operations: uranium ore extraction, conversion, uranium enrichment and reactor. However, the processing and actinide fuel fabrication costs differ.

The reprocessing costs for the RP1-1 scenario include the costs of investment for the construction of the reprocessing plant, operation, up date and dismantling. The construction costs are assessed from the diagram for the process design (see Fig. 3), the material inventory to fix the flow for each reprocessing workshop and the reference cost of the present workshops. The workshop costs are calculated by using three scaling formulae, which take into account the number of additional lines, the variation of the workshop capacity and possible modifications to the workshop. These formulae have been derived from the past experience of CEA. Finally, the overall reprocessing cost for the RP1-1 scenario is assessed to be between 20 and 50% larger than for the R2 scenario with the assumptions taken for the definition of both scenarii.

Adding Np and Am to UOX in a fuel fabrication plant implies that the radioprotection constraints will be larger than in a MOX fuel plant. These constraints can be dealt with by additional shielding and automation. The fabrication costs of Np and Am fuel for the homogeneous RP1-1 scenario are estimated to be between 10 and 30% larger than those of MOX fuel. It would then be recommended to recycle Np and Am in MOX rather than in UOX fuel. As there is no industrial experience in manufacturing Np and Am targets for the heterogeneous mode, the costs have not been assessed.

In conclusion, the global cost of the homogeneous RP1-1 scenario is assessed to be between 20 and 45% larger than that of the R2 scenario. In the R2 scenario, the UOX and MOX fuels are burnt in PWRs and reprocessed with the PUREX process. In the RP1-1 scenario, partitioning is implemented with available technologies and the actinides except Cm are incinerated in PWRs.

3b. Incineration in FRs (RP1-2 scenario)

As the study is not completed yet, only the basic assumptions of this scenario and the results obtained for the physical properties of the fuel are reported here. 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 the homogeneous mode, an amount of Np or Am representing 2.5% of the total mass of heavy isotopes is mixed with the FR fuel. The fuel is reprocessed as the standard FR fuel. The actinide losses during reprocessing are the same as in the RP1-1 scenario.

In the heterogeneous mode, neptunium and americium oxide targets coated with alumina are placed at the first radial breeding row of the core. These targets contain 40% of Np or 20% of Am of the metal mass. They are irradiated during 15 years (3 cycles) and are not reprocessed.

Physical properties of the fuel

The consequences of Np and Am recycling in FRs on the physical properties of the fuel are only investigated in the homogeneous mode. The same maximum burn-up of 150 GWd/t of oxide can be maintained by mixing Np or Am up to 3% of the total mass of heavy metals in a FR fuel without altering the initial amount of plutonium significantly. After 5 year cooling time and compared to the standard FR spent fuel, the minor actinide spent fuel has roughly the same activity, a neutron source and a residual power increased by a factor of about 2. This is linked to the larger production of the $\alpha$-emitter $^{238}$Pu from Np and of the neutron and $\alpha$-emitter $^{244}$Cm from Am.

TRANSMUTATION OF LONG-LIVED RADIONUCLIDES BY ADVANCED CONVERTERS

1. Objectives

Siemens in Germany is analysing the possibilities of advanced fast reactors (FR) and accelerator driven spallation devices for transmutation of actinides and long-lived fission products.

The reference fast reactor is EFR with a thermal power output of 3600MWt, MOX fuel and a 1m core height. The possibility of enhancing transmutation by neutron spectrum hardening via leakage increase (smaller core height, smaller units) will be studied. Metallic and oxide fuel and homogeneous or heterogeneous insertion of waste will be compared. The safety behaviour of the different cores will be evaluated by the sodium void effect and the Doppler effect.

Concerning spallation devices, different fast and thermal systems will be compared.

2. Results

2a. Transmutation in fast reactors

The main results of the study obtained so far can be summarised as follows:

- Reducing the core size might be a possibility to introduce minor actinides (MA) into FR cores without exceeding the sodium void effect level of the European Fast Reactor (EFR); small cores have a smaller MA transmutation rate than large ones because of their lower flux level.

- The maximum fraction of 5% of $^{238}$Pu of the total Pu assumed at present for reprocessing would impose an upper limit of 3% for the MA content in the fresh fuel.

- 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 core and a reduced height (0.7m). Such a core would be able to take 880 kg of MA and to transmute 65 kg of MA per year, which represents the production of about three PWRs.
The capability to burn Pu is larger in smaller cores because of their poorer internal breeding properties. However, when the cores are especially optimised for Pu consumption, there are no differences between small and large cores due to the maximum Pu enrichment limit for the small cores.

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 $^{239}$Pu build-up of 5% of the total Pu. 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.

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 1.2 m core height with 84 $^{99}$Tc target subassemblies and a thermal power output of $2600\text{MW}_{th}$, about 166 kg 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.

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

2b. Accelerator driven transmutation devices

A literature review has been made on accelerator driven spallation devices for transmutation. Four of these projects have been appraised:

- The PHOENIX concept of the Brookhaven National Laboratory is mainly based on proven technology with some extensions and uses transmutation in a fast neutron spectrum.

- The project for Accelerator Transmutation of Nuclear Waste (ATW) of the Los Alamos National Laboratory is a very advanced device with CANDU/slurry fuel technology and makes use of a high thermal neutron flux.

- Los Alamos has now abandoned the above concept for an ambitious hybrid transmuter, which should burn waste on site during the first 30 years and produce energy by burning thorium after. This system is essentially a subcritical thorium burner with the PWR actinide waste serving as a fissile starter.

- The project of the JAERI transmutation plant is more realistic, since it relies more on existing and proven technologies than the first three devices.
NUCLEAR DATA LIBRARIES FOR TRANSMUTATION STUDIES

1. Objectives

The four main aims of the work carried out by ECN Petten in the Netherlands 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 scenarios 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.

2. Main achievements

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.

Two problems have still to be solved:

(i) the cross sections for high threshold reactions like (n, 2n) only depend on the neutron energy above 1 MeV, but ORIGEN-S has no parameter to describe the hardness of the fast spectrum;

(ii) it would be useful to improve the determination of the energy dependence of the branching ratios used in the EAF-3 library. At present, there is a large uncertainty in the branching ratios of some nuclides in the ORIGEN-S library, which leads to large discrepancies in the computation of the density of these nuclides.

Three types of transmutation devices are being compared: an accelerator alone, a reactor alone and an accelerator with a subcritical reactor. The most realistic one in the short term is a reactor alone, economically and technically speaking. Accelerator driven systems for the transmutation of fission products suffer from their lower energy efficiency and their poor cost effectiveness.

CONCLUSION AND SUMMARY

The study of a scenario in which partitioning is implemented with available technologies and the actinides except curium are incinerated in PWRs (RP1-1 scenario) is completed. It
is compared to a reference scenario where UOX and MOX fuels are burnt in PWRs and reprocessed by the PUREX process (R2 scenario). For the RP1-1 scenario, there is a reduction by a factor of 6 in the radiotoxicity of the waste to be disposed of with respect to the R2 scenario between 102 and 103 and between 5.105 and 5.106 year cooling time. However, there is no gain between both scenarios, when considering the total amount of nuclear materials present in the fuel cycle, because of the irradiated fuels and targets in storages and in reactors. Finally, the overall cost for the RP1-1 scenario with homogeneous fuel is evaluated to be between 20 and 45% larger than for the R2 scenario.

Work is progressing on the RP1-2 (incineration in FRs) and on the RP2 (P&T with futuristic technologies) scenarios.

A fast reactor with a large core and a reduced height (0.7m) would be able to transmute around 65kg of minor actinides (MA) per year, which represents the production of about three PWRs, without compromising its safety behaviour. The MA transmutation half-life is 11 years. Concerning long lived fission products, about 166kg of 99Tc corresponding to the production of 5 to 6 1GWe PWRs could be transmuted per year in a FR having a core with an increased height (1.2 m) and a thermal power output of 2600 MWth. The 99Tc transmutation half-life is around 26 years. The safety parameters of such a device are in line with conventional FRs.

PWR and FR cross section data bases have been updated with data from the JEF2-2 and EAF-3 libraries. Cross sections at average burn-up are used to take account of their variation with burn-up. This work has pointed out two deficiencies in the ORIGEN-S fuel depletion code concerning on one hand the treatment of the energy dependence of the (n,2n) reaction cross sections and on the other hand the large uncertainty in the branching ratios of some nuclides.

ACKNOWLEDGEMENTS

The author acknowledges with thanks helpful comments from Drs. K. Abrahams (ECN Petten), W. Balz (CEC), H. Boussier (CEA), N. Cadelli (CEC), J.P. Grouiller (CEA), U.K. Wehmann (Siemens) and A. Zaetta (CEA).
COMPARISON OF POSSIBLE PARTITIONING AND TRANSMUTATION SCHEMES WHEN ADDED TO THE EXISTING NUCLEAR FUEL CYCLE

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Abstract

There have been several concepts proposed for partitioning and transmutation; those relying on existing reactors are by far more advanced. For this purpose, fuels are already under development which contain minor actinides and $^{99}\text{Th}$. They are being tested under irradiation in fast and thermal neutron fluxes. The status will be described. The partitioning of long-lived radionuclides from the present fuel cycles is being developed and flow sheets are under investigation. A comparison of the present to the expected performance will be made. So far, accumulated experience allows for a comparison of partitioning and transmutation schemes with the present conventional reprocessing and the direct storage of spent fuels. An assessment of the possible reduction of the radiological risk - after the release of radiotoxic nuclides from a repository - is given.

INTRODUCTION

Research for partitioning and transmutation mainly focused on the transmutation processes and was giving less attention to the partitioning of radiotoxic nuclides from the waste streams. The development of fuels or targets, which should withstand the extreme conditions of a transmutation process and, moreover, could be reprocessed, has been almost disregarded. The often proposed "once-through transmutation" leading to an acceptable reduction of the radiotoxic inventory is impossible, because any structural material containing the radionuclides during the process would also be transmuted and rupture.

COMPARISON OF THE NUCLEAR FUEL CYCLES

The ongoing discussion on the topic of partitioning and transmutation brought forward a variety of proposals which are summarised in figure 1. Processes based on photon-induced reactions or spallation of minor actinides by charged particles are in a stage too early to justify any target development except those for basic experiments. Other processes, as certain accelerator-driven reactor concepts and the recently revived proposal of transmutation in fusion reactors, are based on similar fuel or target concepts as those proposed for existing reactors.

In the following, fuel cycles in existing nuclear power stations are compared in view of their capabilities to reduce the radiotoxic nuclides in their discharged waste streams. The equilibrium concentrations for the nuclides of Np, Pu, Am, Cm and Tc are given for self-generated nuclide recycling. In order to compare the four concepts, we consider the fuel cycle as a closed system by which energy and radiotoxicity are generated and released, i.e. in the case of the once-through cycle, all nuclides contained in the fuel, but in the other examples only the losses during the remake of the fuel, respectively. Hence, the masses of each discharged nuclide are normalised to the associated energy output of the fuel in table 1.
### Table 1  Masses of selected radionuclides per MWd discharged from different fuel cycles.

<table>
<thead>
<tr>
<th>Fuel cycle</th>
<th>(\mathrm{(^{235}UO)})</th>
<th>(\mathrm{^{239,240}PuO)})</th>
<th>MWd/t</th>
<th>mg/MWd</th>
</tr>
</thead>
<tbody>
<tr>
<td>PWR 1300 MWe &quot;once through&quot;</td>
<td>4</td>
<td>–</td>
<td>50.000</td>
<td>50.000</td>
</tr>
<tr>
<td>PWR 1300 MWe Pu-recycle selfgenerated after 5 cycles</td>
<td>3.75</td>
<td>1</td>
<td>50.000</td>
<td>136.00</td>
</tr>
<tr>
<td>PWR 1300 MWe Pu+MA+ (^{99m}Tc) recycle selfgenerated after 5 cycles</td>
<td>3.75</td>
<td>1</td>
<td>50.000</td>
<td>145.00</td>
</tr>
<tr>
<td>FR 1200 MWe Pu+MA recycle selfgenerated after 16 cycles</td>
<td>–</td>
<td>10</td>
<td>80.000</td>
<td>825.00</td>
</tr>
</tbody>
</table>

#### PWR "once-through" fuel cycle (as a reference case)

Based on the design of the Biblis PWR, the content of Pu and Tc nuclides has been calculated in the spent fuel after a burnup of 50,000 MWd/t and an out-of-pile-time of 7 years [2]. The enrichment of \(^{235}\)U was 4\% in weight (table 1). The discharged concentration after 7a out-of-pile has been normalised to the energy output.

#### PWR self-generated Pu recycle

For the same reactor and operation conditions as above, the discharge concentration after 7a out-of-pile has been calculated in the case of recycling of self-generated plutonium. The data refer to an
"equilibrium" after 5 cycles [2]. It should be noted that at this stage 75% of the discharged fuel is the same as the one in the above case, and 25% consists of recycled plutonium enriched initially to 4% in weight. The figures given in table 1 are averaged for the total core.

**PWR self-generated transuranium recycle**

Again for the same reactor and same conditions, all self-generated transuranium nuclides, together with $^{99}$Tc are recycled. The figures given in table 1 are averaged for the total core.

**FR self-generated TU recycle**

The recycle of TU nuclides in the fast reactor was calculated for SUPERPHENIX assuming an initial charge of TU nuclides stemming from PWR with the following concentrations (table 2). All minor actinides are recycled together with as much of the bred Pu as needed. The concentrations are given after the 16th cycle having a burnup of 80,000 MWd/t. The out-of-pile-time is two years [3]. Again the discharge concentration for the TU nuclides are normalised to the achieved burnup in the table 1. For $^{99}$Tc a heterogeneous recycle is assumed in a thermalised position of the blanket with a transmutation half life comparable to the 960 fpd of the reactor.

For the four fuel cycles, the corresponding compositions of the fuels are:

- "once-through" LWR: $\text{UO}_2$
- Pu recycle self-generated LWR: $(\text{U}_{0.96}\text{Pu}_{0.04})\text{O}_{2-x}$
- TU recycle self-generated LWR: $(\text{U}_{0.96}\text{Pu}_{0.04}\text{Np}_{0.005}\text{Am}_{0.002}\text{Cm}_{0.001}\text{Tc}_{0.003})\text{O}_{2-x}$
- TU recycle self-generated FR: $(\text{U}_{0.8}\text{Pu}_{0.186}\text{Am}_{0.007}\text{Cm}_{0.006}\text{Np}_{0.001})\text{O}_{2-x}$

**Table 2** Initial composition of TU nuclides charged to the SUPERPHENIX-reactor.

<table>
<thead>
<tr>
<th>ISOTOPES</th>
<th>COMPOSITIONS</th>
</tr>
</thead>
<tbody>
<tr>
<td>Np 237</td>
<td>5.12%</td>
</tr>
<tr>
<td>Pu 238</td>
<td>2.15%</td>
</tr>
<tr>
<td>Pu 239</td>
<td>52.37%</td>
</tr>
<tr>
<td>Pu 240</td>
<td>21.54%</td>
</tr>
<tr>
<td>Pu 241</td>
<td>10.15%</td>
</tr>
<tr>
<td>Pu 242</td>
<td>3.49%</td>
</tr>
<tr>
<td>Am 241</td>
<td>3.95%</td>
</tr>
<tr>
<td>Am 242 m</td>
<td>40 ppm</td>
</tr>
<tr>
<td>Am 243</td>
<td>1.15%</td>
</tr>
<tr>
<td>Cm 244</td>
<td>308 ppm</td>
</tr>
<tr>
<td>Cm 245</td>
<td>144 ppm</td>
</tr>
<tr>
<td>Cm 246</td>
<td>163 ppm</td>
</tr>
</tbody>
</table>
Before the equilibrium concentrations in the mentioned fuels are reached, the content of minor actinides will vary. This is especially so in the fast reactor, where minor actinides are charged which have been produced in a thermal flux. In the development, a fuel design should consider an elasticity of the minor actinide content ranging from 1 to 2%.

Instead of having the minor actinides (and Tc) homogeneously distributed in the fuel, one could consider to concentrate them in a few fuel assemblies (heterogeneous concept). The latter approach might ease some fabrication problems, but certainly reduces the transmutation rate due to neutron resonance shielding. Since an addition of minor actinides increases the void coefficient (because of the higher fission threshold of most of the MA, a reactivity increase in case of neutron energy hardening would occur), the heterogeneous concept will not allow for significantly higher MA loading to a reactor core.

MINOR ACTINIDE-CONTAINING FUELS OR TARGETS

The European Institute for Transuranium Elements has been engaged in transuranium element research for 30 years placing main emphasis on the development of fuels and targets. Studies on minor actinide-containing fuels for the purpose of transmutation began in the 70's and led to three irradiation experiments (table 3) [4]. Any design of a specific fuel for given reactor type requires full knowledge of basic data [5], such as:

- phase diagram
- melting point of fuel
- oxygen potential
- thermal expansion
- heat capacity
- vapour pressure
- Young's modulus, etc.

In order to assess the irradiation behaviour, simulation studies are needed, such as:

- fuel restructuring in a thermal gradient by electrical heating

<table>
<thead>
<tr>
<th>Starting (Reactor)</th>
<th>Fuel</th>
</tr>
</thead>
<tbody>
<tr>
<td>1981 (FR2)</td>
<td>(U_{0.5}Am_{0.5})O_2</td>
</tr>
</tbody>
</table>
| 1983 (KNKII) | 1. NpO_2  
               2. (U_{0.5}Am_{0.5})O_{2-x}  
               3. (U_{0.73}Pu_{0.25}Np_{0.02})O_{2-x}  
               4. (U_{0.73}Pu_{0.25}Am_{0.02})O_{2-x} |
| 1986 (PHENIX) | 5. (U_{0.74}Pu_{0.24}Np_{0.02})O_{2-x}  
               6. (U_{0.5}Am_{0.2}Np_{0.2})O_2  
               7. (U_{0.55}Np_{0.45})O_2  
               8. (U_{0.74}Pu_{0.24}Am_{0.02})O_{2-x} |
- fuel-cladding interactions
- compatibility of fuel with coolant

To conduct the pertaining experiments with minor actinide-containing samples, the institute possesses unique facilities which allow the handling of bulk quantities of minor actinides [6]. The increased radiation dose of MA-containing fuels in fresh and spent fuels are discussed in a separate paper given to this meeting (G. Nicolaou, K. Richter, C. Prunier, L. Koch: Experience with Fast Reactor Fuels Containing Minor Actinides: Transmutation Rate and Radiation Doses).

The institute is carrying out an extensive research programme to develop minor actinide-containing fuels apt for the existing reactors. Minor actinides added to fast reactor oxide fuels are tested together with the CEA, France. In continuation of the above mentioned SUPERFACT experiment, further irradiations of fuels are planned [7]. The zirconium alloy based fuel concept suited for the IFR is being adapted - under commercial contract for CRIEPI [8] - to minor actinide transmutation. Irradiation experiments in the fast reactor PHENIX will start soon. As in the case of the UO$_2$ matrix, basic data for thermal conductivity, miscibility of actinides and fission products, fuel cladding compatibility, etc. were obtained from extensive experimental studies [9].

The LWR as a means for transmutation is being studied in the European Union, EU, as well. In spite of obvious disadvantages, the mere existence of such numerous LWR plants is appealing for a short term application of transmutation. In the frame of a co-operation with the CEA, the homogeneous [7] and in a network (with KfK, ECN, CEA, EdF) [10] the heterogeneous concept of minor actinide fuels and Tc are being developed.

PARTITIONING

In the case of the once-through uranium cycle, the total radiotoxic nuclide content is sent to a repository, whereas in the case of the recycling of self-generated plutonium, all of the radionuclides other than plutonium will be discharged (table 1). For the two concepts, the difference between the amount of $^{99}$Tc and $^{237}$Np is not significant. If one takes into account a loss of about 2% of plutonium for reprocessing and MOX fabrication, the plutonium waste from the plutonium fuel cycle amounts to about 5% of the Pu contained in the spent fuel, when directly stored. However, due to the continuous recycling of the plutonium and the subsequent build-up of americium and curium, the amount for the two transplutonium nuclides dramatically increases compared to the direct storage of spent fuel: 460% for americium and 570% for curium. From this follows that, due to the strong build-up of transplutonium elements in the plutonium recycle, this concept produces much more radiotoxic TPu-waste than the direct storage of spent UO$_2$ fuel.

In order to convince the public that partitioning and transmutation is a concept to reduce the future radiotoxic hazard of a repository, a considerable reduction of the radiotoxic inventory must be achieved. In order to assess the separation yields needed by a partitioning scheme, we assume in a first approach, that the radiotoxic hazard would be only 1% of that of the spent fuel of the once-through concept.

The radiotoxicity of a certain nuclide is determined by the ICRP regulations. The radiotoxic hazard, however, must be understood as the probability of radiotoxic
nuclides released from a repository to the biosphere. Here, several parameters influence the migration of a nuclide through the lithosphere, moreover, a general agreement on the assessment of the radiotoxicity is not yet reached (this is the objective of the present co-ordinated research programme to which this meeting pertains). With the lacking data determining the radiotoxic hazard of each nuclide on one hand, but the need to set targets for the partitioning efficiency for a nuclide, we have resorted to the definition of the maximum decontamination factors, which are needed to reduce the radiotoxic hazard by a factor of 100 compared to the once-through concept. I.e., the two partitioning and transmutation concepts for PWR and FR would discharge only 1% of a nuclide present in the spent fuel of the once-through concept. The figures given in table 4 show that for the higher transuranium elements, the required decontamination factors increase due to the stronger built-up of americium and curium during recycling. Again, these targets may be exaggerated, since the radiotoxic hazard is determined only by a few of the radiotoxic nuclides. For example, earlier studies showed that $^{237}$Np and $^{99}$Tc are the determining nuclides [11]. Under such a condition, the decontamination factors for the other nuclides could be considerably less.

There have been several processes developed to partition minor actinides from spent fuel. In order to assess their potential and compare their performance, we have installed in one of the chemical hot cells of the institute, a battery of centrifugal extractors. From highly active waste resulting from the earlier operation of WAK, minor actinides and residual plutonium have been partitioned [12]. These first results prove that decontamination factors as defined in table 4 can be reached.

<table>
<thead>
<tr>
<th>Decontamination Factors for a Partitioning</th>
<th>Tc</th>
<th>Np</th>
<th>Pu</th>
<th>Am</th>
<th>Cm</th>
</tr>
</thead>
<tbody>
<tr>
<td>TU recycle in LWR</td>
<td>350</td>
<td>110</td>
<td>230</td>
<td>530</td>
<td>1900</td>
</tr>
<tr>
<td>TU recycle in FR</td>
<td>310</td>
<td>64</td>
<td>1060</td>
<td>630</td>
<td>2300</td>
</tr>
</tbody>
</table>

The studies will be carried on to include newly developed compounds and, with an extended facility, to study different possibilities to separate lanthanides from the actinides.

REFERENCES


EXPERIENCE WITH FAST REACTOR FUELS
CONTAINING MINOR ACTINIDES:
TRANSMUTATION RATES AND RADIATION DOSES

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Abstract

The preparation and post-irradiation examination of minor actinide-containing oxide fuels used in the irradiation experiment SUPERFACT in the fast reactor PHENIX, have provided the opportunity to investigate the transmutation rate of minor actinides and the occupational dose during handling of such material. The experimental findings on the transmutation rates and the radiation dose levels, and their comparison with calculated values will be reported.

INTRODUCTION

In 1980s, the irradiation experiment called SUPERFACT (SF) was launched, jointly between the European Institute for Transuranium Elements (TUI) and CEA/Cadarache, to study the recycling of MA in a fast reactor (FR) [1]. A series of fast reactor oxide fuels containing Np-237 and Am-241 were fabricated and irradiated in the PHENIX reactor in order to study their material, chemical and neutronic behaviour [2, 3]. The main aim being: to prove the technical feasibility of such fuels for the option of Partitioning and Transmutation of MA.

In this paper, the experimental findings from the preparation and post-irradiation examinations (PIE) of these MA fuels compared with calculated values will be reported on: the transmutation rates of Np-237 and Am-241 during irradiation in PHENIX; the radiation dose levels encountered in the handling of such material containing MA and in particular Am-241 [4].

MA-CONTAINING FUEL FOR A FAST REACTOR

The recycling of MA in a FR is being studied through a series of oxide fuels (SF) containing Np-237 and Am-241 at low and high concentrations. The fuels were fabricated at TUI [5] in accordance with the homogeneous (SF13, SF16) and heterogeneous (SF14, SF15) fuel concepts. SF13 and SF16 contain MA homogeneously distributed in a FR-MOX fuel. Alternatively, the MA are lumped together in special targets fuels which is the case of fuels SF14 and SF15. The specifications of the fuels, together with the final burnup achieved for each fuel, are listed in Table 1.
### Table 1 Fuel characteristics of the SUPERFACT irradiation experiment

<table>
<thead>
<tr>
<th>Fuel</th>
<th>Fuel type</th>
<th>Burnup at%</th>
<th>Fuel density g/cm³</th>
<th>Fuel length (cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>SF13</td>
<td>$^{(0.74}\text{U},^{0.24}\text{Pu},^{0.02}\text{Np})\text{O}_2$-x Unat Pu 238: 1.3% Pu 239: 60.4% Pu 240: 23.4% Pu 241: 10.4% Pu 242: 4.5%</td>
<td>6.4</td>
<td>10.5</td>
<td>85</td>
</tr>
<tr>
<td>SF14</td>
<td>$^{(0.6}\text{U},^{0.2}\text{Am},^{0.2}\text{Np})\text{O}_2$ U 235: 0.3%</td>
<td>4.1</td>
<td>10.3</td>
<td>40</td>
</tr>
<tr>
<td>SF15</td>
<td>$^{(0.65}\text{U},^{0.45}\text{Np})\text{O}_2$ U 235: 0.3%</td>
<td>4.5</td>
<td>10.5</td>
<td>40</td>
</tr>
<tr>
<td>SF16</td>
<td>$^{(0.74}\text{U},^{0.24}\text{Pu},^{0.02}\text{Am})\text{O}_2$-x Unat Pu: idem SF13</td>
<td>6.4</td>
<td>10.5</td>
<td>85</td>
</tr>
</tbody>
</table>

Pellet radius 0.271 cm
Pellet height 0.700 cm
Stainless steel cladding
O.D. : 0.655 cm
I.D. : 0.700 cm

### EXPERIMENTAL

#### Isotopic analyses of MA fuels

Examination of isotopic composition were carried out on samples of approximately 1g cut halfway along the active length of the fuel pins. The samples were then dissolved and aliquots taken for chemical analyses. The concentrations of uranium, plutonium, americium and curium isotopes and Nd-148 were measured by Isotope Dilution Mass Spectrometry (IDMS). The measurement of Np-237 was performed by Inductively Coupled Plasma Mass Spectrometry (ICP-MS). The burnup each fuel achieved during irradiation in PHENIX was derived from the number of final heavy metal atoms and Nd-148 for each sample. A quantitative analyses of the fission products contributing to the gamma output of the fuels was performed by gamma-spectroscopy relative to Cs-137.

#### Dosimetry measurements of MA fuels

The gamma dose rates of the four SF fuel pins were measured during fabrication using a portable ionisation chamber and thermoluminescence dosimeters in contact [6]. After irradiation (cooling time 57 months), the gamma and neutron dose rates were measured using a passive neutron-gamma interrogation unit developed for spent fuel characterisation inside a hot cell at TUI [4, 7].
THEORETICAL

The program KORIGEN [8] has been used to calculate for each of the SF fuels the actinide and fission product concentrations, the contribution to the total burnup for each fissile nuclide and the neutron and gamma outputs at different cooling times. The fission and capture cross-section libraries were adjusted to those for the PHENIX reactor for the transuranium isotopes. The neutron and gamma outputs were also calculated, for comparison purposes, for a standard FR-MOX fuel containing plutonium of the same isotopic composition as the SF homogeneous fuels. The final burnup achieved by each fuel as determined experimentally, was used as a normalisation parameter in the calculations.

The neutron and gamma dose rates, were calculated at TUI for each of the fresh SF fuels. The calculations were performed, using the computer program PUDOL [9], on the basis of the charge composition, certain physical properties (such as density and size) and cladding (Table 1). The program considers self-shielding effects in the fuel itself and attenuation in the cladding. Furthermore, it allows a range of shielding materials for radiation protection purposes.

RESULTS AND DISCUSSION

Transmutation rates

The primary information to be confirmed for a given recycling strategy of MA is the change in the MA inventory during an irradiation. Chemical analyses has been carried out on diluted solutions of dissolved pellets from the SF fuels in order to determine the rate by which Np-237 and Am-241 were transmuted during irradiation in PHENIX.

The isotope vectors of uranium, plutonium, americium and curium, and Np-237 were measured as atom concentration in solution. These results, corrected for decay are converted in % Initial Metal Atoms (%IMA) from which the transmutation rates are deduced. A direct comparison between BOL (begin of life) and EOL (end of life) compositions yields the reduction in Np-237 and Am-241 through both fission and capture. The experimental and theoretical results on the reduction of Np-237 and Am-241 are shown in Table 2. The comparison of BOL and EOL %IMA concentrations, reveals a reduction of about 29% and 28% for Np-237 and Am-241 respectively. This reduction corresponds to transmutation half-lives of about 2 years and 2.2 years respectively for the nominal irradiation period in PHENIX of 375 days.

The agreement between the measurements and theoretical predictions is within 6% except for the recycling scenarios of 2%Np (16%) and 20%Np (20%). The differences observed in the Np-237 results, are due to errors in the ICP-MS determination of neptunium and capture cross-sections (the present accuracy of Np-237 determination by ICP-MS is about 2% which propagates to 8% for the differences). The Pu-238 formed from Np-237 is consistently lower for the theoretical results (Table 3) indicating that the neutron capture cross-section of Np-237 is underestimated.
Table 2 Experimental and theoretical decrease (%) and half-lives in PHENIX

<table>
<thead>
<tr>
<th></th>
<th>SF13</th>
<th>SF14</th>
<th>SF15</th>
<th>SF16</th>
</tr>
</thead>
<tbody>
<tr>
<td>Measured TUI</td>
<td>Np-237</td>
<td>Am-241</td>
<td>Np-237</td>
<td>Am-241</td>
</tr>
<tr>
<td></td>
<td>29.6</td>
<td>27.6</td>
<td>33</td>
<td>27.8</td>
</tr>
<tr>
<td>Predicted TUI</td>
<td>24</td>
<td>30.3</td>
<td>26</td>
<td>26.3</td>
</tr>
<tr>
<td>Experimental half-life (a)</td>
<td>2.05</td>
<td>2.2</td>
<td>1.8</td>
<td>2.2</td>
</tr>
</tbody>
</table>

Table 3 Experimental and theoretical % IMA values

<table>
<thead>
<tr>
<th></th>
<th>%IMA (exp.)</th>
<th>%IMA (theor.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>UPu, 2%Np</td>
<td>Np237: 1.37</td>
<td>Pu238: 0.67</td>
</tr>
<tr>
<td></td>
<td>Np237: 0.020</td>
<td>Pu238: 0.42</td>
</tr>
<tr>
<td>U, 20%Am, 20%Np</td>
<td>Np237: 14.12</td>
<td>Pu238: 6.32</td>
</tr>
<tr>
<td></td>
<td>Np237: 8.5</td>
<td>Pu238: 7.7</td>
</tr>
<tr>
<td>U, 45%Np</td>
<td>Np237: 32.4</td>
<td>Pu238: 8.5</td>
</tr>
<tr>
<td></td>
<td>Np237: 32.9</td>
<td>Pu238: 7.7</td>
</tr>
</tbody>
</table>

Radiation dosimetry aspects

The inclusion of MA, and in particular Am-241, in a fuel would affect the radiation dose levels of the fuels. A significant increase in the gamma and neutron outputs is predicted by KORIGEN for the SF fuels over the standard FR-MOX fuel during fabrication. At discharge, the gamma outputs are comparable; however, the SF fuels containing Am-241 have a significantly higher neutron output due to Cm-242. The neutron outputs decrease with cooling time to that of the FR-MOX fuel at about 5 years after discharge. The total dose rate is now dominated by the gammas [4]. Additional means in order to reduce the occupational dose from the MA fuels would be required certainly during fabrication. Knowledge of these dose levels would allow measures for additional shielding in handling these materials. The gamma dose rates from the SF fuel pins during fabrication are given in Table 4. The high radiation doses from the americium-containing mixtures can be seen in the results.

The recycling of transuranium isotopes has been studied through three strategies: the MA recycling in a FR which corresponds to the SUPERFACT irradiation experiment, the recycling of (MA+Pu) in a PWR and the (MA+Pu) self-generated in FR [10]. Studies are in progress to evaluate the gamma and neutron dose rates from a pellet, fuel pin and fuel assembly characteristic of the reactor type considered in the strategy.
Table 4 Dose rates at 1m from a fresh fuel

<table>
<thead>
<tr>
<th>Fuel</th>
<th>Measured Gamma dose rate (mSv/h)</th>
<th>Predicted Gamma dose rate (mSv/h)</th>
<th>Neutron dose rate (mSv/h)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2% Np</td>
<td>7E-3</td>
<td>8E-3</td>
<td>1.7E-4</td>
</tr>
<tr>
<td>20%Am, 20%Np</td>
<td>0.26</td>
<td>0.25</td>
<td>3.3E-4</td>
</tr>
<tr>
<td>45% Np</td>
<td>&lt;0.01</td>
<td>2E-4</td>
<td>1E-7</td>
</tr>
<tr>
<td>2% Am</td>
<td>0.04</td>
<td>0.056</td>
<td>2E-4</td>
</tr>
</tbody>
</table>

The availability of computer programs enabling the prediction of EOL composition and dose rate aspects of fuels is essential in view of estimating gains and hazards in fuel cycles. The agreement of our measurements with the KORIGEN predictions for the source term gamma and neutron emitting nuclides has been found to be within 20% and 50% for the actinides and fission products respectively [4]. This agreement is sufficient for composition studies associated with dosimetry measurements, but unsuitable for neutron physics calculations. The uncertainties on the cross-section and fission yield libraries are unknown; however, one of the reasons of PIE isotopics is the improvement of these libraries, in progress under a separate study.

In order to assess the reliability of the dosimetry computer program, experimental and theoretical dosimetry results were compared for two sets of experiments. The first is based on the dose rates measured during fuel pin fabrication for the SF fuels (Table 4). The level of agreement is within the accuracy required for radiation protection purposes. The 45%Np case is difficult to assess due to the very low gamma component of the fuel. The second comparison is based on the KNK II/b fresh pellets [11, 12]. Calculations have been carried out at 5 cm distance assumed to correspond to the nominal centre of the ionisation chamber used in the measurements. For comparison purposes, the calculations performed (under contract) by UKAEE, Winfrith, UK [12] have been included. The experimental and theoretical results, together with the specifications of the pellets, are shown in Table 5. The calculations by UKAEE have been based on the sample size and density, and a list of 18-group photon intensities as calculated using KORIGEN. The agreement between the measured and calculated (TUI) dose rates is within 15% which is sufficient for dosimetry purposes. It can be concluded therefore that the TUI dose rate calculations can be considered sufficiently reliable.

Table 5 Characteristics of fresh actinide pellets and gamma dose rates at 5 cm

<table>
<thead>
<tr>
<th>Pellet</th>
<th>Density g/cm³</th>
<th>Gamma dose rates (mSv/h)</th>
<th>predicted TUI</th>
<th>predicted UKAEE</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>measured TUI</td>
<td>predicted TUI</td>
<td></td>
</tr>
<tr>
<td>50%Am, 50%U</td>
<td>9.8</td>
<td>7.8</td>
<td>8.7</td>
<td>11.9</td>
</tr>
<tr>
<td>74%U, 24%Pu, 2%Am</td>
<td>9.94</td>
<td>0.37</td>
<td>0.42</td>
<td>0.52</td>
</tr>
<tr>
<td>74%U, 24%Pu, 2%Np</td>
<td>9.84</td>
<td>0.074</td>
<td>0.089</td>
<td>0.084</td>
</tr>
</tbody>
</table>

Pellet specifications: radius 3.2 mm, height 7.0 mm, \( \frac{Pu_{fiss}}{Pu_{tot}} = 0.8 \)
REFERENCES


[10] L. Koch, G. Nicolaou, this conference


CONTROL OF ACCELERATOR DRIVEN SUBCRITICAL SYSTEMS FUELED BY ACTINIDES

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Abstract

The OECD/NEA and the CEC have been carrying out studies of new transmutation strategies in an international network. It has been shown that an integration of transmutation techniques could reduce substantially the long-term radiation hazard from nuclear waste. Transmutation could also contribute to a safe and even beneficial de-commissioning of nuclear weapons. An additional benefit of this more economic use of fuel could be a reduction of radiation hazards from uranium mining. In the search for transmutation concepts accelerator driven sub-critical actinide and isotope incinerators have attracted a great deal of attention especially in Japan and the US. One reason is the fact that actinide fuelled (critical) reactors pose a particular problem of control as they are characterized by a small delayed neutron fraction coupled with a non-negative Doppler coefficient. In this context a new formula relating the accelerator energy requirements with the amount of fission product transmutation and the multiplication factor of the system was developed. Also one point kinetic studies dealing with fast transients as a function of reactivity insertion in accelerator driven systems were carried out. They show that such boosters behave quite benignly even if they are only slightly subcritical.

INTRODUCTION

The OECD Nuclear Energy Agency and the CEC have been carrying out studies of new transmutation strategies in an international co-operative effort. It has been shown that an integration of transmutation techniques could reduce substantially the long-term radiation hazard from commercial waste. Transmutation could also contribute to a safe and even beneficial decommissioning of nuclear weapons. An additional benefit of this more economic use of fuel could be a reduction of radiation hazards from uranium mining.

In the search for transmutation concepts the first candidates were actinide fuelled (critical) reactors. But soon it turned out that they pose a particular problem of control. This is due to the fact that the fissile isotopes of Neptunium, Americium, and Curium have a considerably smaller fraction of delayed neutron emitters (as compared to the more common fuels U-235 and U-238), a small Doppler effect and a non-negative void coefficient. As is well known, the fraction of delayed neutrons is essential for the control of a nuclear reactor in the critical state.

To overcome these problems various concepts of accelerator driven subcritical systems aiming at the transmutation of actinides and long lived fission products have been proposed in the recent past. Especially in Japan and the US these systems have attracted a great deal of attention.

The safety of a multiplying system depends almost entirely on fast transients caused by accidental reactivity insertions. To study the power changes in accelerator driven systems a kinetic model dealing with fast transients as a function of reactivity insertion was developed and programmed.
The model allows a comparison with an equivalent critical reactor. It was tested by a comparison with a NEACRP benchmark. As a general tendency it turned out that accelerator driven systems behave quite benignly even if they are only slightly subcritical.

THE KINETIC MODEL

In the following considerations we use the conventional point kinetics equation to which the term \( S(t) \) is added. It describes an external source which in our case are the spallation neutrons generated by a proton accelerator.

\[
\begin{align*}
\frac{dN}{dt} &= \rho(t,N) - \beta \frac{N}{A} + \sum_{i=1}^{6} \lambda_i C_i + S(t) \\
\frac{dC_i}{dt} &= \frac{\beta}{A} - \lambda_i C_i; \quad i = 1, 2, \ldots, 6
\end{align*}
\]

where

- \( N \) = number of neutrons in the system (it is considered to be proportional to the power),
- \( C_i \) = delayed precursor concentration of the \( i \)-th delayed neutron group,
- \( \lambda_i \) = decay constant of the \( i \)-th delayed precursor group [sec\(^{-1}\)],
- \( \beta_i \) = delayed neutron fraction of the \( i \)-th delayed precursor group,
- \( \beta \) = total delayed neutron fraction \( (= \beta_1 + \beta_2 + \ldots + \beta_6) \),
- \( \rho(t,N) = \rho_R(t) + \rho_D(N) \) total reactivity variation caused by the time dependent ramp-rate \( \rho_R(t) \) and the power (neutron population) dependent Doppler reactivity \( \rho_D(N) \),
- \( A \) = prompt neutron lifetime [s],
- \( S(t) \) = rate at which external neutrons are inserted. It is chosen such that a certain power level is maintained in the system.

The coupled equations (1) and (2) are solved by a numerical method employing a variable implicit technique [3]. The method yields an efficient and accurate solution. The general features of the program include time dependence of the total reactivity, prompt neutron generation time and time step size, and a maximum of six delayed neutron precursor groups. In addition the total stored energy is also calculated by integrating the reactor power from \( t = 0 \) to the time of interest.

The solution of Equations (1) and (2) is based on the program of Cheng [3] to which the following features were added:

1) The possibility of inserting a linear or quadratic time dependent reactivity ramp. The quadratic time dependent reactivity ramp serves for the simulation of gravity induced accidents like earthquakes, etc.
2) A negative reactivity feed-back mechanism to take the Doppler-effect into account.
3) The possibility to shut down the external neutron source by an exponential, \( \tau \) dependent, decay law of the form \( \exp(-t/\tau) \).
4) A graphical display of the power and reactivity changes.

THE EXTERNAL SOURCE

According to Ref. [1] the power production \( P_{fi} \) of a subcritical assembly fed by spallation neutrons is quantified as:
where:

\[ k = \text{multiplication factor of the subcritical system} \]

\[ a = \text{importance of the target position and target neutron energy distribution} \]

(usually \( a > 1 \) for a central target position)

\[ \nu = \text{mean number of neutrons in a fission process} \]

\[ E_f = \text{power release per fission} (= 3.1 \times 10^{10} \text{ W}) \]

\[ n_{sp} = \text{neutron yield from one proton} \]

\[ I = \text{proton current} \]

\[ C = \text{proton charge} (= 1.6 \times 10^{-19} \text{ A sec}) \]

It can be seen that near criticality a 1 mA current already generates a relatively high fission power. For \( k = 0.97 \) more than 100 MW can be achieved.

One can assume that \( S(t) \equiv -\rho_0 n_{sp} / \Lambda \) is a good approximation since the spectrum of the spallation neutrons is quite similar to the fission spectrum, except for a tail of fast neutrons above 20 MeV. It follows therefore that

\[ S(t) = P_{fi} \frac{\rho_0 \nu(1-k)}{\Lambda} \frac{C}{TE_f} \]

THE EFFECT OF UNPROTECTED REACTIVITY ACCIDENTS

Usually three types of unprotected reactivity accidents are considered:

- Slow reactivity ramp insertion,
- Fast reactivity ramp insertion,
- LOF driven TOP (Fast reactivity ramp insertion due to sodium voiding caused by a loss of coolant accident.)

Slow reactivity ramp insertions without a scram are for example the inadvertent withdraw of a control rod(s) (few cents/sec or .0001 \( k_{eff} \)/sec).

A typical fast reactivity ramp insertion occurred in the EBR-I accident which was caused by an inward bowing of the fuel pins. All later fast reactors were constructed with grids or helical wire spacers to prevent bowing. Other accidents of this category are earthquakes or diagrid failures without a scram (up to a few \$ / sec or .01 \( k_{eff} \)/sec).

The NEACRP Benchmark Problem

As a first example the KfK benchmark problem defined as a rod ejection accident and proposed by the Nuclear Energy Agency Committee on Reactor Physics (NEACRP) is chosen. It consists of a fast reactor made up of a core with a bank of annular control rods, radial and axial blankets and sodium coolant. The essential features of the problem are: Axis-symmetry, two neutron groups and six delayed neutron precursor families and thermal feedback through Doppler effects in capture and fission cross sections.

The transient is obtained through steady control rod bank withdrawal. The reactivity insertion starts at 1 ms and increases at a rate of 170 \( \$ /s \) for the duration of 16 ms. (The speed of the control withdrawal is adjusted to produce a ramp of 0.548 cm /ms.) After this time the reactivity is kept constant.
The reactivity reduction by the Doppler coefficient was calculated from the sample data obtained from Beauwen's paper [2] as a heat generation coefficient of $-0.92108 \text{ S} / \text{GJ}$.

The analysis of this problem allows a comparison with transient calculations obtained by others to validate the code used in our analysis. It also gives a first indication of the mitigating effect of using a subcritical, accelerator driven system.

Figures 1a and 1b show the power and reactivity change in a critical reactor (solid line, curve no. 4) and in systems being sub-critical between $-3 \text{ S}$ and $-1 \text{ S}$ (dotted lines, curves no. 1 to 3). These systems are driven by a spallation source dimensioned such that they generate in steady-state operation the same power as the critical reactor, which is assumed to be 1 GW. Curve 4 corresponds to a critical reactor. Its power oscillates and has two peaks in a short time interval. Super-prompt criticality produces these peaks, as can be seen in Figure 1b. The power rises rapidly during the period of super-prompt criticality and reaches its peak value at the time when the Doppler effect reduces the reactivity to values below the super-prompt limit. (This characteristic is similar to a pulsed reactor).

In the case where the reactor is operated in a subcritical mode the neutron source is determined such that the system generates 1 GW thermal power and this source strength is maintained during the whole time the reactivity is increased. When the time reaches 17 ms, or when the thermal power of the reactor reaches 50 times of the initial power (50 GW), the neutron source is reduced by the shut-off function $exp(-\tau/\tau_r)$ ($\tau = 1 \text{ ms}$).

The case in which the initial sub-criticality is $-3$ and $-2 \text{ S}$ respectively, the power increases only to 2.2 GW and 6 GW respectively after 16 ms and after 17 ms the power decreases almost proportionally with the neutron source strength of $exp(-\tau/\tau_r)$ (curves 1b and 2b). If on the other hand the neutron source is maintained constant (the accelerator is not shut-off), also the power remains almost constant in this time range (curves 1a and 2a). For a sub-criticality of only $-1 \text{ S}$ a single peak-power of 530 GW was calculated. Even though this value is similar to the peak of curve 4, the integrated power, i.e. the total energy release during the excursion is much less than for a critical system.

An interesting result of this analysis is the fact that the power decreases even between the prompt and delayed critical state. This is due to the long time constant of the delayed neutrons. When the reactor is in an under prompt-critical condition, the neutron flux is controlled by prompt neutrons which decrease for a sub-supercritical condition. For the case of a critical reactor two power peaks (curve no. 4) of 700 GW and 500 GW resp. were calculated which is in good agreement with the results of the NEACRP benchmark.

Figure 2 deals with the case of a short neutron life time: only half the value of the case above is used. The time dependence of power in the case of $-3 \text{ S}$ and $-2 \text{ S}$ are almost the same as in Figure 1. This means that in a subcritical system the power change in time is almost independent from the neutron life time. Only when the reactor becomes super-critical, the power increases slightly faster than in systems with longer neutron life time, as shown in 3-a and -b. The power peak value of a system with long neutron life time is slightly higher than the one for a short neutron life time. But the system with short neutron life time has a 'shoulder' around 15 ms which is absent in the case of a long neutron life time. The critical reactor has three peaks instead of two, but the peak power is only slightly higher than the one for a long neutron life time.
Figure 1a: Power excursion after a reactivity insertion accident (170 $/s during 15 ms); 4: critical reactor, 1 to 3: subcritical accelerator driven systems (see text).

Figure 1b: The reactivity behaviour during the accident scenario.
Figure 2a: Power excursion after a reactivity insertion accident (170 $/s$ during 15 ms) and a very short neutron generation time (half the value of Fig.1); 4: critical reactor, 1 to 3: subcritical accelerator driven systems (see text).

Figure 2b: The reactivity behaviour during the accident scenario.
Figure 3a: Power excursion after a reactivity insertion accident (170 $/s$ during 15 ms); 4: critical reactor, 1 to 3: subcritical accelerator driven systems (see text).

Figure 3b: The reactivity behaviour during the accident scenario.
When the neutron life time and delayed neutron portion becomes half the value of the original case, then the power change as well as the reactivity change in units of dollars (as a function of time) becomes identical to the first case. This means that power and reactivity are time scale invariant in both the neutron life time and delayed neutron portion $\beta$. This can be easily verified by an analysis of equation 1, where a simultaneous change of the parameters in the numerator and denominator does not change the numerical value of the coefficients.

When the power change is slow, the reactor can be controlled by a mechanical movement of control rods or by a hydraulic dispersion of liquid neutron absorbers which are dispersed by melting fuel elements like an electric fuse mechanism. In a subcritical reactor operated by spallation neutrons, the power change is much slower than in a critical reactor. This provides a great advantage from the point of view of reactor safety.

CONCLUSIONS

A new one point kinetics program was developed. It allows the simultaneous calculation of a critical and sub-critical, externally driven system using the same input parameters. The code was validated by re-calculating a NEACRP specified benchmark dealing with the example of a rod ejection accident in a fast reactor.

The few examples treated show that even slightly subcritical systems which require only a low accelerator current, respond much more benignly to a sudden reactivity insertion than critical systems. As can be seen from the figures already a subcriticality of $-3$ $\$ \$ reduceuces the energy release to less than a percent of that of a corresponding critical reactor.

It appears that a systems with a $k_{eff}$ of around $0.9 \sim 0.95 \sim -30 \sim -15 \$ for a FR) would even look more attractive from the safety point of view. But these systems would require an expensive high current accelerator. In addition they are characterized by an inhomogeneous power distribution with a sharp peak around the target area.

On the other hand, as we have shown, accelerator driven slightly subcritical systems behave more benignly than the "classical" reactors. They also have a relatively flat power distribution, and require only low current and therefore much less expensive accelerators.

REFERENCES


APPENDIX

Nuclear Parameters used in the Preceding Calculations:

Delayed Neutron Data (used in all calculations)

\[ \beta_{1\text{m6}}: 2.4700E-4, 1.3845E-3, 1.2222E-3, 2.6455E-3, 8.3200E-4, 1.6900E-4 \]
(These values are re-normalized by \( \beta \).)

\[ \lambda_{1\text{m6}}: 0.0127, 0.0317, 0.115, 0.311, 1.400, 3.87 \]

Case 1:
\[ \beta = 0.00378, \Lambda = 3.3E-7, \text{ Reactivity Insertion Rate} = 170 \text{ $\mu$s, Doppler Coefficient} = 0.9211 \]
Curves 1a to 3a, \((1 - k_{\text{eff}}) = 0.01134, 0.00756, 0.00378; \text{ the accelerator is shut-off after } 17 \text{ ms or when the power reaches } 50 \text{ times the initial value.} \)
Curves 1b to 3b, \((1 - k_{\text{eff}}) = 0.01134, 0.00756, 0.00378; \text{ the accelerator is not shut-off.} \)
Curve 4, \((1 - k_{\text{eff}}) = 0.0, \text{ the system is critical, (no accelerator is used).} \)

Case 2:
\[ \beta = 0.00378, \Lambda = 1.65E-7, \text{ everything else like Case 1.} \]

Case 3:
\[ \beta = 0.00189, \Lambda = 1.65E-7, \text{ Reactivity Insertion Rate} = 170 \text{ $\mu$s, Doppler Coefficient} = 0.9211 \]
Curves 1a to 3a, \((1 - k_{\text{eff}}) = 0.00567, 0.00378, 0.00189; \text{ the accelerator is shut-off after } 17 \text{ ms or when the power reaches } 50 \text{ times the initial value.} \)
Curves 1b to 3b, \((1 - k_{\text{eff}}) = 0.00567, 0.00378, 0.00189; \text{ the accelerator is not shut-off.} \)
Curve 4, \((1 - k_{\text{eff}}) = 0.0, \text{ the system is critical, (no accelerator is used).} \)
THE ACTIVITIES OF OECD/NEA ON PARTITIONING AND TRANSMUTATION

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Abstract

The OECD Nuclear Energy Agency (NEA) has been involved since the late 1980s in various activities on Actinide and Fission Product Partitioning and Transmutation. The purpose of this paper is to provide a summary of NEA's past and present projects and highlight its future directions.

1. BACKGROUND

Although Partitioning and Transmutation (P&T) cannot be seen as an alternative to the present policies concerning the back-end of the fuel cycle, it is increasingly recognised that it may contribute to improved efficiency of high level waste disposal and utilisation of resources of the spent fuel. Since 1989, the NEA has stressed that this topic is one of long term scientific and technological development; nevertheless, certain short- or medium-term benefits could also be derived. While a number of bilateral agreements on P&T between OECD countries do exist, clearly, more benefit could be achieved from wider international activities and co-operation.

The activities of the NEA's Nuclear Development Committee (NDC) are conducted under the guidance of Liaison Officers nominated by Member countries. Under the "umbrella" of the NDC, two successful International Information Exchange meetings were held at Mito City and Argonne National Laboratory in 1990 and 1992, respectively. One important outcome of the Argonne meeting was agreement that the next NEA Information Exchange meeting, which will be held at Cadarache in November 1994, should devote some time to an assessment of current approaches to systems studies of introducing actinide partitioning and transmutation into the nuclear fuel cycle. Such studies would form the core of future NDC activities.

It is expected that the NDC will extend its cycle of P&T activities at the end of 1994 for an additional, possibly 5 years, period. To this end, a small advisory group will be formed in early 1994 to assist the NDC with its projects.

The programme of work of the NEA Nuclear Science Committee (NSC) covers several items related to partitioning and transmutation. The main emphasis has so far been on different nuclear data needs for accelerator and reactor transmutation applications. A study involving chemistry questions relevant to partitioning has also been conducted. The Committee has initiated a Task Force to investigate the physics aspects of different transmutation concepts.

A number of projects have been or are being pursued in the field of intermediate energy nuclear data. Two studies were recently completed: one on the availability of experimental data and nuclear model codes, and one on the requirements for an evaluated nuclear data file. An international comparison of the
performance of computer codes used in intermediate energy data calculations is being completed. The results will be published in 1994. A data base of available experimental data will be set up, as well as a cooperative project on an evaluated nuclear data file.

A project on geo-chemical calculations of uranium in natural waters was completed a few years ago. The NSC is at present discussing chemical items related to the fuel cycle, and subjects that have been mentioned include: acid chemistry of Np and Tc, separation of trivalent lanthanides from Am and Cm, and chemical treatment of heterogenous actinide pins for reactor recycling. The NSC will discuss these items further at their next meeting in June 1994.

2. THE ACTIVITIES OF THE NDC

Formally, the NDC's first P&T Exchange Programme was intended to run for 5 years and will thus be concluded in late 1994 with the 3rd International Information Exchange meeting in Cadarache, France. The NDC will consider during its 1994 meetings the continuation of its programme for a further period of, possibly, 5 years. It should be noted that past and present NDC activities were assisted by generous financial contributions provided by the Government of Japan.

The first NEA International Information Exchange meeting was held at Mito City in Japan in November 1990. During the meeting a provocative set of papers had been presented both on policy orientations and on scientic aspects. The discussion, which concentrated on wide ranging ideas regarding future technologies, led to the conclusion that several disparate approaches had already been taken. It was agreed to organize small specialist meetings on suitable topics such as fundamental physics and chemical data requirements, the chemistry of new partitioning processes and the engineering of accelerators. The proceedings of the meeting were published and distributed by the NEA.

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

The specialist meetings held so far have been found most useful by the participants. There had been considerable discussion in which the results achieved in earlier and current work in the USA and Europe had been used to illuminate the more recent work in the vigorous programme being pursued by Japan. One of the points which had come forcibly into focus was that there seemed to be little point in reducing the long-lived radioactivity of the minor actinides, unless, at the same time, attention was paid to reducing markedly the quantity of long-lived radioactive fission
products. There had been a wealth of suggestions for different flow sheets with which to improve the manipulation of actinides into different product streams and for alternative reactor and accelerator-driven transmutation of actinides and fission products.

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

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

The 3rd NEA International Information Exchange meeting will be held at Cadarache, France, in November 1994. Prior to this meeting a small advisory group will be formed under NDC auspices to:

(a) advise and assist the NDC with its P&T activities and long-term orientations;

(b) provide technical assistance for the 1994 NEA General Exchange meeting and, in particular, address central issues and define criteria oriented towards establishing a comparison framework for different P&T systems studies;

(c) review country positions on P&T and group cases accordingly.

The active involvement of NEA's Radioactive Waste Management Committee (RWMC) will continue to be sought.

As indicated above, it is expected that a significant part of the 1994 General Exchange meeting would be dedicated to evaluating a limited number of different P&T systems and their integration into the nuclear fuel cycle. Other areas of interest would be the effects of progress made with various national P&T activities, technical advances, economic assessments, objectives of the P&T and environmental impact considerations. It is currently intended to invite participants from the Russian Federation to the 1994 General Exchange meeting.

Last, in consultation with other NEA Committees, national laboratories and other International Organisations, it will be attempted to find suitable ways and appropriate fora to discuss and evaluate the general health and environmental benefits and detriments of different P&T options.
3. THE ACTIVITIES OF THE NSC AND THE DATA BANK

The NSC and the NEA Data Bank have carried out several programmes related to Partitioning and Transmutation. Some of them have been performed within the NSC's own programme of work, whereas others were carried out using funds from the Japanese voluntary contributions to the NEA.

I. Completed programmes

1) Review of Fission Product Yields and Delayed Neutron Data from the Actinides (July 1990)

A critical review of available fission product yields and delayed neutron data for the actinides of interest to the transmutation, i.e., $^{237}\text{Np}$, $^{242}\text{Pu}$, $^{242m}\text{Am}$, $^{243}\text{Am}$, $^{243}\text{Cm}$ and $^{245}\text{Cm}$ has been performed. The study has identified gaps and inconsistencies in the existing data base and also identified priority areas for further experimental, theoretical and evaluation efforts.

2) Uranium in Natural Waters: Some Geochemical Calculations with the NEA Thermochemical Data Base (July 1991)

The NEA has been working on setting up a Thermochemical Data Base, which could also be of use for studies relevant to the partitioning of actinides. The Uranium data, present in this data base, were used in a validation calculation of uranium compounds in aqueous solutions.

3) Review of Intermediate Energy Data and Model Codes for Accelerator-Based Transmutation (December 1992)

This study investigated the availability of experimental intermediate energy data. The need for a comprehensive compilation effort of these data was stressed. The most important nuclear theories and some of the associated nuclear model codes were described, and their applicability to intermediate energy nuclear data calculations was discussed.

4) Requirements for an Evaluated Nuclear Data File for Accelerator-Based Transmutation (June 1993)

The importance of evaluated intermediate energy nuclear data files as part of a global calculation scheme for accelerator-based transmutation was discussed. The resulting report contains a proposal for setting up the following three data libraries:

- a data library from 0 to 100 MeV (first priority),
- a reference data library from 20 to 1500 MeV,
- an activation library from 0 to about 100 MeV.
II On-going programmes

1) Comparison of Codes for Calculation of Intermediate Nuclear Data

An international comparison study is being coordinated. The aim is to assess the predictive ability of computer codes used in calculating intermediate energy charged particle data. Two cases have been defined: one where thin target data of $^{90}$Zr and $^{208}$Pb should be calculated and one with the objective to predict the neutron yield and mass distribution of spallation products from 800 MeV proton bombardment of Bi. The results, to be compared to experimental data, are being collected and analysed. A report will be published in 1994.

2) International Evaluation Co-operation

The NSC Working Party on International Evaluation Co-operation has started an activity to evaluate intermediate nuclear energy data. The project will partly build on the results from the study on the Requirements for an Evaluated Nuclear Data File for Accelerator-Based Transmutation mentioned above. The Working Party will discuss items such as the type and format of the data to be included in the library, before starting the actual evaluation effort. The first data files are expected to be ready for testing in 1994.

3) Review of the Physics Aspects of Different Transmutation Concepts

This programme aims to analyze discrepancies and uncertainties with respect to the scientific performance of different transmutation concepts, and to identify the crucial issues for the technical feasibility of these devices. A first overview report, restricted to programme participants, will be ready in early 1994. The final report is expected to be published at the end of 1994.

III Proposed future programmes (to be decided by the NSC)

1) Fuel Cycle Chemistry

The NSC has set up a Task Force to review the Committee's future activities in fuel cycle chemistry. Items concerning chemical data needed for partitioning, for example, chemical treatment of heterogeneous samples for reactor recycling, have been suggested in the preliminary discussions.

2) A Data Base for Experimental Intermediate Energy Data

According to a recommendation in the report on Review of High Energy Data and Model Codes for Accelerator-Based Transmutation mentioned above, it has been proposed to set up a data base for experimental intermediate energy data. The new data base will be based on available information contained in existing data bases, supplemented by information concerning data of essential interest to accelerator transmutation needs.
5. SUMMARY

The NEA has provided and will continue to offer a forum for technical discussions on Partitioning and Transmutation activities and national programmes. A number of science and data collection activities have been completed and new projects will be undertaken in the future. The NEA's P&T programme of work will be strengthened by adding new activities associated with comparisons of systems studies and the evaluation of the overall impacts of various P&T options.
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LIST OF RELATED IAEA PUBLICATIONS

1982  Evaluation of Actinide Partitioning and Transmutation
      (Technical Reports Series No. 214)

1989  Feasibility of Separation and Utilization of Ruthenium, Rhodium and Palladium from
      High Level Wastes
      (Technical Reports Series No. 308)

1993  Feasibility of Separation and Utilization of Caesium and Strontium from High Level
      Liquid Waste
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