SESSION I

OPENING REMARKS AND INTRODUCTORY PAPERS

CHAIRMAN: L.H. BAETSLÉ

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The introductory papers clearly showed a growing interest in Partitioning and Transmutation (P&T) from the international community. The OECD-NEA announced the continuation of the P&T assessment study which will be focused on Accelerator Driven Transmutation Systems (ADS). The EURATOM programme managed by the European Commission and the five successive OECD-NEA information exchange meetings have greatly contributed to international collaboration.

The International Atomic Energy Agency (IAEA) has conducted a more basic programme aimed at the coverage of P&T activities from non OECD-NEA member countries and focused on safety, environmental and non-proliferation aspects of P&T.

The Japanese OMEGA and French SPIN programmes are the engines of the world-wide current R&D activities.

The OMEGA programme produced a broad spectrum of basic and applied research and pioneered the concept of the double strata fuel cycle and Fast Burner Reactors. The programme has been reviewed and will be revised in 1999. The new major direction of activity is oriented towards ADS and its associated fuel cycle. A powerful ADS machine is under construction in Japan.

The French SPIN programme achieved breakthroughs in the new partitioning methods and in the global impact of P&T on the current fuel cycle. The future evolution of the French R&D programme will be based on the comprehensive investigation of the double strata concept and its impact on the nuclear waste programme. The French P&T programme aims at reducing the waste radiotoxicity by a factor of 100. Long-term fundamental studies in the ADS field (GEDEON association) are programmed for the next decade.

A European partitioning programme, NEWPART, coupling institutes with chemical expertise, has led to very promising results:

- the DIAMEX process has been tested with success in the two different institutes.
- the combination of DIAMEX-CYANEX and SESAME can lead to the complete separation of actinides.

Partitioning activities in Russia are based on the proven methods to separate heat emitting nuclides (^{137}Cs and ^{90}Sr) and minor actinides. An industrial extraction experiment using cobalt dicarbollide at the Mayak plant showed excellent results with real high-level waste.

The OECD/NEA P&T status and assessment report is currently the most extensive database for P&T and comprises a critical analysis of the impact of P&T on the nuclear fuel cycle and waste management. Partitioning and transmutation can conceptually reduce the long-term radiotoxicity but it will require new fuel cycle plants of new dedicated reactors and an increased need for specialised intermediate storage plants. Geologic disposal structures for high active waste remain inevitable.

WELCOME ADDRESS

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Ladies and Gentlemen,

It is my pleasure to welcome you at our Belgian nuclear research centre in Mol for the Fifth International Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation organised by the Nuclear Energy Agency of OECD and the European Commission.

The concept of partitioning and transmutation is emerging since some years as a logical result of the application of a commonly accepted ethical principle.

Those who enjoyed the benefits of nuclear energy have to ensure that no risk will be transferred to future generations. In addition, a more developed fuel cycle might extract more energy per unit of initial mass of fuel, preserving as much as possible natural resources.

Each alternative that might help to optimise energy production and its associated waste management deserves to be fully considered by our research programmes.

Research on partitioning and transmutation is rather seductive to all of us. It requires new reprocessing techniques, new fuel developments, additional nuclear data, new reactors and irradiation facilities, new waste treatment and disposal concepts, and specific safety studies. The global nuclear scientific and engineering community is challenged by this opportunity.

Here at SCK•CEN we are also looking forward to fuel and materials testing in the BR2-reactor, code validations in VENUS and last but not least, to the design and the construction of a pilot Accelerator Driven System.

Everybody realises however that this voyage to the promised land will pass a desert with a lot of mountains and that we are not so sure that the horizon will be as bright as one can hope. On one hand, investments in reprocessing plants and fast reactors are going against the current of today’s nuclear policies in many countries. Moreover the return of those investments will only become apparent within several decades, by a reduction of an already small potential impact of disposed long-living waste. On the other hand, the demonstration of the technical feasibility of several theoretical options will still require important research activities and technological developments.

Successes of the P&T technology will only be possible, if the associated research will be supported by a systematic optimisation of the use of resources. National decisions on future fuel cycles will also require an international consensus on comparative risk assessments, allowing for instance an objective trade-off between potential climate changes and waste disposal risks.

In this context I want to congratulate the work of the P&T working group steering committee of NEA for the status and assessment report that will be presented at this information exchange meeting. Those international review activities will allow to consolidate gradually the conclusions of research and to draft a new menu of relevant work for the future.

Ladies and gentlemen, I am sure that your discussions here in Mol will allow you to make the point of the present status of actinide and fission product partitioning and transmutation and give the onset for the next step to internationally concerted actions.

I want to thank the organisers and all who will contribute to this challenging workshop.

I wish you a successful meeting and a pleasant stay in Mol.

OPENING REMARKS

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Ladies and Gentlemen,

It's a great privilege and pleasure to welcome you on behalf of NEA to the Fifth Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation. It's a particularly interesting occasion because we are able to present to you during this meeting the outcome of the systems study 'Status and Assessment Report on Actinide and Fission Product Partitioning and Transmutation', the result of fruitful work of the expert group.

Partitioning and transmutation has gained in interest during the past decade. As a result of the initiative taken by the Japanese government to launch a long-term research and development programme on the recycling and transmutation of actinides and long-lived fission products (called the OMEGA programme) the OECD-NEA was invited in 1988 to conduct an international project related to actinide separation and use. These information exchange meetings are one of the outcomes of this international project and intend to give the floor to current developments and discussion of issues in this very interesting field. Other incentives involve specialist meetings on particular scientific aspects as the NEA Nuclear Science Committee is conducting. From the outset of this programme, the involvement of and reporting to the NEA Radioactive Waste Management Committee was included, as the potential application of Partitioning and Transmutation would have impact on waste management. Here again, we have to emphasise that P&T would not at all replace the need for geological disposal for high-level waste but has potential to reduce the radiotoxic inventory there.

Four International Information Exchange Meetings have been held, differing in scope and conclusions as well as in attendance. The first meeting was held at Mito City (Japan) in November 1990. Various scientific and policy aspects of P&T were addressed and highlighted several disparate approaches which had been taken, covering a variety of aqueous and non-aqueous chemical procedures and a number of different reactor and accelerator based transmutation schemes. The second meeting took place in November 1992 at the Argonne National Laboratory (Illinois, USA). This meeting revealed a broader scope of discussed issues, including legal background, incentives and implications of P&T for the whole fuel cycle according to different nuclear policies and the need for guidance on the research and priorities. One of the main conclusions was that a comparison of systems studies in the field of P&T, some of them already in progress, should form the central part of the P&T activities under the Nuclear Development Committee of the OECD-NEA.

The third meeting was hosted by CEA in Cadarache in December 1994 and this meeting revealed the wide international interest in this subject and the great progress made throughout the OECD Member countries in understanding the implications of P&T. The fourth meeting was hosted by the Science and Technology Agency in combination with JAERI, PNC and CRIEPI and was held again in Mito City, Japan, in September 1996. This meeting could be summarised by the following:

- Goals of P&T were clearer: P&T would not replace geological disposal; potential hazard reduction was mainly associated with TRU elements; and reduction of the dose impact to man would come from mobile fission product radionuclides such as ^{129}I and ^{135}Cs.
- The main motivations for P&T were ethical reasons for the future generations and public claims concerning geological waste disposal sites.
- There was a need to better define: performance evaluation with criteria e.g. feasibility, credibility or safety gains; cut-off period; the best ways for industrial implementation; and reasonable level of extra costs.
- It would be necessary: to continue performing technical studies; to continue evaluating the results obtained; to continue with systems and strategic studies; and to continue with the necessary economical evaluations.

Today, a first phase in the assessment of P&T has been finalised and will be presented by Dr. Leon Baetslé this morning. The report "Status and Assessment Report on Actinide and Fission Product Partitioning and Transmutation", at your hand, highlights the results of a systems analysis on the complex issues involved in the P&T option. On this occasion I would like to address my special thanks and gratitude to the members of the expert group and to Dr. Leon Baetslé who has chaired this group in a remarkable way, resulting in a new publication which, I am sure, will be a standard reference in the coming years.

The conclusions of this report, although addressing the specific merits of P&T, would lead to new questions about the cost/benefit of the different highlighted partitioning and transmutation options. Especially transmutation in fast reactors and accelerator driven systems demand a comparative study, including the question of technical feasibility and projected performance of the latter. Both options will have their feedback on reprocessing needs and constraints and, in some respect, will be different in reprocessing impact.

The next phase, kindly sponsored by the Japanese government, will address these requirements, the feasibility and the reliability of Accelerator-Driven Systems as transmutation devices and this compared to fast reactors in principal. This second phase will again be co-ordinated by the Nuclear Development Committee and will start during the coming months. In addition, other studies as task forces under the umbrella of NSC will highlight the feasibility and reliability of high-power proton accelerators, the nuclear data needs at intermediate energies and calculation benchmarks on transmutation concepts, and the actinide separation chemistry which was published recently. Collaborative effort with IAEA will involve specific topics of P&T.

Dear colleagues, it's apparent that P&T is one of, if not the main issue in the coming decades. Increased reliance on electricity in an ever developing world, especially in the developing countries, will ask for readiness of production means, and most probably addressing specific demand of nuclear producing capacity if sustainability concerns, environmental maybe first, become more and more involved in policy questions. Nuclear will need to demonstrate its capacity and should at that moment be able to propose solutions to some remaining issues, with HLW-management being key issue. P&T addresses these sensitivities and our research and development should bring the answer to the questions posed by society. The role of OECD-NEA as an international organisation, is to conduct appropriate studies in these fields and to stimulate international collaboration, a practice which has been common within this P&T community from the earliest outset. In this context we encourage information exchange between the participants.

I thank you for all coming, and I particularly wish to thank Dr. Leon Baetslé and his colleagues of SCK•CEN, as our hosts who have, I am sure, laid the basis for a successful meeting. Finally, I would like to express my special gratitude to the Organising Committee members as well as to the European Commission for their collaboration to this meeting.

I wish you all an interesting, instructive and profitable three days.

IAEA'S ACTIVITIES IN P&T

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Abstract

The paper gives an overview of the IAEA's activities on Partitioning and Transmutation in the Division of Nuclear Power and the Division of Nuclear Fuel Cycle and Waste Technology, carried out since the information provided at the previous Information Exchange Meeting held in 1996.

Introduction

There is considerable interest among a number of Member states in the Partitioning and Transmutation (P&T) of long-lived radionuclides as a potential complement to the reference concept of the closed nuclear fuel cycle comprising: fuel fabrication, energy generation, intermediate storage of spent fuel, reprocessing, plutonium use in fuel and disposal of solidified high-level waste (HLW) in a deep geologic repository. This is the reason why P&T activities have been regularly dealt with in the IAEA Programme of Activities since the International Nuclear Fuel Cycle Evaluation (INFCE), conducted in the late seventies.

The IAEA reports on P&T published between 1982 and 1997 are listed at the end of this paper. Since the first OECD-NEA Information Exchange Meeting in 1990, the IAEA has been reporting on it’s P&T activities, which covered at that time mainly non-OECD countries. This paper gives an overview of these activities carried out in the Division of Nuclear Power and the Division of Nuclear Fuel Cycle and Waste Technology, since the previous Information Exchange Meeting held in 1996.

Early 1998, the Director General of the IAEA established a Senior Expert Group (SEG) to carry out an in-depth review of the main IAEA Programme of Activities. In its report to the DG, the SEG identified in the Programme for Nuclear Power and the Nuclear Fuel Cycle specific strategic objectives and recommended adjusting the Programme of Activities, which is expected to be done in 1999. Amongst others, it identified three specific strategic objectives on P&T:

- Further development and introduction of advanced power reactors with inherent safety and passive mechanism and of emerging nuclear energy systems (ADS).
- Potential and development of partitioning and transmutation techniques for high-level long-lived wastes.
- Potential, development and introduction of new proliferation resistant, actinides burning and transmutation fit fuel cycles.

IAEA activities

Status report on actinide and fission product transmutation studies

While intensive R&D programmes in the field of actinide and fission products transmutation are carried in OECD countries, there is a broader international interest in this area. Many non-OECD countries either already rely on nuclear power as a very important source of energy, or consider seriously plans for nuclear development as a way to meet the energy needs of their large and fast growing population (China, India). Two non-OECD states (Russia and China) possess nuclear weapons and face problems connected with the disposal of radwaste of defence nuclear facilities. These factors form an incentive for transmutation related research and at least four non-OECD countries have projects of different scale under way: Czech Republic, China, India, and the Russian Federation.

In view of this, the IAEA has taken an initiative to make a survey of the research activities in non-OECD countries upon recommendation by a Technical Committee Meeting (TCM) held in Vienna in 1994. The report was prepared by a TCM in September 1995 and published in 1997 (IAEA-TECDOC-948).

Accelerator-driven systems: energy generation and transmutation of nuclear waste (status report)

Participants of a Special Scientific Programme on “Use of High Energy Accelerators for Transmutation of Actinides and Power Production” held in Vienna in 1994, in conjunction with the 38th IAEA General Conference recommended the IAEA to prepare a status report on **Accelerator Driven Systems** (ADS). The general purpose of the status report was to provide an overview of ongoing development activities, different concepts being developed and their status, as well as typical development trends in this area and to evaluate the potential of this system for power production, Pu burning and transmutation of minor actinides and fission products. This document includes the individual contributions by the experts from six countries and two international organisations, as well as executive summaries in many different areas of the ADS technology. The document was published and distributed by the IAEA in 1997 (IAEA-TECDOC-985).

Co-ordinated Research Project (CRP) on the potential of Th-based fuel cycles to constrain Pu and to reduce long-term waste toxicities

This CRP started in 1996 with participation of China, Germany, India, Israel, Japan, Republic of Korea, the Netherlands, Russia and the USA with a purpose to examine the different fuel cycle options in which Pu can be recycled with Th to get rid of the Pu, or replace the Pu with materials that are less unacceptable to the public. The potential of the Th matrix will be examined through computer calculations. Each participant would choose his own fuel cycle, and the different cycles will be compared through certain predefined parameters (e.g. annual reduction in Pu inventory). As a final recommendation, the CRP could suggest the demonstration of Th-Pu burning in a reactor in one of the Member states. The toxicity accumulation and the transmutation potential of Th-based cycles for current, advanced and innovative nuclear power reactors including hybrid systems will be investigated.

To achieve the target of the CRP, it was decided to set up three stages. During the first stage, two benchmark exercises should be performed. Benchmark 1 would deal with calculation of the isotopic composition, cross-sections and fluxes for a typical PWR-cell loaded with $(Pu\text{-}Th)O_2$ fuel, as a function of the fuel burn-up. Benchmark 2 would deal with the same calculations, but for PWR lattice. Results of the benchmark exercises were discussed at the first Research Co-ordination Meeting (RCM) held in October 1996. Generally, the benchmark results of the participating countries showed good agreement regarding the destruction of Pu, and the isotopic composition of the remaining Pu. Some discrepancies appeared in the calculated neutron multiplication factor of the fuel, and in the build-up of minor actinides for high burn-up. The results of this stage were reported at the 9th International Conference on Emerging Nuclear Energy Systems (ICENES ‘98), held in Tel-Aviv, Israel, 28 June – 2 July 1998.

A critical review of the basic data and of the methods were performed during the second stage of the CRP. Participants of the CRP agreed that during the second stage of the CRP, three types of reactors should be investigated in terms of their potential to burn plutonium. The participants were divided into three groups, of which each group would investigate one reactor type. This procedure allowed a comparison of the computed results among several participants investigating the same type of reactor, and thus served as an additional benchmark exercise. The results (Calculations on Burning of Plutonium in LWR, HTR and MSR), obtained during the second stage of the CRP, were presented and analysed at the Second Research Co-ordination Meeting (RCM) held in Tel-Aviv, Israel, from 23-25 June 1998. Generally, the results showed a range of fuelling strategies on the basis of current and near-future-technologies.

For the third stage of the CRP, a Pu generating unit combined with a corresponding Pu burning unit was selected to assure a complete burning of the generated plutonium. A complete Pu-flow cycle will be considered. On the basis of these data, overall radio–toxicities of the combined system of PWRs and Pu burners will be compared with the U-fuelled PWR. The third RCM is planned in April 1999.

CRP on the use of Th-based fuel cycle in Accelerator Driven Systems (ADS) to Incinerate Pu and to Reduce Long-term Waste Toxicities

The purpose of the CRP is to assess the uncertainties of the calculated neutronic parameters of a simple model of Th or U fuelled ADS, in order to get a consensus on the calculation methods and associated nuclear data. Participants identified a number of issues which should be considered to get a better understanding of the ADS and agreed that some points, such as comparison of the different approaches and tools used by the different groups, should be reviewed at a later stage.

The first stage of the ADS benchmark was devoted to neutronics analysis of those ADS which are under development for nuclear energy production with a low long-term radioactivity of waste. As a background of this concept, Energy Amplifier (EA-ADS) simplified model was under inter-comparison.

The first RCM was held in March 1997 in Italy. The purpose of the meeting was to review the results of the first stage of ADS neutronic benchmark and to elaborate a programme for the second stage. Results of this RCM were reported to the IAEA TCM on Feasibility and Motivation for Hybrid Concepts for Nuclear Energy Generation and Transmutation held in Madrid, Spain, 17-19 September 1997.

During the next stage, an inter-comparison of the transmutation potential of ADS: transmutation rates of MA and LLFP, actinide and fission product inventories in a modular fast spectrum ADS, will be investigated. Results of this stage are very important in order to:

- Expand the benchmark option of the CRP to ADS new application.
- Cover the most essential activities (conducted now in many counties) to clarify the potential of ADS in transmutation of long-lived actinide and toxic fission products being produced by nuclear power in using the of U-Pu fuel cycle.
- Find an international consensus on the future role of ADS in a nuclear power scenario.

The second RCM will be held from 2-4 December 1998 in Petten, the Netherlands.

Technical committee meeting on feasibility and motivation for hybrid concepts for nuclear energy generation and transmutation

The purpose of this TCM was to assess the advantages and disadvantages of hybrid concepts for nuclear energy generation and transmutation of minor actinides and their potential role relative to the current nuclear power programmes and potential future direction to promote these concepts world-wide. The TCM was hosted by CIEMAT (Centro de Investigaciones Energeticas Medicamentales y Tecnologicas) and held at its headquarters in Madrid, Spain, on 17-19 September 1997.

Several major programmes/concepts on ADS development were presented:

- CERN Concept of ADS (C. Rubbia).
- OMEGA Programme & Neutron Science Project For Developing Accelerator Hybrid System at JAERI (T. Mukaiyama).
- Los Alamos ATW Programme (F. Venneri).
- Hybrid Systems For Nuclear Waste Transmutation Project In France (M. Salvatores).

The most salient observations resulting from the TCM were:

- Several accelerator systems and sources concepts can be developed for ADS.
- It is however important to have a very reliable source of neutrons coupled to the reactor system.
- The associated sub-critical reactor will likely be liquid lead-cooled (or lead/bismuth cooled), with efforts to use natural convection for lead circulation.
- Effort to develop neutronic benchmarks and codes for ADS should be pursued at the international level under the aegis of the Agency.
- Even if ADS is tentatively presented by some as a way to solve all nuclear waste issues, ADS is not an alternative to geological disposal. However, they have the potential to drastically reduce the waste toxicity, thanks to their capacity to burn minor actinides and fission products. As a reprocessing stage will be required, non-proliferation concerns should therefore be addressed.
- Further development of ADS requires the building of a demonstration device with a thermal power, in the 100-300 MW range. Efforts should be co-ordinated at international level on this matter.
- This pre-industrial test should provide input on the feasibility of the industrial deployment of ADS, including fuel cycle requirements, and a better understanding of the safety issues to be addressed.

Data-base of experimental facilities and computer codes for ADS related R&D

The needs for strengthening international co-operation in the field of the R&D for accelerator-driven systems was emphasised at several international forums:

- Scientific Programme on “Use of High Energy Accelerators for Transmutation of Actinides and Power Production”, Vienna, 21 September 1994 (in conjunction with the 38th IAEA General Conference).
- The Second International Conference on Accelerator-Driven Transmutation Technologies and Applications, Kalmar, Sweden, 3-7 June 1996.
- The 8th International Conference on Emerging Nuclear Energy Systems (ICENES’96), Obninsk, Russian Federation, 24-28 June 1996.

Consultants on Hybrid Concepts for Nuclear Energy Generation and Transmutation held in Vienna, on 16-17 December 1996 noted that:

- An increasing number of groups is entering this field of research.
- Many of these groups are not embedded in wider national activities.
- For these groups there is a need for co-ordinating their efforts and jointly funding projects as also for getting access to information from nationally or internationally co-ordinated activities.

Discussing organisational aspects of a possible IAEA involvement, consultants came to the conclusion that an effective co-ordination would necessitate the creation of an information document on existing and planned experimental facilities which can be used for ADS related R&D. The information would be provided by the Member states and distributed to all interested institutions. To facilitate this recommendation, two consultancies were organised in 1997 to work out and finalise a format of the document.

In June 1998, a draft of the database (DB) was distributed by the Agency to all contributors in the form of working material. A consultancy, was convened from 27-31 July 1998 in Moscow to review the draft.

The consultants made the following recommendations:

- The “hard” copy of the DB should be followed by an “electronic” version of the DB publicly accessible on the Internet or on CD-ROM.
- The IAEA should continue the ADS activities in CRPs and several possible topics for future CRPs have been proposed for consideration.
- The status on ADS should be regularly upgraded in an IAEA TECDOC, accessible to the general public and re-issued every 3-4 years.
- International meetings on ADS R&D need stronger international co-operation and harmonisation. There are already a large number of different international meetings and activities in this area. In this context, it is desirable to harmonise and closely co-ordinate the IAEA ADS activities with OECD-NEA.
- The IAEA co-operation in the preparation of the coming ADTT conference in Prague (June 1999) is highly appreciated.
- To establish an IAEA Advisory Group on ADS Concepts for Nuclear Energy Generation and Transmutation and Related Fuel Cycles for guidance on the above recommendations.

Co-ordinated Research Project (CRP) on Safety, Environmental and non-proliferation aspects of partitioning and transmutation of actinides and fission products

The overall objectives of the CRP are to reduce the long-term hazard arising from the disposal of high level waste. More specifically, the CRP aims to identify the critical nuclides to be considered in a P&T strategy, to quantify their radiological importance in a global nuclear fuel cycle analysis and to establish a priority list of radionuclides according the hazard definition.

During the first Research Co-ordination Meeting, held in October 1996, the Chief Scientific Investigators (CSI) made the following recommendations:

- The CSIs noted that the P&T studies initiated at OECD-NEA have up to now been focused on the development of the P&T processes within France and Japan and those countries (UK, USA) being interested in its evolution or which were engaged in these activities in the past. The OECD-NEA intends to make a system analysis study of the entire P&T field except for some very fundamental issues, e.g.: radionuclide toxicity ranking and influence of the geosphere on the dose-to-man generated by a radionuclide inventory. Keeping these principles in mind it is obvious that the IAEA with a world-wide scope in nuclear matters, will cover those aspects of P&T which are explicitly outside the OECD-NEA area (China, India, Korea, Republic of Russia) or which are not treated in the OECD-NEA system's analysis. In this context the detailed technological process analysis will not be studied within this CRP and the direct safety impact on workers and environment will be restricted to non-OECD countries. The contribution of the IAEA CRP on P&T will focus on the radiological impact of partitioning on the disposal issues.
- The CSIs pointed out that there are still no proposals to cover non-proliferation aspects of P&T processes and suggested to leave this subject out of the scope for the time being.
- The CSIs pointed out that in Russia in August 1996, the first stage of an industrial separation facility started operation at RT-1 in Chelyabinsk. This stage is based on the dicarbollide process and separates Cs and Sr from HLW. The second stage which is under design, will be based on a generic phosphine oxide process to separate actinides and Tc. The main aim of this separation facility is to simplify handling of HLW in Russia. However, the experience with this facility in the future could be useful for the investigation on P&T. This technology could be included in comparative analysis with TRUEX, DIDPA, DIAMEX and others. This facility could also be used to investigate operational safety aspects of partitioning facilities and secondary waste arising in a real process. Therefore, the CSIs proposed to include operational safety aspects in the Russian contribution.
- The CSIs recommended that the Republic of Korea should try to determine the required hazard reduction factor with and without P&T in addition to their proposed work which is relevant to the disposal of spent nuclear fuel.
- The CSIs proposed that in addition to its present work on quantification of radiological source terms and hazard reduction by partitioning of HLW with reference to PHWR fuel arisings, India should evaluate the radiological source terms of the critical nuclides generated in the thorium fuel cycle for their long-term hazard potential.
- The CSIs stressed that up to now the P&T community has considered the critical nuclides from the potential radiotoxicity point of view. In this respect, Pu and Am are the most important hazardous nuclides. However, taking into account the very low solubility of Pu and Am in disposal conditions, it is necessary to review the ranking and to investigate the risk of the nuclides for the biosphere and mankind. Other nuclides, e.g. ^{129}I, ^{237}Np, ^{99}Tc, ^{135}Cs, ^{93}Zr, ^{90}Sr and ^{137}Cs, will be also comparatively studied. In the framework of P&T, recycling of actinides implies an increase of the ^{238}Pu level in the fuel recycling processes and of ^{241}Am, ^{243}Am, ^{244}Cm in the MA recycling. The handling and treatment of industrial quantities puts the ^{238}Pu and ^{244}Cm at a much higher critical level than the Am isotopes in the safety assessment. The CSIs suggested that Belgium and UK should consider these aspects in their contribution to the CRP.

- The CSIs pointed out that the TRPO and DHDECMP-HDEHP processes for separation of MA from HLW are being investigated in China. It could be recommended to compare decontamination factors for both processes and their advantages and disadvantages and include these results in China's contribution to the CRP.

A second RCM is scheduled from 1-4 December 1998, where progress reports will be presented based on the proposals in the research agreements/contracts and the recommendations of the first meeting.

Conclusions

- The IAEA will continue to assist Member states in activities on partitioning and transmutation according to their needs.
- Adjustments in the IAEA programme of activities will be made based on the recommendations of the SEG.
- The IAEA will continue to co-ordinate activities with international organisations such as OECD-NEA and EC.

LIST OF IAEA PUBLICATIONS

[1] "*Evaluation of Actinide Partitioning and Transmutation*", Technical Report Series No. 214, Vienna, 1982, International Atomic Energy Agency.

[2] "*Feasibility of Separation and Utilization of Ruthenium, Rhodium and Palladium from High Level Wastes*", Technical Report Series No. 308, Vienna, 1989, International Atomic Energy Agency.

[3] "*Feasibility of Separation and Utilization of Caesium and Strontium from High Level Liquid Wastes*", Technical Report Series No. 356, Vienna, 1993, International Atomic Energy Agency.

[4] "*Use of Fast Reactors for Actinide Transmutation*", IAEA-TECDOC-693, Vienna, 1993, International Atomic Energy Agency.

[5] "*Safety and Environmental Aspects of Partitioning and Transmutation of Actinides and Fission Products*", IAEA-TECDOC-783, Vienna, 1995, International Atomic Energy Agency.

[6] "*Advanced Fuels With Reduced Actinide Generation*", IAEA-TECDOC-916, Vienna, 1996, International Atomic Energy Agency.

[7] "*Status Report on Actinide and Fission Product Transmutation Studies*", IAEA-TECDOC-948, Vienna, 1997, International Atomic Energy Agency.

[8] "*Accelerator Driven Systems: Energy Generation and Transmutation of Nuclear Waste*", IAEA-TECDOC-985, Vienna, 1997, International Atomic Energy Agency.

THE EU RESEARCH ACTIVITIES ON PARTITIONING AND TRANSMUTATION: FROM THE 4TH TO THE 5TH FRAMEWORK PROGRAMME

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Abstract

The European Commission is partly supporting research work on partitioning and transmutation of radioactive waste under the Fourth Framework Programme (1994-1998). This work includes nine research projects. Five strategy studies are evaluating the capabilities of various burners and fuel cycles to limit the production and even destroy the stock of actinides (plutonium and minor actinides). Two experimental projects are aiming at developing techniques for the chemical separation of actinides and two others are dealing with the investigation of transmutation of americium and long-lived fission products. The objectives of these studies are described together with the main results already obtained. The European Union should adopt the 5th Framework Programme (1998-2002) at the end of 1998. The broad lines of the research activities foreseen in partitioning and transmutation and future systems under the 5th Framework Programme are briefly presented.

Introduction

The European Union has launched in 1994 a specific programme on Nuclear Fission Safety in the Framework Programme for the European Atomic Energy Community (1994-1998). This specific programme covers research activities on reactor safety, radioactive waste management and disposal and radiation protection. It includes in particular research work on partitioning and transmutation (P&T) which is divided in three research tasks: strategy studies, partitioning techniques and transmutation techniques. Nine projects are covering these tasks with five in the field of strategy studies, two in partitioning techniques and two in transmutation techniques. They involve different European research organisations, which are partly funded by the European Commission. This paper presents the objectives of these projects and summarises the main results obtained so far.

The 5th Framework Programme (1998-2002) should be adopted by the European Union before the end of 1998. The specific programme for research and training in the field of Nuclear Energy includes activities on both controlled thermonuclear fusion and nuclear fission. In the field of nuclear fission, it is foreseen to increase the support to the research activities devoted to partitioning and transmutation and to open a new chapter on future systems. The future systems considered in the programme are in particular innovative or revisited concepts for reactors, fuels and fuel cycles. The content of the 5th Framework Programme and the broad lines of the activities on P&T and future systems are briefly presented in the paper.

Partitioning and Transmutation (P&T) under the 4th Euratom Framework Programme

The objectives of the nine research projects on P&T [1] are described together with the main results obtained so far [2,3]. These projects are five strategy studies on P&T (four shared-cost actions and one concerted action), two experimental investigations of partitioning techniques and two experimental studies of transmutation techniques. The strategy studies are aiming at assessing the capabilities of different burners and fuel cycles to limit the production or even destroy the stock of actinides (plutonium and minor actinides). The following order has been adopted to present the research projects in this section: first the both experimental studies of partitioning techniques, then three projects related to transmutation in reactors, followed by a study dealing with the thorium fuel cycle and finally three projects on accelerator-driven systems (ADS).

Experimental work on partitioning

New partitioning techniques (NEWPART)

CEA Marcoule is co-ordinating this project, which involves seven other partners: Reading University, Chalmers University in Göteborg, JRC-ITU Karlsruhe, ENEA Ispra, Univ. “Politecnico” Milano, FZK Karlsruhe and FZJ Jülich [4].

The objective of the project is to develop solvent extraction partitioning processes of minor actinides (mainly americium and curium) from the high level liquid waste (HLLW) generated by spent fuel reprocessing using the PUREX process. Because of the difficulty of the problem, the partitioning process is based on two extraction cycles. The first cycle consists in the co-extraction of actinides and lanthanides from the HLLW using malonamides as extractants (DIAMEX process). The second cycle aims at separating the actinides from the lanthanides. Several extraction systems are being investigated: (i) nitrogen polydentate ligands; (ii) sulphur bearing extractants; (iii) synergistic combination of (i) or (ii) with other chemical compounds.

Progress has been made in the comprehension of the basic chemical reactions between malonamide extractants and trivalent actinide and lanthanide nitrates. This could be achieved thanks to crystal structure determinations, X-ray absorption spectroscopy and molecular modelling. In addition, several flowsheets using different malonamides have been designed and tested to further develop the DIAMEX process. Active tests of these new flowsheets will be carried out shortly.

Concerning the separation between actinides and lanthanides, new nitrogen polydentate ligands have been synthesised and tested, which exhibit outstanding selective extraction properties for actinides. In particular, the pyridine-bis-1,2,4-tri-azines are able to selectively extract actinides from lanthanides in 1 to 2 molar nitric acid solutions. A synergistic mixture of newly synthesised sulphur bearing extractants and TOPO has enabled to obtain an effective separation between actinides and lanthanides in a 0.5 molar nitric acid solution during a counter-current test in a bank of centrifugal contactors [5].

The results obtained in the framework of this project suggest that it might be possible in a near future to develop a process with a single cycle allowing the direct extraction of the minor actinides from the very acidic HLLW.

Extraction and selective separation of long lived nuclides by fonctionalized macrocycles

This research project involves CEA Cadarache as co-ordinator and eight European universities (ECPM Strasbourg, Univ. Mainz, Univ. Parma, Univ. Twente, Univ. L. Pasteur in Strasbourg, Univ. Valencia, Univ. Liège, Univ. Autónoma in Madrid).

The objectives of the project are to synthesise and test macrocyclic compounds for the selective extraction of radionuclides with two purposes:

- Decategorisation of waste by selective removal of strontium and actinides.
- Separation of trivalent actinides from lanthanides.

Five of the universities have been synthesising a large number of extractants (mainly calixarene and bis-crown ether derivatives) for the removal of strontium, actinides and the separation between actinides and lanthanides. After testing these compounds, the most promising ones are described in what follows.

Strontium complexation and extraction experiments indicate that methyloxy and octyloxy calix[8]arene octa diethyl amide are good extractants for strontium from acidic medium with a high selectivity with respect to sodium.

Calix[4]arenes bearing acetamido phosphine moieties on the wide rim or “wide rim CMPO like calixarenes” have an excellent complexing and extracting ability with lanthanides and actinides in very acidic solutions, much larger than that of CMPO used at a concentration 250 times higher. In addition, the “wide rim CMPO like calixarenes” allow lanthanides to be discriminated and actinides to be separated from the heaviest lanthanides.

The crystal structure of two complexes of strontium with the p-tertbutylcalix[8]arene octaamide and the p-methoxycalix[8]arene has been determined by X-ray diffraction. It indicates that two strontium cations are inside the cavity of the calixarene. In addition, the behaviour in solution of several lanthanide and actinide complexes with different calixarenes has been studied by NMR.

Quantum mechanics calculations have been performed to understand the selective complexation of the cations with the extractants. In particular, the comparison between the interaction energies for several complexes of cations with alkyl (or phenyl) phosphine oxide indicates the crucial role of phenyl groups compared to alkyl groups, to enhance the interactions with lanthanides and to discriminate between them.

Transmutation in reactors

Evaluation of possible P&T strategies and of associated means to perform them

This project is co-ordinated by CEA Marcoule and involves ten other European laboratories (FZK Karlsruhe, AEA Technology Harwell, BNFL Risley, Belgonucléaire Brussels, SCK•CEN Mol, NRG Petten, JRC-ITU Karlsruhe, GRS Köln, ENEA Roma, Univ. "Politecnico" Milano).

The aim of this study is to give indications on what can be expected from partitioning and transmutation (P&T) strategies. The project includes five work packages: (WP1) global evaluation of different P&T scenarios and assessment of (WP2) partitioning techniques, (WP3) the feasibility of transmutation, (WP4) advanced fuel or target fabrication and (WP5) geological barrier efficiency.

The five scenarios investigated in WP1 are: (1) "once-through" cycle; (2) implementation of P&T with Highly Moderated Pressurised Water Reactors (HM-PWRs); (3) implementation of P&T with CAPRA type Fast Neutron Reactors (FNRs); when equilibrium is reached for scenario (3), it can be followed by either (4) a progressive introduction of EFR type FNRs or (5) a gradual decrease of nuclear power. Recycling of plutonium (Pu) and minor actinides is implemented after a 40-year period during which Pu is only recycled once in current MOX-fuelled PWRs. The total installed electric power remains fixed at 120 GWe.

The evaluation of the feasibility of transmutation indicates that HM-PWR cores could consume a significant amount of plutonium with a moderator-to-fuel ratio of about 4, but that the safety of such cores seems doubtful in view of the present requirements. However, the simulation of Scenario 2 shows an impossibility to recycle both plutonium and minor actinides with only HM-PWRs. In the case of CAPRA burners (Scenario 3), the envisaged recycling of actinides seems quite feasible from the reactor standpoint. The scenario evaluation confirmed that mining and milling and reactor operation were the main sources of short-term risk to the public in the fuel cycle. The risk is smaller for Scenario 3 than for Scenario 1, as the additional risk due to reprocessing operations is largely compensated by the decrease of mining activities.

In WP5, three geological host formations in which the waste resulting from the P&T scenarios would be disposed of have been evaluated on the basis of specific sites (Mol for clay, Gorleben for salt) and a generic site (granite). Normal evolution and human intrusion scenarios have been considered for geological disposal. It turns out that the waste from Scenarios 1, 3, 4 and 5 only have a minor effect on the normal evolution scenario, and that the doses are acceptable even in the case of direct disposal of spent fuel. To obtain a large reduction of the dose, long-lived fission products need to be transmuted, but this is difficult for technical and scientific reasons. The effects of P&T implementation on the consequences of the human intrusion scenario are only important if the efficiency of partitioning is very high and if it is possible to transmute most of the inventory of all actinides including curium [6].

Preliminary assessments of PUREX-based and pyro-metallurgical reprocessing techniques applied to CAPRA fuels were performed (WP2). For the PUREX process, the main issue is the dissolution of fuel containing high plutonium concentration in nitric acid for which experimental data are expected. Concerning the pyro-metallurgical process, developed at Argonne National Laboratory, the oxide-to-metal conversion required for its implementation seems possible, but the influence of the accumulation of fission products in molten salt during recycling needs further assessments.

Three processes for fabrication of fuels and targets containing minor actinides have been investigated in WP4. The MIMAS process (powder blending) could be used without significant difficulties for this purpose. The sol-gel precipitation process could be safer (it produces less dust) and better for recycling (more homogeneous U-Pu mixture). Impregnation techniques tested in the framework of the EFTTRA programme proved to be suitable for target fabrication (see below).

Supporting nuclear data for advanced MOX fuels

Belgonucléaire Brussels co-ordinates this study, which involves five other European partners (NRG Petten, SCK•CEN Mol, CEA Cadarache, JRC-ITU Karlsruhe, ENEA Bologna).

Its objective is to provide more accurate nuclear data for the P&T scenarios aiming at reducing the waste toxicity investigated in the strategy study described above. The use of advanced MOX fuels, either in PWRs or in FNRs, is studied. The necessary nuclear data working libraries are updated with new information from basic data evaluations and available integral experiments. The accuracy of the strategy studies to minimise wastes will be assessed at the end of the study.

Since the beginning of the project, a comprehensive compilation of irradiation results for high burn-up fuels has been carried out and experiments have been recalculated with modern computer codes and data sets.

The analysis of the composition of MOX fuels after irradiation at high burn-up (80 GWd/t in the highly moderated reactor BR3 in Mol and up to 46 GWd/t in the Saint Laurent B1 reactor) provides the experimental basis for PWRs. The values measured for the isotopic masses in the discharged fuel are being compared with the recalculated ones. Fairly good agreement is being found. In addition, the assessment of modelling errors has been completed.

In the case of the analysis of irradiation in FNRs, work has been progressing well. The comparison of computed and measured values for Phénix irradiations (PROFIL experiment) has been completed. Concerning the irradiations in KNK II in Karlsruhe, the chemical analysis of the fuel pins has been also completed and that of the single nuclide samples has started.

Joint EFTTRA experiment on americium transmutation

The objective of the project is to study the transmutation of ^{241}Am embedded in an inert matrix (spinel) in an irradiation experiment in the High Flux Reactor (HFR) at Petten. It includes the fabrication of a target pin, the execution of irradiation, post-irradiation examinations (non-destructive and destructive) and the interpretation of the results [7].

The work is being performed by the EFTTRA group, a collaboration of laboratories and research institutes in Europe (CEA and EDF in France, FZK in Germany, NRG in the Netherlands and ITU and IAM of the JRC of the European Commission).

The project is progressing according to plan. The irradiation of the target in HFR and the non-destructive post-irradiation examinations have been completed. The destructive examinations will be carried out by ITU soon.

The target contained 10 pellets of spinel (aluminium-magnesium oxide) with 11.5% in weight of americium oxide (about 0.7g of americium in total) in a titanium steel alloy cladding. It has been irradiated for 359 days in HFR, which represents a fluency of $1.7 \times 10^{26} m^{-2}$ for neutrons with an energy above 100 keV.

The first non-destructive post-irradiation examinations indicate that:

- the distribution of americium is not homogeneous inside the pellets; there is more americium at the edge of the pellet than inside; this is due to the impregnation technique used to fabricate the pellet;
- the pellets have swollen in volume by 15 to 18%, which is too much compared to a swelling of about 3% for uranium oxide; the swelling is caused by the alteration of the spinel matrix by the fission products.

The results of the calculations of the radial and axial transmutation effects can be summarised as follows:

- the flux profile is flat in the vertical direction;
- 92.3% of americium has been transmuted;
- the total content of actinides in the sample has decreased by 32%;
- the shielding effect leads to a higher transmutation rate for americium at the outer region of the pellets than at the inner region.

Thorium cycles as a nuclear waste management option

This project is co-ordinated by NRG Petten. Six other European laboratories are participating to this study: CNRS Orsay, Belgonucléaire Brussels, CEA Cadarache, FZJ Jülich, JRC-ITU Karlsruhe, ENEA Roma.

The goal of the project is an assessment of thorium cycles with a view to limit nuclear waste production and to burn waste, keeping in mind that thorium will be, in any case, a long-term option. The major fuel cycle steps have to be reviewed, focused on the European situation with thorium fuelled PWRs and FNRs as candidate reactors. The case of one thorium fuelled hybrid system is also investigated.

The six work packages of the project are covering the different steps of the thorium (Th) fuel cycle. The results obtained so far can be summarised as follows.

From the study of “Mining” (WP1), it appears that the thorium cycle leads to less mining waste production than the uranium cycle and that the waste could be disposed of in geological formations, which is not possible in practice for uranium waste. The final extracted thorium oxide product, however, has a higher gamma ray, alpha dose and radiotoxicity than uranium oxide.

“Fabrication of fresh thorium fuels” (WP2) does not lead to dose rates higher than those of conventional MOX fuel.

The study of “PWR (once-through) scenarios with Th/U fuel” (WP3.1) indicates that, compared to the reference low-enriched uranium scenario, the radiotoxicity is strongly reduced for the waste in all cases up to about 20 000 years, with best results for Th with highly enriched uranium at high burn up. For longer storage times, decay products of ^{233}U dominate. Recycling will be studied in the next phase. “Th-assisted (once-through) Pu-burning in PWRs” (WP3.2) is a very good option, since such cores can consume twice more TRU compared to U/Pu MOX. Recycling will be studied in the next phase. “Fast reactors with Th and full recycling of actinides” (WP3.3) offer excellent possibilities to burn Pu and to breed ^{233}U simultaneously. A self-sustaining core with Th/^{233}U is possible with very small void coefficient. Important waste radiotoxicity reductions are possible below 30 000 years. “Accelerator-driven fast reactors” (WP 3.4) loaded with Th/^{233}U are being studied; computational methods have been validated and nuclear data problems have been identified. Waste radiotoxicity with recycling of actinides will be studied in the next phase.

“Reprocessing” (WP4) using the single-cycle THOREX process can lead to U, Pu losses of 0.1% and to protactinium (Pa) losses of 1 to 2 %. High decontamination factors can also be reached for fission products. The pyrochemical method seems promising for Th fuel as well.

The “Residual risks of geological disposal” (WP5) were evaluated for once-through PWR scenarios with Th/^{233}U and Th/Pu fuels. The total dose of the actinides (most important ^{231}Pa) turned out to be rather small. In fact, long lived fission products (^{129}I, ^{135}Cs) gave the highest contribution, followed by activation products (^{59}Ni, ^{94}Nb). There was not much difference with U-based fuels.

Finally, a systematic approach has been developed to assess the “Technical aspects of non-proliferation” (WP6). Some concerns with respect to possible proliferation of ^{233}U in case of recycling and the misuse of accelerators were formulated.

Transmutation in accelerator-driven systems (ADS)

Impact of the accelerator-based technologies on nuclear fission safety (IABAT)

KTH Stockholm is co-ordinating this project, to which participate nine other European research institutions (CEA Cadarache, NRG Petten, FZJ Jülich, ENEA Roma, FZK Karlsruhe, JRC-ITU Karlsruhe, AEA Technology Harwell, Univ. Uppsala, Univ. Chalmers Göteborg).

The overall objective of the IABAT project is to make a European assessment of the possibilities of accelerator-driven hybrid reactor systems from the point of view of safe energy production, minimum waste production and transmutation capabilities. The work programme has three main items. The progress achieved in each of them is given in what follows.

In the first item “System and fuel cycle studies”, different ADS systems are investigated with a wide range of parameters like: neutron spectra varying from thermal to fast neutrons, fuel form from solid through suspension in liquid lead to molten salt solutions. A lead-bismuth spallation target has been optimised by performing thermal hydraulic calculations and making a pre-conceptual design. A number of code packages is being developed, e.g. a burn-up module for Monte Carlo codes and a kinetic code for liquid and solid fuels. In addition, some theoretical studies of ADS dynamics are carried out to interpret the reactivity control and to develop a methodology for subcriticality control. Finally, the radiotoxicity and proliferation resistance of different fuel cycles for ADS (thorium based plutonium burners, LWR waste incinerators and minor actinide incinerators) are being assessed.

Concerning the second item “Assessment of the technology and cost of accelerators”, the basic requirements for accelerators for transmutation (power and performance) have been formulated. Cost models for linear and circular accelerators have been developed. The linear accelerator cost model has been benchmarked against the European Spallation Source cost estimate. The cost model for circular accelerators will be completed shortly.

The third item deals with “Basic nuclear and material data”. First experiments on ^{232}Th fast fission yields and ^{233}U fission yields have been performed at OSIRIS mass-separator facility in Sweden. Models are validated with experimental data for nuclear reactions on ^{208}Pb, ^{54,56} Fe and ^{90}Zr induced by neutrons and protons. The creation of a cross-section library for neutrons and protons in the (0-150 MeV) energy range has started with data for $^{204, 206, 207, 208}$ Pb, ^{232} Th and ^{238}U.

Finally, it is worth mentioning that the analysis of different conceptual designs points in the direction of fast spectrum and liquid lead (or lead/bismuth) cooling for ADS.

Neutron driven nuclear transmutation by adiabatic resonance crossing (TARC)

This project is carried out by CERN as co-ordinator, Univ. Autónoma in Madrid, CNRS Grenoble and the Universities of Athens and Thessaloniki [8].

The main aim of this project is to develop both theoretically and experimentally a new method, the adiabatic resonance crossing (ARC), which enables to enhance strongly the capture rate of neutrons by the radionuclides to be incinerated. The peak cross sections for neutron energies corresponding to the resonance region are much larger than for the other neutron energies. For instance, the cross section of ^{99}Tc is 4 000 barn at the peak of a resonance at 5 eV, but it is only 20 barn at thermal energy. Access to the resonance region can be achieved by using a transparent medium like lead where the neutrons lose their energy in very small decrements. Then, the neutron energy decreases slowly through the resonances, leading to very large probability of capture during crossing. An experimental test has been carried out on ^{99}Tc with a neutron spallation source driven by the CERN proton synchrotron.

The project has 6 work packages:

- setting-up of the lead assembly and preparation and instrumentation of the beam line;
- experiments on ARC including timing experiments (CeF_3 counters) and activation experiments (delayed γ counting);
- study of advanced neutronics in lead by electronic experiments, activation measurements and temperature measurements;

- development of an appropriate formalism and computational tools for ARC;
- conceptual design of an incinerating device based on ARC;
- other applications of ARC.

The project has been completed by carrying out the following tasks. The detailed analysis of the data (work packages 2 and 3) has concentrated on the following items: (i) measurement of ^{99}Tc transmutation rate and of the ^{99}Tc neutron capture cross-section as a function of neutron energy with the CeF_3 detector; (ii) measurement of the production rate of ^{233}U from ^{232}Th and ^{239}Pu from ^{238}U using delayed γ spectroscopy; (iii) neutron flux measurements over the lead capture resonance region with ^{3}He chambers working in the ionisation mode; (iv) measurements of the high neutron energy flux (>1.4 MeV) with fission detectors; and (v) thermoluminescence measurements of the neutron flux below 0.5 eV. The computational tools that are indispensable for the interpretation of these data (work package 4) have been developed in parallel. The various detectors have been simulated and the results of simulation have been compared with experimental data in a systematic way. The present assessment of the experimental data confirms clearly the possibility of destroying in an effective fashion ^{99}Tc and ^{129}I by using the ARC method, as well as extending this concept to other applications.

Physical aspects of lead as a neutron producing target for the accelerator transmutation devices

The University of Louvain-la-Neuve is co-ordinating this concerted action, which involves twelve other European research institutions (Univ. Uppsala, Univ. Hannover, PTB Braunschweig, CEA Saclay, CNRS Nantes, Univ. Groningen, NRG Petten, GSI Darmstadt, FZJ Jülich, CNRS Caen, CNRS Orsay, Univ. Liège).

This concerted action started in the second half of 1998 for a two year duration. Its aim is to evaluate lead as a spallation target for the production of high intensity neutrons. The available experimental data are being compiled and compared to predictions from different theoretical models as well as the new measurements, which will be obtained in the EU laboratories in the next two years. The concerted action will be concluded by recommendations on possible additional measurements on lead and on the best codes to evaluate the reaction mechanisms.

The work programme includes the following items:

- Reactions induced by protons (30-70 MeV) and by neutrons (30-65 MeV);
- Production of residual nuclei by protons and by neutrons;
- Elastic scattering of 100 MeV neutrons on lead;
- Fission in proton induced reactions at 190 MeV on lead;
- Spallation induced by protons and deuterons at 0.8, 1.2 and 1.6 GeV;
- Fragmentation of ^{208}Pb ions of 1 GeV.A on hydrogen;
- Particle multiplicities on lead targets induced by protons from 0.8 to 2.5 GeV.

P&T and future systems under the 5th Euratom Framework Programme

The Commission adopted its proposal for the Fifth Framework Programme (1998 to 2002) in April 1997 (content) and in July 1997 (overall budget of 16 300 MECU including 1 467 MECU for Euratom research activities). After a first reading by the Parliament in December 1997, the proposal was amended by the Commission in January 1998. The Council reached unanimous political agreement on a common position concerning the Fifth Framework Programme (FP5) at its meeting on 12 February with an overall budget of 14 000 MECU, of which 1 260 MECU is allocated to the Euratom part of FP5. Because of the difference between the proposal of the Council and that of the Parliament (16 300 MECU as the Commission) for the overall budget of FP5, the conciliation procedure between both institutions started at the end of September to reach a final agreement on an overall budget of 14 960 MECU on 17 November (see Table 1). The adoption of FP5 is expected for December 1998.

Table 1

5th Framework Programme
Agreement between Council and Parliament on 17 November 1998

•	Quality of life and management of living resources	2 413	MECU
•	User-friendly information society	3 600	MECU
•	Competitive and sustainable growth	2 705	MECU
•	Environment	1 083	MECU
•	Energy	1 042	MECU
•	Confirming the international role of Community research	475	MECU
•	Innovation and encouragement of participation of SMEs	363	MECU
•	Human research potential and socio-economic knowledge	1 280	MECU
•	Activities to be carried out by means of direct actions (JRC)	739	MECU
•	***Euratom Framework Programme***	***1 260***	MECU
	Total	**14 960**	MECU

The specific programmes implementing the Fifth Framework Programme were proposed by the Commission in May 1998. The Council reached provisional agreements on the content of the specific programmes at its meetings on 22 June and 13 October. The final decision should be taken at the end of 1998. The work programmes are being prepared by the Commission. Targeted calls for proposals should be launched from the beginning of 1999.

There are two specific programmes implementing the Euratom part of FP5 [9]. The first one, named "Nuclear Energy", has two key actions, one on controlled thermonuclear fusion and one on nuclear fission. In addition, there are generic activities on radiological sciences and support for research infrastructures. The second specific programme is concerning the direct actions to be implemented by the Joint Research Centre. Table 2 shows the breakdown for the budget of the Euratom part of FP5 decided by the Council.

Table 2

EURATOM 5th Framework Programme *Decision by the Council*	
Key action Thermonuclear fusion	788MECU
Key action Nuclear fission	***142MECU***
Generic research on Radiological Sciences	***39 MECU***
Support for infrastructures	***10 MECU***
Joint Research Centre	281MECU
Total	**1 260 MECU**

The overall objectives of the key action on nuclear fission are to enhance the safety of Europe's nuclear facilities and to improve the competitiveness of Europe's nuclear industry. The more detailed aims are to ensure the protection of workers and the public from radiation, to assure a safe and effective management and disposal of radioactive waste, to explore more innovative concepts and to contribute to the maintenance of a high level of expertise in nuclear technology.

The key action on nuclear fission comprises four chapters: (i) operational safety of existing installations (ageing effects; severe accidents); (ii) safety of the fuel cycle (waste management and disposal, decommissioning; partitioning and transmutation); (iii) safety and efficiency of future systems and (iv) radiation protection.

In the area of partitioning and transmutation, strategy studies are foreseen to investigate its benefits and compare different methods such as critical and sub-critical systems taking into account the whole fuel cycle. New efficient and selective processes will be developed for the separation of the critical long-lived radionuclides from high level and medium level waste (e.g. hydro-metallurgical processes, chromatography and pyro-chemical processes). Basic nuclear data essential for transmutation and the development of ADS will be measured and computed. The radiation damage induced by spallation reactions in materials will be investigated. It is foreseen to develop and test fuels and targets for actinide and long-lived fission product incineration. The preliminary study of an ADS is also considered in the programme with supporting research work on sub-critical mock-ups, safety, coolants (liquid metal or gas), the confinement of the accelerator/reactor window and high power accelerators. Finally, new specific matrices could be also developed for the conditioning of long-lived radionuclides, which cannot be transmuted.

The future systems foreseen in the programme are innovative or revisited concepts. The objective is to assess new or previously discarded reactor concepts, that would be potentially cheaper, safer, more sustainable, producing less waste and reducing the risk of diversion (e.g. gas-cooled fast neutron reactors, high temperature reactors). Small reactors will be evaluated, identifying those of potential interest to the market beyond the next decade. The commercial potential of special applications of nuclear energy, such as combined heat and electricity production, desalination, hydrogen and methanol production will be investigated. Innovative fuels will be studied with advantages in terms of use of fissile materials, robustness to severe accidents and long-term storage. New fuel cycle concepts will be evaluated and developed (e.g. thorium fuel cycle and integral reactor with on-line reprocessing and fuel fabrication).

Conclusion

Under the 4th Framework Programme, the European Commission only devoted limited funding to research work on partitioning and transmutation. Nevertheless, about 40 European research laboratories are participating to these studies, which cover a wide range of scientific areas from partitioning techniques to transmutation in accelerator-driven systems. Most of the projects will be completed in the spring of 1999. Significant results have already been obtained: for instance, in the fields of partitioning of minor actinides by aqueous processes, strategy studies of P&T involving critical reactors and ADS, assessment of the thorium fuel cycle and transmutation experiments. These results could be used as an input for research projects in the next Framework Programme.

In the key action on nuclear fission of the 5th Framework Programme, a substantial part of the research work should be devoted to partitioning and transmutation and future systems. The 5th Framework Programme should be adopted by the European Union at the end of 1998. It is foreseen to make targeted calls for proposals from the beginning of 1999.

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RECENT TOPICS IN THE FIELD OF RADIOACTIVE WASTE MANAGEMENT IN JAPAN

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Abstract

In Japan, several organisations have carried out R&D of Partitioning and Transmutation (P&T) technology under the OMEGA programme (Options for Making Extra Gains from Actinides and fission products) which was initiated in 1988. In 1999, the Atomic Energy Commission of Japan (AEC) will check and review the outcome of the OMEGA programme up to now to decide how to proceed the P&T technology further.

The AEC established two committees for both technical and socio-economic aspects of geological disposal, and each of them materialised a report. The Advisory Committee on Nuclear Fuel Cycle Backend Policy, AEC, laid down the guidelines until 2000 on future research and development of the disposal of high-level radioactive waste (HLW), and the Special Committee on High-Level Radioactive Waste Disposal, AEC, published a report on the disposal of HLW discussed from social and economical points of view.

Status of Partitioning and Transmutation Study in Japan

P&T technology of long-lived radionuclides could convert HLW into useful resources and reduce the environmental impact associated with its disposal. In Japan, the Japan Atomic Energy Research Institute (JAERI), the Japan Nuclear Cycle Development Institute (JNC; former Power Reactor and Nuclear Fuel Development Corporation (PNC)), and the Central Research Institute of Electric Power Industry (CRIEPI), etc. have carried out the basic research and development activities of P&T technology under the OMEGA programme.

The OMEGA programme was initiated in 1988 in accordance with the national programme "Long-Term Program for Research, Development and Utilisation of Nuclear Energy" defined by AEC, aiming at efficient disposal of radioactive waste, effective use of natural resources and improvement of safety level of geological disposal. The OMEGA programme consists of the research and development subjects as follows:

- Partitioning:
  - Partitioning technology of high activity liquid wastes.
  - Recovery technology of useful metal from insoluble residue.
  - Effective utilisation technology of separated nuclides.
- Transmutation:
  - Transmutation technology by using reactor.
  - Transmutation technology by using accelerator.

In 1999, the AEC will check and review the results of the OMEGA programme, make clear the position of study on P&T technology within the HLW disposal policy at present, and revise the programme.

In addition, international collaboration with France and with Russia, concerning transmutation by using FBR, is now under way.

Current Activities for Radioactive Waste Management

Japanese basic policy regarding disposal of HLW is to solidify it into stabilised form, to store it for 30-50 years to be cooled, and to dispose of it deep to the underground (geological disposal). Japan is planning to set up an implementing entity for geological disposal around the year 2000, and thereafter we are looking to start operation of the repository between 2030 and the mid-2040s at the latest. In order to realise the plan harmoniously, the AEC has been discussing the technical and socio-economical aspects of HLW disposal.

The study of P&T technology is considered to provide the improvement option of geological disposal, not to be the replacement option of geological disposal. The results of the OMEGA programme must be checked and reviewed in accordance with these discussion for the basic policy of HLW disposal.

After the last (4th) International Information Exchange Meeting held in Mito in 1996, intensive discussions have been done concerning HLW disposal, and such efforts materialised two reports. Main recommendations and future plans described in these reports are summarised.

Technical Aspects

In April 1997, the Advisory Committee on Nuclear Fuel Cycle Backend Policy, AEC, laid down the guidelines on future research and development of the disposal of HLW. The report, entitled “Guidelines on Research and Development relating to Geological Disposal of High-Level Radioactive Waste in Japan”, states the basic technical issues relevant to geological disposal of HLW in Japan, the matters to be discussed in the second progress report, released by JNC, and the technically important issues which have to be covered by the second progress report.

In accordance with the report, JNC will release the report on the outcome of R&D activities to elucidate technological reliability of the geological disposal and to provide technical ground for selecting repository sites and for establishing safety requirement in the form of the second progress report before the year 2000, with the co-operation of related institutions, such as JAERI, CRIEPI, the Geological Survey of Japan (GSJ), the National Research Institute for Earth Science and Disaster Prevention (NIED), and university researchers. The first draft of the second progress report was made known to the public on September 1998, and the second one will follow around next spring, reviewed by experts in Japan. The second draft will be available in English for international reviews, and after that final version of the second progress report will be finalised before the year 2000.

In parallel with the R&D programme, there is also a plan in the national programme that the implementing organisation for HLW disposal will be established around the year 2000 to initiate the siting activities. The programme will then move from the generic into the site-specific phase. Development of site characterisation methodologies will become a more important issue as the national programme progresses.

Socio-economic Aspects

The Special Committee on High-Level Radioactive Waste Disposal, AEC, has considered various aspects of HLW disposal, including social and economic aspects, with a view to ensuring that such disposal will be accepted by the Japanese people in the coming century. Based on its discussions, the Special Committee released a report on how to implement HLW disposal in May 1998. In this report, the Committee considered the needs for discussing HLW disposal issues and the strategies for making information open to the public for their acceptance. And then, the Committee discussed an institutional arrangement on how to establish a reliable and transparent system for the HLW disposal project funding and the establishment of the entity to implement the disposal project and on how to make sure of the most appropriate siting process for harmonious co-existence between an eventual repository and host communities. So that, the Committee recommended various strategies for agencies concerned to enforce in particular. The Committee would like strongly to request those agencies concerned to take urgent steps to resolve the following four policy issues; (1) to secure HLW disposal project funds; (2) to set up the HLW disposal project-implementing entity; (3) to develop deep underground research laboratories; and (4) to prepare and establish the safety criteria and the guidelines on safety assessment of HLW disposal.

The report ,entitled “Basic Approach to the Disposal of High-Level Radioactive Waste”, consists of two parts. Part 1 discusses general considerations relating to the implementation of HLW disposal. Part 2 highlights four specific issues essential to further implement disposal of HLW: 1) how to promote public understanding of HLW disposal, 2) how to build public confidence in disposal technology and construct a financial and social supporting system for implementation, 3) how to coexist harmoniously with local communities at the disposal site and 4) how to proceed with site selection.

For the construction of the institutional system, to collect fund for disposal and the establishment of the implementing entity, Steering Committee on High-Level-Radioactive-Waste Project (SHP: the committee established by the Japanese government, JNC and the Federation of Electric Power Companies for preparing the establishment of implementing entity) is continuously investigating.

For the development of deep underground research laboratories, JNC has already started to the research programme in deep underground research laboratory at Mizunami city, Tono area, Gifu prefecture, in the central part of Japan as a crystalline rock site. JNC is also planning another deep underground research laboratory at Horonobe town, Hokkaido prefecture, in the north of Japan as a sedimentary rock site.

The report also states that “At present, the geological disposal of HLW is technically the most practical. The research on the volume reduction of HLW and utilisation of HLW as resources (P&T, etc.), aiming at safer and more efficient geological disposal, should be reviewed periodically, and the disposal system should be flexible to adopt outstanding progresses in these technologies, if any.”

NEWPART:
A EUROPEAN RESEARCH PROGRAMME
FOR MINOR ACTINIDE PARTITIONING

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Abstract

The programme NEWPART (1996-1999), was set-up in the framework of the 4PCRD European research programme "Nuclear Fission Safety" in order to develop solvent extraction partitioning processes of minor actinides, mainly americium (Am) and curium (Cm), which are contained in the high active raffinate generated during spent nuclear fuel reprocessing by the PUREX process. Owing to the difficulty of the separation problem, it was decided to design a partitioning process based on two extraction cycles: (i) the first cycle should separate the mixture of trivalent actinides (An(III)) and lanthanides (Ln(III)) fission products (FPs) from the bulk of the FPs. Malonamide extractants have been selected for this cycle, named DIAMEX process; (ii) the second cycle aims to separate selectively An(III) from Ln(III). Several extraction systems have been chosen for this goal. They include: (i) nitrogen polydendate ligands used alone or in synergistic mixtures with carboxylic acids; and (ii) sulphur bearing extractants (*di*-thiophosphinic acids) used alone or in synergistic combination with neutral oxygen bearing extractants. This lecture will briefly present the main achievements obtained in the framework in this research.

Introduction

In the framework of the 4PCRD "Nuclear Fission Safety" European Programme, the NEWPART research contract (1996-1999) was set-up for the definition of processes for partitioning minor actinides (MAs) contained within nuclear waste mixtures generated after reprocessing of spent nuclear fuels by the PUREX process.

The main guides for the definition of partitioning methods for MAs within NEWPART are the following: (i) the minor actinide target for partitioning are americium (Am) and curium (Cm). Neptunium is not fully considered for partitioning within NEWPART (the partitioning of this element is considered feasible by an adapted version of the PUREX process); (ii) the waste mixture to be treated is the aqueous high active raffinate (HAR) from the reprocessing of spent nuclear fuel by the PUREX process; (iii) essentially no (or only minor) modification of the composition of HAR is needed prior to implement the partitioning process. This point is a prerequisite to prevent the formation of disturbing radioactive precipitates; (iv) Am and Cm will be separated from the complex mixture of nuclear wastes (essentially the fission products, FPs) without modification of their stable oxidation state (i.e. M(III)) in aqueous nitric acid medium; (v) liquid-liquid extraction will be the preferred method of partitioning selected, owing to its potentially high efficiency for the decontamination of HAR in MAs and for the purification of the recovered MAs; and (vi) as far as possible, all the chemicals to be used, such as extractants, diluents and aqueous complexing agents, must be destructible, at the end of their uses in the process, into gases that can be released freely into the atmosphere. In this case, no secondary solid waste is expected to be generated from the partitioning operations.

The selective separation of Am and Cm from the HAR is a complex problem owing to: (i) the high concentration of nitric acid (~3 mol/L) of this effluent; and (ii) the presence of high concentrations of trivalent lanthanide FPs (molar ratio Ln/An is about 40 for the UOx1-type fuel). So, the main strategy selected in NEWPART relies in the development of a two step process: (i) the first step aims to separate the mixture of An(III)+Ln(III) from 2/3 of the FPs and from the bulk of nitric acid; and (ii) the second step is intended for the An(III)/Ln(III) group separation by selective extraction of An(III), which is the most difficult goal to achieve. To perform the first step, the DIAMEX process, based on the use of a malonamide extractant was selected, while for the second step several selective systems are under study. They include: (i) synergistic mixtures of α-substituted carboxylic acid and nitrogen polydendate ligands, like substituted oligopyridines, *tri*-pyridine-*tri*-azines, amino-*di*-pyridine-*tri*-azines, pyridine-*bis*-1,2,4-*tri*-azoles; (ii) pyridine-*bis*-1,2,4-*tri*azines, and (iii) synergistic mixtures of substituted *di*-thiophosphinic acids (CYANEX 301 like) and oxygen neutral donor ligands.

The most important results obtained so far during the fundamental and process development studies will be highlighted in this lecture.

Results and discussion

DIAMEX process

Malonamide extractants, with the formula: $(RR'NCO)_2$ CHR'', where R, R' are alkyl, cyclohexano or aryl groups and R'' is alkyl or oxy-alkyl groups, were selected for the design of the first An(III)+Ln(III) extraction cycle. Several types of work have been carried out: (i) synthesis of malonamide extractants, performed mainly at Reading and by small industrial companies; and (ii) basic research involving malonamides solvent extraction mechanism, including thermodynamic and kinetic aspects, co-ordination chemistry of Am(III) and Ln(III) with malonamide ligands. This work involves, in particular, crystal structure determination and X-ray absorption spectroscopy (XAS) related to M(III) (M = An or Ln) malonamide complexes in solution, molecular modelling, DIAMEX process development, including tests of the process with synthetic spiked effluent and genuine HAR.

Fundamental studies

Extraction thermodynamics, mechanisms and kinetics

One of the major questions to be solved for the extraction of lanthanide (III) and actinide (III) from aqueous nitrate solutions by malonamide extractants concerns the knowledge of the nature of the extracted species. Is there a dual mechanism of extraction i.e. formation of solvates with formulae $M(NO_3)_3L_n$, (with M(III) = Ln(III) or An(III) and L = diamide *bi*-dendate extractant), for low aqueous nitric acid concentration and alkali nitrate solutions (e.g $LiNO_3$), and formation of ion-pairs, of formula $[LH^+]_n$, $[M(NO_3)_{3+n}]^{n-}$, for concentrated aqueous nitric acid solutions, as encountered when extracting U(VI) or actinide (IV) from nitric acid media by monoamide extractants? Several experiments, involving UV-visible and ^{13}C NMR spectroscopies and solvent extraction, have been conducted to answer this question. From the data obtained so-far one can conclude that, if a dual extraction mechanism exists, the second mechanism does not seem to be an ion-pair mechanism involving a protonated diamide. So, one can conclude that the occurrence of an ion-pair mechanism is unlikely.

The M(III) extraction reaction by malonamides from aqueous nitrate solutions can thus be written as follows:

$$M^{3+}_{aq} + 3\ NO^-_{3\ aq} + n\ L_{org} \Leftrightarrow M(NO_3)_3L_{n\ org} \qquad (1)$$

n is usually 2 for most of the malonamides.

Several diamides have been compared in their ability to extract An(III) or Ln(III) from aqueous nitrate media. For a series of malonamides with different R’ groups (butyl, phenyl and chloro-phenyl), it was shown at Chalmers that the less basic is the malonamide the better its M(III) nitrate extracting properties. At Marcoule, new malonamides have been also studied that contain either function(s) in the central R’’ group. These malonamides, especially the *di*-methyl-*di*-octyl-hexylethoxy-malonamide, DMDOHEMA, exhibit better affinities for M(III) nitrates in comparison with malonamides with R’’ alkyl groups. DMDOHEMA has been selected to replace *di*-methyl-*di*-butyl-*tetra*-decylmalonamide, DMDBTDMA, in a new version of the DIAMEX process (see below).

A comprehensive study of the extraction kinetics of Ln(III) and An(III) nitrates by DMDBTDMA have been carried out at Marcoule (M. Dal Don Ph.D. thesis). The main conclusions of this work can be summarised as follows:

- Using a constant area stirred cell of the Lewis type, evidence of chemically limited M(III) nitrate extraction rates was obtained from the knowledge of the values of the Arrhenius activation energies, which, for all M(III) studied, were found to be close to 40 kJ/mol (while for a diffusion limited kinetic regime, this energy is generally found close to be 20 kJ/mol).

- M(III) extraction rate laws were established for Ln(III) and An(III). All of them exhibit the same form: rate : k [M(III)] [L]$[NO_3^-]^3$.
- Noticeable differences exist in the values of k for the series of Ln(III) ions. Experimental data can be sorted out in several sections; light, medium and heavy Ln. Kinetic constant k for An(III) (An= Am and Cm) was found between those of the light and middle Ln for Am, and close to that of Eu for Cm.
- Evidence was obtained for the location of the chemical limiting reaction at the interphase. This was made clear, in particular, when studying the effect of surfactants on the Nd(III) extraction kinetics; decrease of the Nd(III) extraction rate was observed with both cationic or anionic surfactants.
- Extraction kinetics of M(III) nitrates by DMDBTDMA were found much slower than extraction of U(VI) or Pu(IV) nitrates by tri-n-butylphosphate, TBP (extractant of the PUREX process). The relative slowness of the M(III) extraction rates by malonamides may have an influence on the definition of the DIAMEX process implementing conditions.

Structure determination for M(III) nitrate malonamide solvates

Numerous highly valuable structural informations have been obtained by crystal structure determination and by X-ray absorption spectroscopy.

Crystal structures related to the following complexes have been obtained at Reading: $M(NO_3)_3L_n$ with Nd/1 DMDPhMA, Yb/1 DMDPhMA, Nd/ 2 DMDCHMA, Nd/ 2 BDMDPHMA, Yb/ 1 BRTEMA stoichiometries (with D = di; M = methyl; Ph = phenyl; CH = cyclohexyl, E = ethyl; MA = malonamide and BR = bromo). Moreover, crystal structure of BRTEMA was also determined. Various Ln(III) co-ordination polyhedra were thus evidenced.

Experiments aiming to determine and compare M(III) ion co-ordination polyhedra in crystals and solutions have been carried out using X-ray absorption spectroscopy. Experiments were done at two synchrotron facilities, LURE (Orsay, France) and Daresbury (GB). For example, $Nd(NO_3)_3L_n$ crystals or solutions in ethanol were studied for L = *tetra*-ethylmalonamide, TEMA, *di*-methyl-*di*-phenylmalonamide, DMDPhMA, *di*-methyl-*di*-butyl-ethoxyethylmalonamide, DMDBEEMA, and DMDBTDMA. For all these systems, Nd(III) co-ordination polyhedron seem to be the same with about 10 to 11 oxygen atoms at 2.55 ± 0.01 A. It is important to note that no evidence of involvement of ether oxygen in the bonding with Nd (III) was observed for DMDBEEMA. While very interesting, these XAS data demonstrate the difficulty to precise the co-ordination modes of Ln(III) nitrate solvates with malonamides.

Molecular modelling

Molecular modelling studies have been conducted at Reading aiming:

- To compare calculated structures and X-ray determined crystal structures.
- To propose structural explanations to experimental differences observed during extraction of M(III) metallic nitrates by several malonamides.

Using Quanta/CHARM package, it was found that the lowest conformation calculated structures for DMDPhMA, DMDCHMA and BDMDPhMA, were similar to the experimentally determined crystal structures. Differences exist between structures of DMDPhMA and BDMDPhMA and that of DMDCHMA, which were confirmed by calculations. So, an important conclusion was drawn from that study; the differences in M(III) extraction efficiency between cyclohexano (DMDCHMA) and phenyl substituted (DMDPhMA and BDMDPhMA) malonamides can be correlated with the difference of the preferred conformations of the malonamide extractants.

Using Gaussian 94 programme, protonation of cyclohexano (DMDCHMA) and phenyl (DMDPhMA) substituted malonamides was studied. Results are equivalent for both malonamides and show that *mono*-protonated malonamide contains an intramolecular hydrogen bond, while the *di*-protonated malonamide does not.

Quantitative Structure-Activity Relationships (QSAR) study related to the extraction of Nd(III) nitrate by a set of 17 malonamides was carried out in 1998 at Marcoule in collaboration with Reading. For each diamide in Zg-Z conformation a molecular dynamics, *in vacuo*, at 500 K for 200 ps has been made. Then, a conformation has been selected from the trajectory by QSAR techniques and minimised with AM1 semi-empirical method (MOPAC). The main result of this QSAR lies in the presence of an oxygen ether atom in the R’’ substituent which confers to the corresponding malonamides better M(III) nitrate extracting properties.

DIAMEX process development

Reference DIAMEX process

The reference DIAMEX process is based on the use of DMDBTDMA in solution in TPH at a concentration equal to 0.5 mol/L.

The radiolysis and hydrolysis of DMDBTDMA were studied in parallel at the CEA/Marcoule with that of an other malonamide; the *di*-methyl-*di*-butyl-dodecylethoxymalonamide, DMDBDDEMA, which differs from the former malonamide by the presence of an ether function in 2 position within R’’ group.

The main results obtained so-far can be summarised as follows:

- Radiolysis:
  - Degradation of malonamides is observed under radiolysis.
  - The rate of degradation is accelerated slightly by the presence of an aliphatic diluent and more importantly by that of an aqueous nitric acid solution.
  - Main radiolysis products consist mainly in amide-carboxylic acid and long chain carboxylic acids.
  - Even if degradation of the diamides occurs under a radiation field, their rates, while larger than that of the TBP, are not so important.

  So, we can conclude that diamides seem to resist radiolysis sufficiently so that one can consider their use in a real process for HLLW treatment.

- Hydrolysis:
  - Hydrolysis of DMDBTDMA was found limited.
  - Hydrolytic degradation of DMDBTDMA increases with temperature.
  - Modification of the nitric acid concentration in the aqueous phase does not seem to have any influence on the DMDBTDMA degradation rate.
  - The main hydrolytic products of DMDBTDMA are an amide-acid compound and a monoamide.

Next studies to be carried out in this field will concern the regeneration of the degraded solvents.

Reference DIAMEX process flowsheets have been designed for tests performed at the ITU using centrifugal contactors with real HAR. To prevent the co-extraction of Mo and Zr FPs, oxalic acid was introduced into the feed and the scrubbing solution. Several cold tests were conducted and two hot tests were performed. Most of the goals assigned to this test were reached: almost quantitative extraction of Am, and Ln; very limited extraction of most of the FPs, including Zr and Mo; and limited extraction of ruthenium. It should be noted however that some other FPs were found to be extracted. They included technetium and palladium. Solutions to prevent contamination of the purified An(III)+Ln(III) fraction will be defined in the future.

Different DIAMEX processes using DMDBTDMA or DMDOHEMA

ENEA/Polimi (Italy) developed a new version of the DIAMEX process based on the reference DMDBTDMA molecule, but for a 1 mol/L diamide solution in TPH, instead of 0.5 mol/L DMDBTDMA solution for the reference DIAMEX process. The HAR is also different; it corresponds to an UOx-2-type used nuclear fuel, whereas the fuel considered is of an UOx-1-type for the reference DIAMEX flowsheet.

The modification of the concentration of the extractant within the solvent may have at least two major advantages in:

- Minimising the occurrence of third phase problems.
- The ability to treat feeds with higher nitric acid concentrations than that of the reference DIAMEX process.

The above mentioned advantages were also observed for the flowsheet developed at CEA/Marcoule using DMDOHEMA but for a lower extractant concentration (0.65 mol/L instead of 1 mol/L for DMDBTDMA). The flowsheets designed were, like the reference DIAMEX flowsheet, based in particular on the use of oxalic acid to prevent the co-extraction of Zr and Mo fission products with the mixture An(III)+Ln(III).

ENEA/Polimi flowsheet using 1 mol/L DMDBTDMA was tested successfully in mixer-settlers in collaboration with G. Modolo and R. Odoj at Jülich on synthetic spiked solutions. DMDOHEMA flowsheet was tested at CEA/Marcoule on cold synthetic solutions. In both cases the tests were highly successful. Most of the goals were reached without noticeable problems. Computer codes of these processes were established and calculated solute concentration profiles along the cascade of extractors correspond fairly well with experimental data.

One can conclude that several possibilities exist for the selection of a DIAMEX process, either based on the reference diamide DMDBTDMA, with possible several extractant concentrations, or on DMDOHEMA optimised malonamide. Progress should be made in the future to solve the small remaining problems that have been highlighted during the hot tests at the ITU.

Group separation of actinides (III) from lanthanides (III)

Fundamental studies

Heterocyclic nitrogen compounds

Extraction thermodynamics with various molecules

Work was done in this field at Chalmers/Göteborg, at CEA/Marcoule and FZK/Karlsruhe.

An(III)/Ln(III) selective extraction of the An(III) using mixtures of α-substituted carboxylic acids (HA) with nitrogen polydendate ligands (L) were studied. The nitrogen ligands used were from different origins:

- Prepared at Reading for most of the ligands used at Chalmers, i.e. polypyridines, amino-*di*-pyridyl-*tri*-azine (ADPTZ) derivatives.
- Prepared at Reading and obtained from French sources (PANCHIM and University laboratories) for the CEA, i.e. polypyridines, ADPTZ, pyridyl-pyrimidine, etc.
- Prepared at the laboratory for FZK work, i.e. for pyridine-*bis*-1,2,4-triazoles.

For HA+L mixtures, most of the experiments conducted were aiming to determine the M(III) ion extraction mechanism(s) and to expect to obtain good selectivity for the extraction of An(III) over Ln(III) for sufficiently high nitric acid aqueous feed.

The method used to determine the M(III) ion extraction mechanism(s) was generally the slope analysis method which consists in studying the effect of the variation of one chemical parameter on the distribution ratios and separation factors of the metal ions at trace levels. The determination of such extractions mechanism(s) is complicated by the following facts:

- Interaction between HA and L occurs in the organic phases.
- Protonation of L, inducing its transfer into the aqueous phase, is observed for low pH aqueous solutions.

The following preliminary conclusions can be drawn from these studies:

- Polydendate nitrogen ligands L are effective for inducing a selectivity in the separation of An(III) vs. Ln(III) when mixed in synergistic combination with a-substituted carboxylic acids.
- Substituted terpyridines are generally less effective than terpyridine itself, when one considers the DM(III).

- Substituted terpyridines are less effective than tri-tertio-butyl-pyridine-tri-azine, TtBPTZ, studied previously, in terms of DM(III), while SFAm/Eu are almost the same for both systems (≅ 10).
- Quaterpyridine are more effective than terpyridine for both DM(III) and SFAm/Eu values. This seems to be connected with the denticity of the ligand.
- ADPTZ derivatives, easy to lipophilise in comparison with TPTZ, were found to be almost as good as TPTZ derivatives. This is in particular the case for tri-methyl-hexanoyl-amino-di-pyridyl-tri-azine, BADPTZ.
- Extracted complexes are of the form MA3Ln.
- Reasonably good SFAm/Eu were obtained; 10 (terpyridines and ADPTZ), 20 (quaterpyridine), 10 to 20 (bis-1,2,4-triazole derivatives).
- None of these systems is able to perform a selective extraction of Am(III) over Eu(III) from a high nitric acid concentration aqueous solution, but BADPTZ in synergistic combination with a-cyanodecanoic acid in solution in a diluent made of TPH + a modifier was considered to be suitable for process development (see below). In this case the feed containing An(III)+ Ln(III) will contain 0.2 mol/L of nitric acid, which is a reasonably high acid concentration.

Other nitrogen polydendate ligands were studied by Z. Kolarik at FZK. This is the case, in particular, for the 3-(2-pyridyl)-5-(4-*ter*-butyl-2-pyridyl)-1,2,4-*tri*-azole (TpTP). That nitrogen polydendate ligand was found to be a cationic exchanger, which exhibits very interesting selective extraction properties for Am(III) over Eu(III). $SF_{Am/Eu}$ equal to 60 was observed, but unfortunately for pH = 5.7 for the feed aqueous solution. Nevertheless, this very unusual molecule seems very interesting to study.

Moreover, An(III)/Ln(III) group separation was also studied by Z. Kolarik *et al.* at Karlsruhe with home-made pyridine-bis-1,24-triazines derivatives (BTPs). These molecules have interesting properties. For example, they are able to extract selectively An(III) from Ln(III) with separation factor as high as $SF_{Am/Eu} = 140$ and this for high aqueous nitric acid concentration. An(III) extraction proceeds through a solvation mechanism involving three molecules of BTP, as shown in the following reaction:

$$An^{3+}_{aq} + 3\ NO^-_{3\ aq} + 3\ L_{org} \Leftrightarrow AnL_3(NO_3)_{3\ org} \quad (2)$$

with L = BTP.

These properties of BTPs extractants correspond potentially to a breakthrough in the field of An(III)/Ln(III) group separation. Syntheses of new BTPs were carried out at CEA/Marcoule, and their M(III) extraction mechanisms were also studied. Research will be devoted in the future for the study of these very interesting extractants.

Structural determinations and molecular modelling of the complexes

Numerous crystal structures involving both pure new polydendate nitrogen ligands L and their solvates with Ln (III) salts have been determined at Reading.

Among them, one can cite:

- The structure of the solid TPTZ and of its Sm(III) nitrate monosolvate. For the first time, the tridendate character of the bond between the TPTZ and one Ln(III) ion was established.

- The structure of the protonated form of the 2-amino-4,6-*di*-(2-pyridyl)-1,3,5-*tri*-azine (ADPTZ).
- The structure of solvates formed between the ADPTZ and most of the Ln(III) nitrate salts. Variations of the crystal structures were observed with an increase of the atomic number of the Ln, leading to very interesting considerations about the complexation of these M(III) metal ions with these terdendate nitrogen ligands.
- The structure of ion-pairs type formed between protonated forms of the terpyridine and Ln(III) nitrato complex anions.
- Structures of solvates formed between Ln(III) nitrates and pyridine-*bis*-1,2,4-*tri*-azoles derivatives were determined. With an increase with Ln atomic number the structural types of the crystals change. So, a systematic was carried out with respect to structures along the Ln series.

All these structures are very helpful for the design of extractant molecules.

Molecular modelling of these systems were carried out both at Reading and CEA-Marcoule. We will mention only few studies done in the field of terpyridine conformations studied *in vacuo* or in a water box, alone or in the presence of protons or M(III) metal ions. The main results obtained in this field can be summarised as follows:

- The *trans-trans* conformation of terpy is preferred *in vacuo.*
- The presence with terpy *in vacuo* of M(III) ion, induces a reorganisation of the terpy molecules which are now *cis-cis*, i.e. they exhibit a potential *tri*-dendate binding site.
- In a water box, terpy exhibits a *cis-cis* conformation because water molecules (1 or 2) are bound to the nitrogen atoms. One can conclude that water is able to pre-organise the terpy which is a highly favourable phenomenon for subsequent ion binding.
- In a water box, $M(terpy)(NO_3)_3$ complexes were found to be unstable. Hydrolysis of the complexes occur leading to the complete dissociation of the M(terpy) moieties. This has been demonstrated by molecular dynamic simulations.
- On the contrary, complexes of the type $M(terpy)A_3$, with A = α-bromocaprate anion, were found to be stable in a water box. This important observation is in agreement with the observed extraction efficiencies of M(III) in the corresponding experimental systems.

Sulphur bearing cation exchange extractants

Following the discovery by Zhu (Beijing, China) in 1995 of the highly efficient properties of the purified CYANEX 301, a commercial *di*-thiophosphinic acid, two partners of the NEWPART collaboration decided to work in this field. The first study was carried out at FZJ/Jülich, which involved:

- Confirmation of Zhu’s results and extension of the work in creating new CYANEX 301 like extractants with improved properties in comparison with the original molecule.
- Extension of the work towards process development, which was done in collaboration with ITU/Karlsruhe.

The second study which was carried out by CEA/ Marcoule in collaboration with PNC Japanese scientists concerns:

- The verification of the Zhu's results.
- The determination of the mechanism for the synergistic extraction of Am (III) and Eu(III) by CYANEX 301+ TBP synergistic mixtures.

The main results obtained in this important field can be summarised as follows:

- It was confirmed at FZJ/Jülich and CEA/Marcoule, that the results obtained by Zhu are real. $SF_{Am/Eu}$ as high as 6000 were obtained.
- The solvates formed when M(III) ions are extracted by the purified CYANEX 301 possess the following formula: MA_3HA, with M = Am(III) or Eu(III) and HA standing for the CYANEX 301 molecule. The corresponding extraction constants were determined. They are in agreement with those published by Zhu in 1996
- Extraction mechanisms of Am(III) and Eu(III) by a CYANEX 301 + TBP synergistic mixture were determined. The composition of the extracted complexes are different for Am(III) and Eu(III). Although a slight increase of the $SF_{Am/Eu}$ is observed for low TBP/CYANEX 301 ratios, further increase of this ratio leads to an important decrease of this separation factor.
- New CYANEX 301 like extractants with electron withdrawing substituents, such as phenyl, chlorophenyl, fluorophenyl or tolyl, exhibit, in synergistic combination with neutral organophosphorous extractant, like TBP, *tri*-butylphosphine oxide TBPO, *tri*-octylphosphine oxide TOPO, a high and selective affinity for Am(III) over Ln(III). This holds even for high aqueous nitric acid concentrations, i.e. 0. 5 to 2 mol/L. These results represent an important breakthrough in the field of An(III)/Ln(III) separation. This work was done by G. Modolo and R. Odoj at Jülich.

Process developments

Nitrogen polydendate extractants

Several systems involving nitrogen polydendate extractants were considered for An(III)/Ln(III) group separation process development. The common goal of all these systems is to extract selectively the An(III) from a feed with a reasonably high concentration of nitric acid (i.e. at least higher than 0.15 mol/L), leaving the bulk of the Ln(III) in the aqueous raffinate. Up to now the most advanced system considered is based on the mixture of BADPTZ and α-cyanodecanoic acid in solution in TPH in the presence of a modifier. Development work has been done to design the flowsheet which will be operated for a feed with a 0.2 mol/L nitric acid concentration, and corresponding to a An(III)+Ln(III) solution generated by the DIAMEX process. The flowsheet has been defined by P. Baron at CEA/Marcoule for runs which will be carried out in a bank of 16 miniature centrifugal-contactors installed in a hot-cell at the ITU/Karlsruhe. Hot tests are foreseen to be carried out in December 1998 or in the early beginning of 1999.

Owing to the extraordinary properties of the BTPs and of the very interesting properties of the *bis*-1,2,4-*tri*azoles derivatives synthesised by Z. Kolarik *et al.* in consultation with Reading at KZK/Karlsruhe, processes for An(III)/Ln(III) group separation will be also developed by

CEA/Marcoule and ENEA/Polimi and tested in the near future. At the moment, the research concerns the scaling-up of the synthetic procedure to be able to have sufficient quantities of extractants to run the processes in centrifugal-contactor loops. Results in this field are expected in 1999.

Substituted di-thiophosphinic extractants

Experiments have been conducted at FZJ/Jülich, and also at the ITU/Karlsruhe, in order to define an An(III)/Ln(III) separation process based on the use of the outstanding properties of the *di*-thiophosphinic extractants.

The main results obtained so far can be summarised as follows:

- At Jülich:
  - The radiolytic degradation of the CYANEX 301 has been studied. It was found that the instability of the extractant was not extensive, but the radiolytic degradation products have a detrimental effect on the $SF_{Am/Ln}$. Consequently, this point is of great importance for process design.
  - The radiolytic degradation of the substituted phenyl *di*-thiophosphinic acids was studied. It was found that their radiolytic instability was lower than that exhibited by the CYANEX 301 itself. These molecules can thus be considered as promising candidates for the design of an An(III)/Ln(III) separation process.
  - An(III)/Ln(III) separation test was made on synthetic spiked solution using a synergistic mixture made of *bis*-chlorophenyl-*di*-thiophosphinic acid and TOPO. The flowsheet was defined in collaboration with P. Baron from CEA/Marcoule and the test was performed using a bank of 8 stages of centrifugal contactors. Good separation performances were obtained. This represents a very encouraging result because the feed was 0.5 mol/L in nitric acid. One may observe that no such performances have been ever published to date.
- At ITU (in collaboration with FZJ/Jülich)
  - Tests of an An(III)/Ln(III) separation process based on the use of CYANEX 301 in synergistic combination with TBP was carried out with a genuine liquid waste. Highly successful results were obtained for purified CYANEX 301+TBP mixture; in only three stages, the effluent was decontaminated vs An(III) (Am and Cm) by a factor higher than 10 and the Am+Cm fraction was decontaminated from the Ln (III); DFAn/Ln > 400 was reached. These first results are very encouraging.

Conclusions

Considerable progress have been made in the field of minor actinide partitioning during the NEWPART research programme. Among the main achievements one can cite:

- DIAMEX process — Definite progresses have been made in the understanding of the basic chemistry of malonamide extractants in their ability to react with trivalent actinide and lanthanide nitrates. This was achieved with the systematic use of crystal structure determinations, X-ray absorption spectroscopy and molecular modelling. As far as the DIAMEX process development is concerned, one can mention that we extended the choice for a good process by designing three flowsheets with the first reference malonamide extractant, DMDBTDMA, and by optimising the formula of a new malonamide, the DMDOHEMA, suitable for the design of an improved DIAMEX process flowsheet. Active tests of theses new processes are foreseen in the near future.

- An(III)/Ln(III) group separation — Here also the results obtained are quite impressive. New, very interesting nitrogen polydendate ligands were prepared and studied which exhibit far better properties than those corresponding to the previously known ligands of this type. In particular, the most important results correspond to the pyridine-*bis*-1,2,4-*tri*-azines prepared by Z. Kolarik at FZK. These molecules are able to selectively extract An(III) nitrates from Ln(III) and this for a 1 to 2 mol/L nitric acid feed. Very interesting results were also obtained in this field with *bis*-chlorophenyl-*di*-thiophosphinic acid and TOPO mixture. Good separation between An(III) and Ln(III) were obtained for a 0.5 mol/L nitric acid feed during the implementation of a counter-current test carried out with a bank of centrifugal contactors with synthetic spiked solution.

In conclusion, we can say that we are confident with the fact that the processes under study can be developed to reach the goals required for modern efficient industrial minor actinide partitioning processes.

Acknowledgements

This work was done in the framework of the E.C. 4th PCRD research programme under the contract FI41-CT-96-0010. CEA also received support from Cogéma and EDF. Special thanks are given to the numerous scientists belonging to the eight different organisations listed above who carried the experimental research.

R&D STRATEGY FOR PARTITIONING & TRANSMUTATION UNDER OMEGA PROGRAMME AND NEUTRON SCIENCE PROJECT OF JAERI

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Abstract

JAERI developed the double-strata fuel cycle concept and JAERI's OMEGA activities have been devoted for developing technologies which are needed to realise this concept. In this concept, partitioning is based on aqueous separation of high level liquid waste from the reprocessing plant and a dedicated system with a very hard neutron spectrum is used for transmutation of minor actinides. Accelerator-driven subcritical system(ADS) and Actinide Burner Reactor(ABR) are designed. The system design of these systems is based on nitride fuel. Technology of nitride fuel fabrication and dry reprocessing of spent nitride fuel are under development. For 1000MWth-ADS plant, a proton accelerator of about 1GeV, 40mA will be needed for subcritical core of k_{eff} of 0.95 for the support factor of 12 to 14. To develop an accelerator hybrid system, an intense proton accelerator is needed for various experiments and also to demonstrate the high power accelerator technology. JAERI started the development of a super-conducting proton linac. In the course of developing of an accelerator for transmutation experiments, we recognised that neutron scattering community desires to have very high intensity neutron beam, two order of magnitude stronger than that of existing ones. Also, JAERI has been one of major neutron suppliers for basic science community in Japan with its research reactor JRR-3M, and is asked to continue to be so in the 21st century.

Neutron scattering has achieved some notable successes in recent years, such as unravelling the crystal structures of high-temperature superconductors, and is now exciting a lot of interest among biologists for probing large organic molecules. A limiting factor for neutron scattering research is the intensity of the neutron beams. High intensity of neutron beams allows researchers to carry out experiments that would otherwise be impossible.

Under these circumstances, JAERI has launched the Neutron Science Project. The objective of the project is to construct a 5MW spallation neutron source which consists of a high intensity proton accelerator with proton energy of 1.5 GeV, proton beam current 5.3mA (8 MW), a 5MW mercury spallation target and research facility complex for basic science such as neutron scattering and for nuclear technology. The R&D on accelerator-driven transmutation is performed under both the Neutron Science Project and the OMEGA programme.

The paper discusses the strategy for developing ADS and the recent R&D achievements of P&T under the Neutron Science Project and the OMEGA programme.

ADVANCED OPTIONS FOR TRANSMUTATION STRATEGIES

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Abstract

Advanced options for transmutation strategies currently investigated in France are presented. Accelerator driven systems are the major item. In this field, the French National Initiative GEDEON is presented together with some relevant technical results, both in the experimental and in the scenario study fields.

Introduction

In the present paper, we will shortly recall that the motivation for hybrid system development at CEA and in France is related to the waste management optimisation by advanced options.

In this context, we will make a short presentation of the national programme ("Groupement de Recherche") GEDEON.

An example of the role of advanced options for transmutation will be given, in terms of possible scenarios, according to the "double strata" fuel cycle approach.

The physics of transmutation and role of ADS

Accelerator Driven Systems (ADS) can find a role in:

a) Energy production (widely discussed by C. Rubbia in the frame of the EA proposal).

b) Symbiosis with standard reactors (PWRs with UOX and MOX fuel, Fast Reactors with MOX fuel), in order to manage minor actinides (Np, Am, Cm) and selected Long Lived Fission Products (LLFP) in a separate "stratum" of the fuel cycle.

c) Symbiosis with standard reactors (PWRs with UOX fuel), in order to manage Pu, Minor actinides and LLFP.

These scenarios have been described in Reference [1], and the decisive role of fast neutron spectra has been underlined.

In the "transmutation" scenarios (b and c), when compared to the corresponding critical reactor, an ADS has the following features:

a) *Same* transmutation rates for actinides (since proportional to the power in the core).

b) Some extra-neutron availability to transmute LLFP (a neutron consuming process).

c) Subcriticality can allow to have a core with a very small fraction of delayed neutrons (case of the dedicated cores of scenario b), or a core with a significant reactivity loss/cycle (scenario c), possibly within a deterministic safety approach.

These arguments motivate the research in the field of ADS, and require further detailed intercomparison of critical and subcritical cores, to build-up a convincing case and a reasonable framework for future R and D.

The GEDEON Initiative

In order to co-ordinate several activities in the ADS domain, the "Groupement de Recherche" GEDEON has been created.

GEDEON is a meeting point for CEA and CNRS researchers, proposed by EDF in 1995, on ADS and thorium studies. Recently it has been extended to Framatome, and several other institutes and industries have applied to join it.

A network for basic research has been set up with no strong financial commitments but with more than 100 scientists involved.

Research programmes are discussed and, for some, carried out in common e.g.:

a) SATURNE programmes for spallation physics (ended in 1997).
b) Subcritical studies at MASURCA at CEA-Cadarache.
c) Material studies (in particular for lead alloys).

The motivation for setting up GEDEON can be summarised as follows:

a) The CEA is in charge of R&D in nuclear waste incineration and up to now carried it out mostly in the frame of critical reactors, with some exploratory studies in the ADS field.
b) The CNRS is traditionally not involved in such studies, but shares with CEA an expertise in nuclear and accelerator physics.
c) The EDF has a medium term policy with respect to spent fuels management and has an open scientific watching activity as far as new options for the back-end of the fuel cycle.
d) Finally, Framatome proposes his know how to be used in the framework of new reactor development (e.g. HTR), and in case a hybrid test reactor should be decided.

The focus of GEDEON is on following scientific issues:

- Spallation target physics
  - Neutron production from heavy targets.
  - Residues measurement (heat, radiotoxicity, corrosion).
  - Theoretical efforts to qualify intra- and inter-nuclear cascade codes.

- Nuclear data acquisition
  - Special cross sections, differential or integral (measurements at Geel LINAC, or irradiations in power reactors), for minor actinides, long-lived fission products, lead and bismuth.
  - Extending neutron cross-section libraries from 20 to 200 MeV.

- New material studies
  - Window : radiation damage in n and p fields.
  - Pb, Pb/Bi : corrosion, embrittlement, irradiation effects.

- Subcritical neutronics studies with the Masurca reactor at Cadarache (see Paragraph 4)

- Scenarios studies and system studies (demo experiment, targets etc.), including the assessment of the role of pyroprocesses (see Paragraph 5)

- IPHI project between CEA and CNRS for High Power Proton Accelerator development
  - An injector of 10 MeV × 100 mA (first stage) is presently considered.

Activities of GEDEON

The main activities of GEDEON have been devoted to experimental studies in the field of spallation physics, with experiments performed at the SATURNE accelerator (thin and thick targets; neutron production and double differential cross-section measurements for several materials and several proton energies) and at GSI-Darmstadt (inverse kinematics experiments for spallation residual measurement).

The MUSE experiments at MASURCA have been a major undertaking between CEA and CNRS, to investigate the neutronics of subcritical cores. Three experiments have been performed: MUSE-1 and 2 (References [2] and [3]), where a ^{252}Cf spontaneous fission neutron source was used, and MUSE-3 (see Figure 1) where a 14 MeV neutron generator was used to provide the external source, in pulsed and continuous mode.

The future configuration MUSE-4 (1999) will use a new accelerator (GENEPI, see Figure 2) developed at ISN-Grenoble, which will allow both neutrons from (d,t) and (d,d) reactions, with high neutron intensity (up to 10^{10} n/s) and sharp pulses (~ 1 μsec). This will enable to perform dynamic measurements of the reactivity, besides the measurement of the neutron source importance, core subcriticality, reaction rate distributions, etc.

Finally, in the frame of GEDEON, workshops are organised. Two of these workshops were organised in 1998:

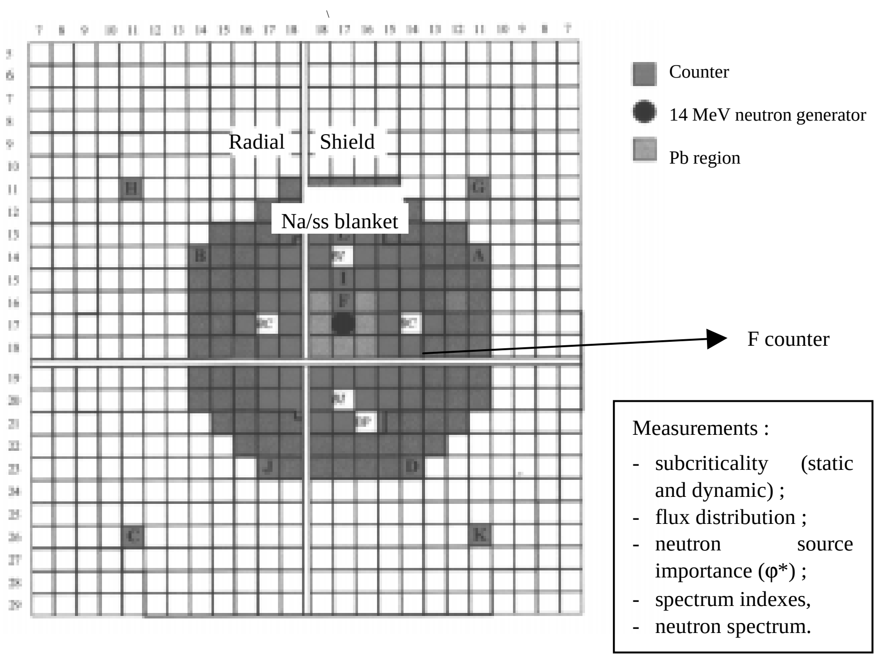
- Role of pyrochemistry for advanced options in waste management.
- Nuclear data needs and experiments (for minor actinides, long-lived fission products, lead and bismuth).

Figure 1. **MUSE3 configuration (1998)**

Core composition :
50 % fuel (75 % U)
(25 % Pu)
50 % Na

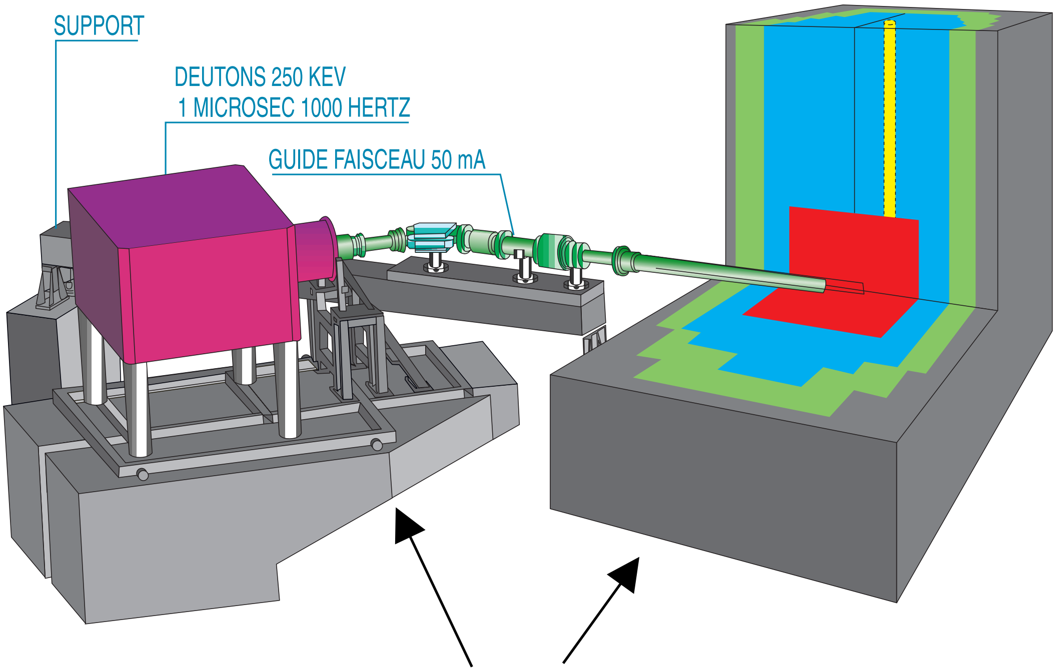
Blanket :
50 % Na
50 % Steel

Radial Shield :
100 % Steel



Measurements :
- subcriticality (static and dynamic) ;
- flux distribution ;
- neutron source importance (φ*) ;
- spectrum indexes,
- neutron spectrum.

Figure 2. **A new development: the GENEPI installation developed at CNRS, ISN-Grenoble for neutron generation by (D,T) and (D,D) reactions (early 1999)**

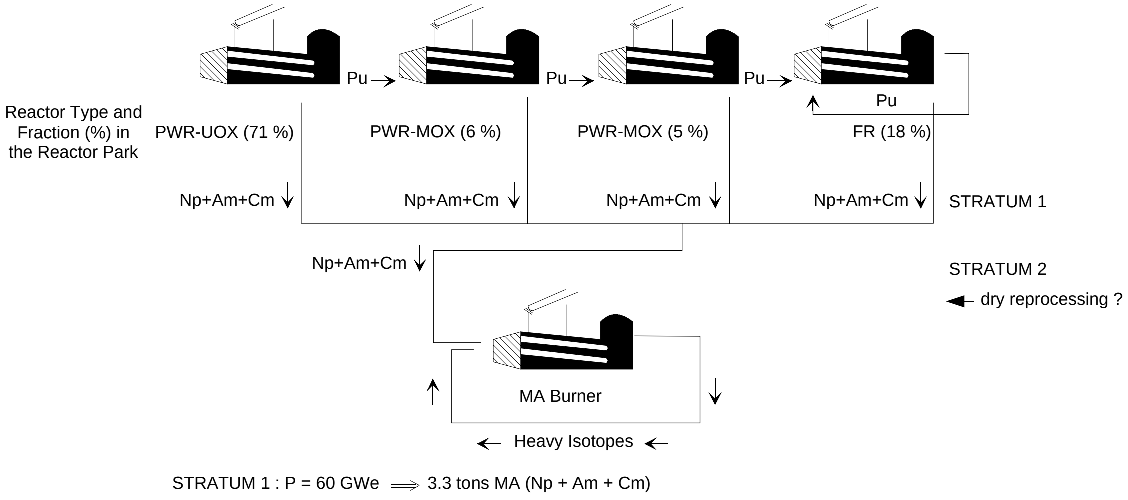


Scenario studies

The “double strata” approach, initially introduced at JAERI [4], has been developed in the frame of the studies described above. In Reference [5], a first quantitative assessment was given of the size of the separated stratum equipped with dedicated reactors, in terms of the share of the total power produced in the park.

The following scenario, with Np, Am, and Cm transmutation was obtained.

Figure 3. **Double strata fuel cycle**



This reactor park allows to reduce waste potential radiotoxicity by a factor of 100 with respect to a reactor park operated in open cycle and producing the same amount of energy.

The limit is due to the level of losses during reprocessing (here assumed to be 1% for all MA, and 0.1% for U and Pu). If dry reprocessing is used, a compact dedicated fuel cycle can be envisaged.

In order to specify the characteristics of the dedicated burner cores, critical and subcritical (ADS) dedicated cores have been specified in preliminary studies (Reference [6]). They have the following characteristics:

	Critical	**Subcritical (ADS)**
Power	130 MWt	130 MWt
Neutron spectrum	fast	fast
Coolant	Pb/Bi	Pb/Bi
Fuel	Pu + MA	Pu + MA
MA burning rate	- 78 kg/TWhe	-99 kg/TWhe
β_{eff}	~ 170 pcm	–
Target/beam	-	Pb/Bi target - Beam : E_p = 1 GeV - I_p = 1 mA (1.6 mA at EOC)

Versions of the same cores, but with gas cooling are under study.

If Long Lived Fission Products (LLFP) (^{129}I, ^{99}Tc) are introduced in “moderated flux” S/A at the periphery of the core (LSD concept see Figure 3), the following transmutation rates are obtained:

^{99}Tc	-30 kg/TWhe
^{129}I	-18 kg/TWhe

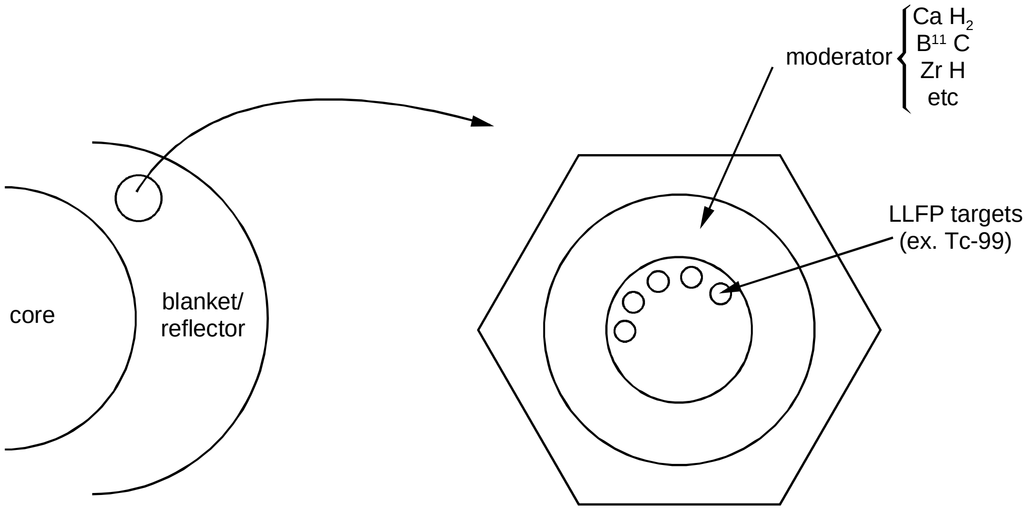
both for critical or subcritical (ADS) cores.

When these core are used, the following double strata scenario can be obtained:

- First fuel cycle stratum, with PWR (UOX) and both PWR (MOX) and CAPRA-type fast reactors to manage Pu.
- “Dedicated” second stratum, with the dedicated burners of Table I.

The characteristics of these scenarios (with critical or ADS in the second stratum) are given in Figures 4 and 5.

Example. **Reaction rate of ^{99}Tc**



Transmutation in a "moderated" S/A

The neutron surplus available leaking out of the core, is used in the blanket/reflector.

These neutrons are slowed down to the required energy ("spectrum tailoring") using moderator (not-absorbing) materials in the specific S/A.

Figure 3. **"Leakage with Slowing Down" (LSD) concept for LLFP transmutation**

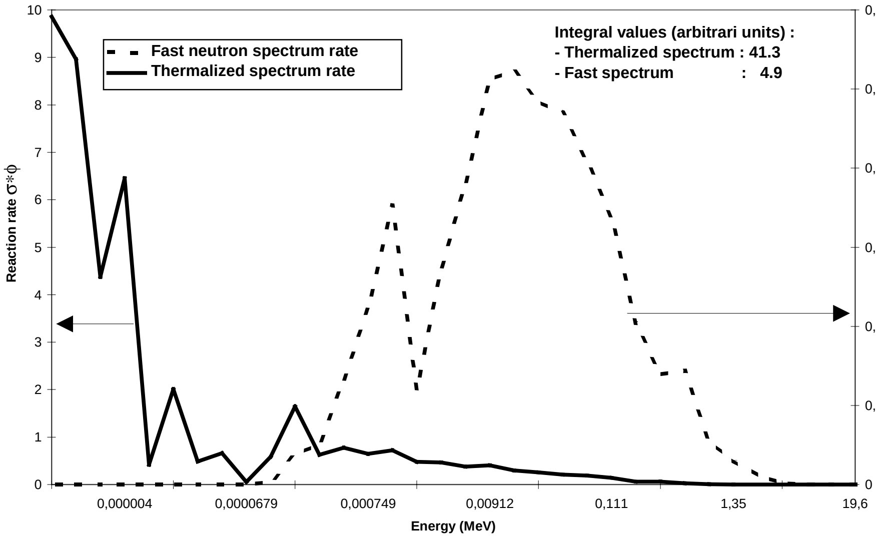


Figure 4

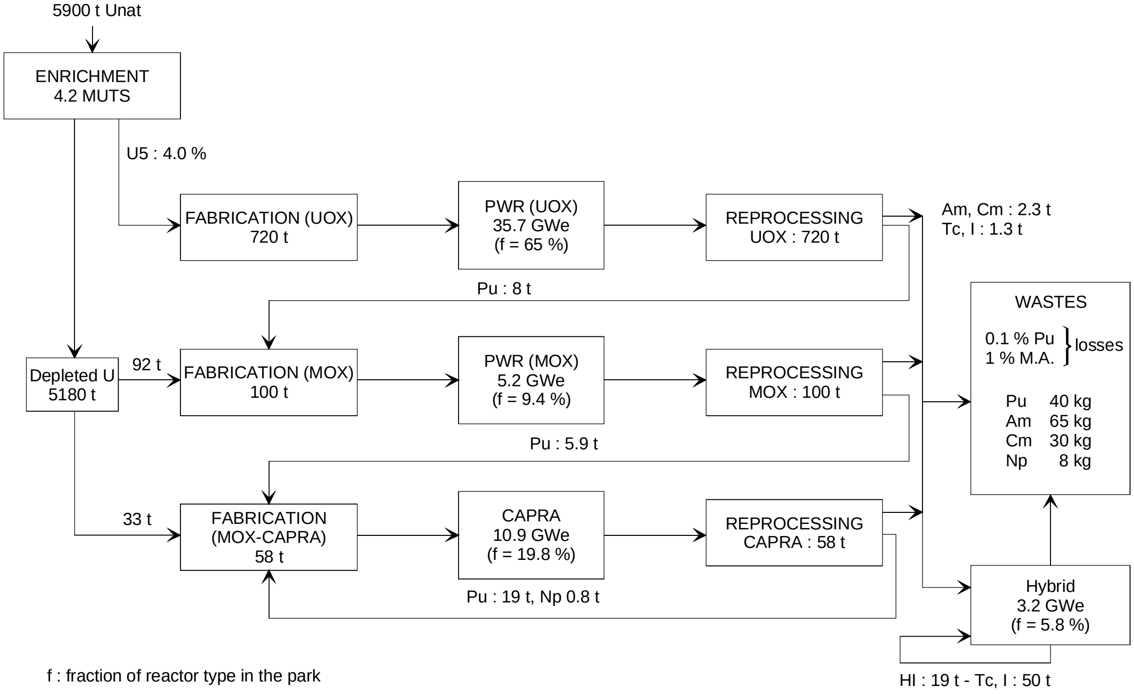
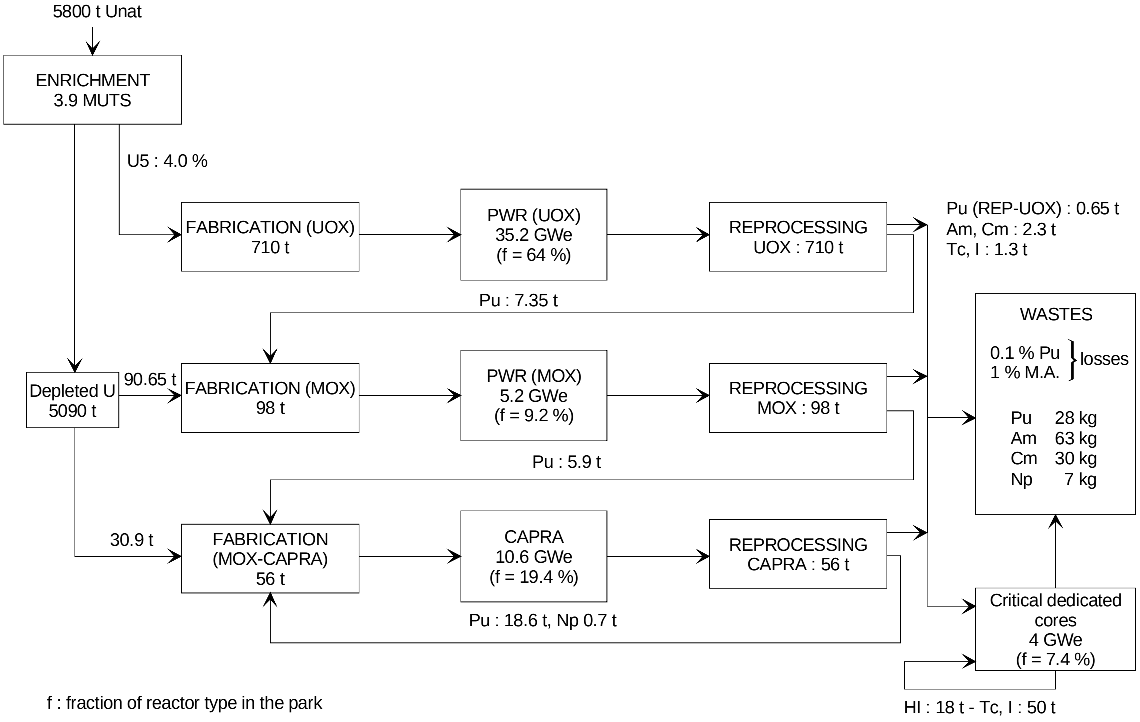


Figure 5



Conclusion

Advanced options for transmutation strategies are actively investigated in France. The ADS is a major field of R and D, at present performed mostly in the frame of the GEDEON initiative.

As far as CEA, these activities are part of the wider SPIN programme which is supported and co-financed by the industrial partners, EDF/Cogéma/Framatome, according to the 1991 Law on Waste management.

Finally, the construction of an eventual "demo" ADS of limited power and the performance of the associated R and D, is presently discussed by France together with Italy and Spain (technical group under the leadership of Prof. C. Rubbia).

A wider European initiative is expected in the very near future.

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R&D ACTIVITIES ON PARTITIONING IN RUSSIA

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Abstract

A concept of closed nuclear fuel cycle (NFC) accepted in Russia is presented. The Russian operating plant for spent nuclear fuel (SNF) reprocessing with the recovery of long-lived radionuclides is outlined. The results of the most promoted Russian developments on partitioning, including the sorption and extraction studies, are described. The results on operation of the world's first industrial extraction facility for the separation of long-lived radionuclides from high-level wastes (HLW) are given.

Introduction

Russia has accepted the concept of closed NFC with allowance for SNF reprocessing [1]. It is known that the possibility to solve radically the problem on long-term safe management of long-lived radionuclides is one of the advantages of the closed NFC, because the reprocessing of SNF enables to recover these nuclides and to treat them as an individual fraction.

Transmutation is an efficient method for management of long-lived radionuclides. Another promising way is to create the extra strong matrices being disposed into geological formations. In particular, the technology for production of synthetic materials is now under development in Russia with the use of high-temperature synthesis of minerals of zircon group, garnet, cubic zirconium oxide a.o. [2]. The principal feature of these minerals consists of including into mineral matrix an individual element or at least chemical analogs, but not non-separated mixture of nuclides contained in SNF. Such compositions make these materials very close to natural minerals with stability tested during millions of years.

Hence, just as in the case of transmutation, so also in the synthesis of highly strong matrices for geological disposal, there is a need for selective recovery of long-lived radionuclides contained in SNF. This could be provided by the availability of reprocessing plants equipped with special facilities for recovery of long-lived radionuclides in the framework of NFC infrastructure.

In Russia there is now in operation the plant RT-1 for reprocessing of SNF, predominantly from WWER-440 reactors [2]. End product of the plant’s uranium line is uranyl nitrate melt passed to fabrication of fuel elements for RBMK reactors; in such a manner the fuel cycle of WWER-440 becomes closed as applied to uranium even today. Next task is to return plutonium into the fuel cycle as MOX-fuel for thermal and fast reactors.

Management of long-lived radionuclides at the plant RT-1 is similar to other reprocessing plants functioning in the world: non-separated mixture of long-lived radionuclides is vitrified, and the resultant glass-blocks are directed to interim monitored storage in a special store facility. Thus, the efficient separation technologies for radical management of long-lived radionuclides are to be developed for the use in the future.

However it should be noted that even the present practice in management of radwastes requires selective recovery of long-lived radionuclides. This is caused by the fact that the accumulated and current high-level wastes have very complicated composition and contain, as a rule, great amounts of salts. In principle, such non-separated mixture of radwastes could be transferred to solidification without any preliminary treatment, but in this case the following drawbacks inevitably emerge:

- Large volumes of “ballast” material to be vitrified along with radionuclides increase expenses at the stage of glass boiling, as well as at the stage of glass-blocks monitored storage.
- Many macrocomponents of non-separated radwastes have harmful effect on glass boiling process, its safety and the quality of obtainable glass blocks.

Owing to this, there exists an alternative method for HLW management by solidification as follows:

- Long-lived radionuclides are separated from the HLW bulk and are concentrated in small volumes of solutions to be solidified, particularly by vitrification for interim monitored storage and subsequent disposal.

- Ballast mass of radwastes remained after recovery of radionuclides is sent to solidification, by means of cementing in particular, for subsequent near-surface storage as low-level waste (LLW).

The necessary recovery degree of long-lived radionuclides is specified by the limiting standards on their concentrations in LLW for near-surface storage.

The attained standard specifications provide a significant economic effect. For example, the calculations performed by the American specialists have shown that a variant with separation of radionuclides from Hanford HLW would require 12 000 containers for vitrified blocks instead of 40 000 ones needed for wastes without separation; in the case of separation, the saving of expenses would be around US$ 14 billion. The cost comparison of HLW and LLW management at different stages could offer another index of economic efficiency provided by separation of radionuclides from HLW. These data indicate that the production cost of waste in oxide form is equal to 2.126 US$/kg for HLW in comparison with 64 US$/kg for LLW.

Thus, summarising the urgency of efficient technologies for radwaste management, one can advance the following main arguments in favour of such approach:

- A radical promising technique for isolation of long-lived radionuclides from the biosphere by means of transmutation envisages their individual separation from the products of SNF reprocessing for subsequent burning in reactors or accelerators.
- The perspective extra-strong matrices for immobilisation of long-lived radionuclides for long-term safe storage and disposal require selective or fractional separation of the latter.
- The existing technologies for HLW management with vitrification and subsequent monitored storage of glass-blocks require the separation of radionuclides from the bulk of non-radioactive waste for higher glass quality, process safety and economy in operations of glass boiling and storage of solidified materials.

Russian R&D on recovery of radionuclides

In accordance with the foregoing, the systematic studies and developments of efficient methods for separation of long-lived radionuclides from HLW of spent fuel reprocessing have been conducted in Russia for the last 20 years.

These studies are carried out at several Russian Institutes (V.G. Khlopin Radium Institute, Institute of Chemical Technology, Institute of Physical Chemistry, Institute of Geochemistry and Analytical Chemistry a.o.) in collaboration with the radiochemical plants of the RF MinAtom.

Key operation of technologies for separation of long-lived radionuclides involves selective recovery of cesium, strontium, technetium, rare-earth and transplutonium elements, as well as residues of uranium, neptunium and plutonium from HLW remained after Purex-process. In the world’s practice different processes (precipitation, sorption, extraction, chromatography a.o.) for recovery of these components are under development. The object of this communication is to consider those Russian developments which are now most feasible for application.

Ferrocyanide Sorbents for Cesium Recovery

As an example of successful use of sorption processes for recovery of radionuclides from HLW one could consider the method for sorption recovery of cesium by inorganic ferrocyanide-containing sorbents developed by Institute of Physical Chemistry in co-operation with "Mayak" PA. The chemical composition of these sorbents is described by the following bruto-formula [3]

$$K_{1.0}Me^{II}_{1.5}Fe^{II}(CN)_6 \cdot nSiO_2$$

where Me^{II} – Ni, Cu or Zn.

Copper-potassium ferrocyanide was chosen for practical use due to its highest stability in the cycle of sorption – desorption – regeneration. The systematic studies on using this sorbent enabled to develop a technology for cesium recovery from HLW of SNF reprocessing [4]. A sorption column with volume 120 l was used in the technology tests at the "Mayak" PA. The recovery degree of cesium at the sorption stage was more than 98%, cesium yield into desorbate was 98-99%, concentrating degree attained 100. After 15 cycles of operation, the sorption properties of the sorbent remained unchanged. In the course of pilot-industrial tests there were recovered from HLW about 7 mln Ci of cesium.

Crown-ethers for Recovery of Strontium and Cesium

For recovery of strontium from HLW, a method with the use of macrocyclic polyethers (crown-ethers) is elaborated. In particular, the studies of the Institute of Chemical Technology with the use of dicyclohexyl-18-crown-6 (DCH-6) were also brought up to the level of pilot-industrial tests at the "Mayak" PA. As result of these tests there were reprocessed about 90 m^3 of HLW and were obtained more than 0.5 mln Ci of radiostrontium with 6-fold concentrating degree [3]. The recovery degree of strontium was 96%. The characteristic advantage of the DCH-method is the ease of strontium stripping operation being efficiently performed by water.

Investigations are also pursuing with the aim of using crown-ethers for efficient recovery of cesium from HLW [5]. In particular, dibenzo-21-crown-7 in polyfluorosubstituted alcohols extracts cesium in a wide range of acidity and provides rather low distribution coefficients in water stripping operation. 99% recovery of cesium with three-fold concentrating was achieved during the tests on HLW solutions performed at the Radium Institute.

Monodentate organophosphorus reagents for recovery of actinides and technetium

Deep recovery of actinides and lanthanides from HLW may be achieved by means of monodentate neutral organophosphorus extractants. Among them the phosphine oxides possess the most extraction ability; due to high solubility of these compounds in organic diluents, preference was given to different-radical phosphine oxide (POR), i.e. isoamyl dialkylphosphine oxide wherein alkyl radicals have normal structure with 7-9 carbon atoms [6,7].

The possibilities of this extractant were tested on real HLW, affording high recovery degree of actinides (up to 99,9%). Solid extractants wherein the macroporous polystyrol-divinylbenzene matrix contains up to 50% of phosphine oxide were synthesised on the base of POR. Trials of this sorption variant were conducted on industrial scale in 60 L-column. The obtained results have confirmed not only the high recovery degree of actinides, but also the efficient concentrating of them in desorbate. A distinguishing feature of the POR-based reactants is the possibility for recovery of technetium along with actinides. The recovery degree of technetium from actual HLW was about 70% [8].

TRUEX-process for recovery of actinides and lanthanides

In the modern world's practice the possibilities of the so-called TRUEX-process based on the use of bifunctional neutral organophosphorus compounds are studied thoroughly. An interesting modification of TRUEX-process was developed by the Radium Institute in collaboration with the Institute of Geochemistry and Analytical Chemistry [9]. In classical TRUEX-process phenyloctyl-isobutyl-carbamoylphosphine oxide is diluted by dodecane with addition of TBP as solubilizator. In the proposed modification diphenyldibutyl-carbamoylphosphine oxide diluted by a polar organofluoric compound (fluoropol) is used as extractant [10]. Elimination of solubilizator from the extractant composition allows to obviate thus the necessity for washing operation of extractant from TBP destruction products. The technology of the modified TRUEX-process was tested in the facility of centrifugal contactors to assess the possibility for deep recovery of actinides and lanthanides [11].

Feed solution contained 5 M/L HNO_3 and 13 g/L of lanthanides and actinides. Fe(II), Zr(IV) and Mo(VI) were stripped by acetohydroxamic acid; TPE and lanthanides were stripped by 0.01 M HNO_3. The recovery degree of actinides and lanthanides was more than 99.5% at concentrating factor of 4-6 and at purification coefficients from iron, zirconium and molybdenum above 50. These results were confirmed by the tests under static conditions, but on actual industrial HLW [12].

Chlorinated cobalt dicarbollyde in HLW reprocessing

The extraction technology with the use of chlorinated cobalt dicarbollyde (ChCoDiC) in a polar diluent is now the most promoted technology for recovery of radionuclides from HLW. Fundamentals of the extraction process on the basis of ChCoDiC were developed by the specialists of the Radium Institute and the Institute of Nuclear Research (Czech Republic) [13], whereupon the RI in collaboration with the "Mayak" PA brought this development up to the level of industrial use at the radiochemical plant RT-1 [14].

ChCoDiC in polar diluent extracts efficiently cesium from aqueous solutions; addition of polyethylene glycol (PEG) to the extractant makes it possible to extract strontium, as well as TPE and RE. Selective recovery of cesium was tested on actual HLW [6,7], the recovery degree therewith was 99.998%. Different variants of the flowsheets with combined and separate recovery of cesium, strontium, TPE and RE were put through a series of tests from laboratory to pilot-industrial scale [15,16]. The developed and tested pseudomembrane method which enables to increase the recovery degree of trivalent elements from HLW by a factor of tens should be considered as an original modification of the extraction process with the use of ChCoDiC. This is achieved by changing the topology of connections between apparati in the membrane variant as compared to a linear one, providing therewith the increase in efficient capacity of extractant [17,18].

The universal flowsheet for recovery of cesium, strontium, TPE and RE was tested in a test facility in hot cells of the RI with the use of 24 l of actual raffinate from reprocessing of WWER-1000 spent fuel. The tests results on recovery degrees into extract were as follows: Cs – more than 99.999%, Sr – 99.998%, An – over 99.992%. Concentrating degree of recovered radionuclides was equal to six.

The pilot-industrial tests of ChCoDiC-process at the "Mayak" PA confirmed high efficiency in recovery of long-lived radionuclides from HLW. As a result of these tests, the megacurie quantities of cesium and strontium were recovered, the TPE concentrate containing 240 g of americium-241 and 21 g of curium-244 was obtained as well [16].

Industrial Facility for HLW Partitioning at the "Mayak" PA

The most achievement in the use of ChCoDiC-process in Russia is now the introduction of partitioning technology for HLW with different composition at the "Mayak" PA. By means of this technology, the first in the world commercial facility UE-35 for recovery of radionuclides was put into operation in August 1996. The first line of this facility envisages selective recovery of cesium and strontium from HLW. The facility was successfully functioning for three months, then it was stopped because of financial difficulties. However in September of this year the facility was restored to operation and is now in service at present.

During its operation there were reprocessed approximately 400 m^3 of HLW; the concentrates of cesium and strontium with total activity of about 15 mln Ci were produced. The average indices on the recovery degree of cesium and strontium were at a level of 98.5%. The cesium and strontium concentrates were passed to vitrification; this enabled to increase the specific activity of glass blocks up to 550 Ci/kg. Thus, the increase of expenses for the partitioning facility by 5% led to the decrease of the production cost of high-level active glass by 60%.

The second line of the UE-35 facility at the "Mayak" PA was intended for deep recovery of actinides and technetium, along with cesium and strontium from HLW. Three feasible extraction processes are now under investigation for these purposes.

The first process involves the recovery of actinides, RE and technetium by POR from raffinates arising upon the recovery of cesium and strontium by ChCoDiC. This process was successfully tested by joint efforts of the specialists of the Radium Institute and the Idaho National Laboratory at a set-up of centrifugal contactors with the use of actual acidic Idaho HLW [6,8].

The second variant for separation of An and RE from HLW at the "Mayak" PA is based on the Russian TRUEX-process described above. The results of dynamic tests on simulated HLW of the "Mayak" PA were confirmed by static tests on actual raffinates arising from recovery of cesium and strontium at the UE-35 facility. The 99.7% recovery degree of alpha-nuclides was achieved in 4 contacts in the course of these tests.

The third alternative variant of the second line of the UE-35 facility at the "Mayak" PA envisages the use of an unified process for recovery of cesium, strontium, actinides and RE. The fundamentals of this process were elaborated by the specialists of the Radium Institute and the Idaho National Laboratory and were brought up to the level of dynamic tests in multi-stage centrifugal contactors on actual acidic HLW of the Idaho Center [19,20].

Universal Extractant for Separation of Cesium, Strontium, Actinides and Rare-earth Elements from HLW

The universal extractant (UE) is a mixture containing chlorinated cobalt dicarbollyde, diphenyl-dibutyl-carbamoyl phosphine oxide with addition of polyethylene glycol and specific diluent. This solvent extracts effectively from acidic HLW cesium, strontium, uranium, neptunium, plutonium, americium, curium and RE.

The flowsheet of this process was tested on actual HLW of the Idaho Center with the use of 24-stage cascade of centrifugal contactors including 8 extraction stages, 2 stages for extract scrubbing, 6 stages for cesium and strontium stripping, 3 stages for An and RE stripping, 5 stages for extractant regeneration.

In the tests conducted in June of this year the recovery degrees were as follows: Cs – 99.95%, Sr – 99.985%, gross alpha-activity – 95.2%.

In view of potential prospects for introducing the universal extractant at the “Mayak” PA in Russia and the Idaho National Laboratory, efforts are now made to optimise this process. Problems on operational safety, i.e. explosion and fire-safety of the process, toxicity and corrosion agressivity, are also under study. It should be noted in this connection that these problems are elaborated in detail, as applied to the ChCoDiC-process with the aim of cesium and strontium recovery.

Conclusion

The concept of closed nuclear fuel cycle is accepted in Russia; this enables to solve radically the problem of long-term safe management of long-lived radionuclides. In accordance with this concept, there is created the appropriate infrastructure including spent nuclear fuel reprocessing and management of radwastes in the nuclear industry of the Russian Federation.

To reduce the volumes of HLW being vitrified, a facility for recovery of long-lived radionuclides has been constructed at the reprocessing plant of the “Mayak” PA. The efficient separation technologies are under development not only for reduction of HLW volumes, but also for obtaining of the fractions of individual radionuclides. This creates prerequisites for the future use of radical methods for management of the most hazardous radionuclides by transmutation techniques or by final disposal in the form of very strong matrices of Synrock type.

Several Russian institutes are developing the sorption and extraction processes which provide the deep recovery of radionuclides. Among the methods under development, the most progress has been made in using the ferrocyanide sorbents, mono- and bidentate organophosphorus compounds and crown-ethers.

The most advanced method for HLW reprocessing is now the extraction by chlorinated cobalt dicarbollyde. This method which affords deep recovery of cesium and strontium has been introduced on industrial scale at the “Mayak” PA (UE-35 facility). The introduction of the first partitioning stage has already given an economic effect due to reduced volumes of HLW being vitrified.

Three process variants for the second line of the UE-35 facility are now under development for recovery of actinides, RE and technetium. The first variant is based on POR, the second – on the Russian modification of TRUEX-process, the third – on the use of the so-called universal extractant (mixture of ChCoDiC, CMPO and PEG in special diluent).

Further efforts of the Russian specialists in separation technologies are aimed at obtaining the individual fractions of long-lived radionuclides, as well as at affording the safety of processes and equipment being in service.

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STATUS AND ASSESSMENT REPORT ON ACTINIDE AND FISSION PRODUCT PARTITIONING AND TRANSMUTATION

On behalf of the OECD/NEA Working group on P&T

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Abstract

A working group composed of 30 scientists representing 12 countries and three international organisations have prepared a comprehensive status and assessment report on the Partitioning and Transmutation (P&T) of actinides and fission products in the frame work of an advanced fuel cycle option and as an alternative or complementary waste management option.

The report is subdivided into two main parts: the first part gives a general overview of P&T with a critical evaluation and general conclusions; the second part covers a technical discussion and a systems analysis of partitioning, transmutation and risk analysis within an advanced fuel cycle and in a long term waste management option. Separate chapters are covering the Japanese and French R&D programmes which are the leading programmes within the OECD countries.

Introduction

For about 10 years the Partitioning and Transmutation (P&T) option has been revived due to the OMEGA initiative taken by the Japanese government in 1988. In 1990 the French government decided to launch a similar programme called SPIN (*Séparation - Incinération*) which was requested by law to investigate the alternative high-level waste (HLW) treatment by P&T in parallel with the development of the geological disposal option.

In the mean time important R&D efforts have been accomplished throughout the world to investigate means to reduce the long-term radiotoxic inventory resulting from the nuclear power generation.

Partitioning is a complex series of chemical and/or metallurgical operations, intended to separate selected radiotoxic nuclides or groups of nuclides occluded in the spent fuel elements. The separated fractions can be stored as such or transformed into new fuel elements or irradiation targets.

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

Fuel cycle options

Fuel cycles

At the industrial level there are two major fuel cycle options:

- The “Once-through-cycle” (OTC) or direct disposal option of spent fuel.
- The “Reprocessing fuel cycle” (RFC) with MOX recycle and disposal of HLW.

The OTC is an option taken by the US, Sweden, Spain, Canada, Korea and some other countries. The RFC is the traditional nuclear fuel cycle which limits the recycling to uranium and plutonium and leaves the minor actinides (MA: neptunium, americium and curium) in the HLW mixture, this option is industrially available in France, United Kingdom and Japan. Some other countries: Russia, China and India have important activities in this field.

The P&T option is an extension of the RFC. Partitioning requires additional separation operations to extract the MAs and some selected Long-Lived Fission Products (LLFP: ^{99}Tc, ^{129}I, ^{79}Se, ^{93}Zr and ^{135}Cs) from HLW. In order to recycle and burn the MAs and to transmute LLFPs, dedicated nuclear facilities have to be developed. The P&T option is part of a so called “Advanced Fuel Cycle” (AFC) which could be realised as an extension of the present fuel cycle industry and the nuclear power plant (NPP) park.

Alternatively the AFC could be accomplished by separate operation of, on the one hand, the electricity producing reactors and their associated fuel cycle facilities and, on the other hand, the recycling and burning facilities of MAs and LLFPs operated independently from the major industrial nuclear-power activity. This type of AFC option is called “the double strata fuel cycle” and was pioneered by Japanese nuclear research organisations.

Fuel cycle issues

The OTC is undoubtedly the cheapest nuclear energy production but it leads to the accumulation of large inventories of spent fuel containing long lived actinides and fission products. The long-term impact has to be controlled by man-made systems and natural barriers which should provide protection for as long as the life of the radiological source term they confine.

The RFC option results in a partial recycling of major actinides (U and Pu) and produces in terms of residual radiotoxicity essentially vitrified high level waste (HLW) which has to be disposed of in suitable repositories. However, these glasses contain MAs and all the fission products. The radiotoxic inventory of these wastes is significantly lower than spent fuel but their toxic lifetime is determined by the presence of some MAs and LLFPs. The MAs have half-lives which range from several hundreds to million years and some LLFPs have half-lives of hundred thousand to several million years. Recycled U and Pu in fresh LWR-MOX fuel constitutes, in comparison with the OTC option, a reduction of the Pu-containing spent fuel volume but does not decrease the overall long-term radiotoxicity.

The AFC comprises all the fuel cycle and reactor sequences of the RFC but involves additionally a complex set of chemical separations to remove the MAs and some LLFPs from HLW before vitrification. Apart from the use of LWRs for energy production the AFC option requires the industrial deployment of Fast Reactors (FRs) in order to reduce the inventory and the radiotoxic half-life of separated MAs by transmutation or “incineration”. The LLFPs could, in principle, be transmuted to inactive or shorter-lived isotopes in dedicated LWRs or in accelerator-driven systems (ADS).

The combined use of LWRs and FRs would stabilise the actinide inventory and reduce the MA content in the vitrified HLW. However, the gradual increase of the Pu+MA inventory in the reactors and fuel cycle facilities calls for additional actinide destruction tools such as ADS in order to eliminate the accumulated actinides and possibly to transmute some LLFPs.

Whatever option taken, it will always be necessary to build and operate geological repositories in order to isolate the HLW, the α bearing medium level waste and ultimately residual reactor cores from dedicated reactor systems.

Advanced reprocessing and partitioning

Improved aqueous reprocessing

Industrially aqueous reprocessing is carried out according to PUREX process by which spent fuel is dissolved in HNO_3 and treated by liquid extraction with tributylphosphate (TBP). Uranium and plutonium enter into the TBP phase and the other components remain in aqueous solution which is concentrated into HLW. This highly active waste stream contains the MAs and most of fission products (FP). Eventually this waste stream is vitrified and stored until disposal.

The additional operations to realise an AFC process can partially be carried out, on line, within the current reprocessing facilities and partially, off line, from the stored liquid HLW stream before vitrification.

The easiest adaptation of the conventional reprocessing process is by addition of the neptunium separation to the first cycle of the PUREX process. The neptunium separation process could be included in the current large reprocessing plants if some additional modifications of the separation flowsheet by REDOX reactions would be performed.

Some LLFPs constitute the very long-term risk for the disposal of HLW and are, or could be separated, during the PUREX process. The most important nuclides are: ^{129}I and ^{99}Tc.

The current reprocessing process involves the separation of ^{129}I ($t_{1/2}$ =15.7×10^{6}) from dissolver off-gases and its discharge into the ocean as medium-level liquid waste. The iodine scrubbing could be complemented by a specific conditioning operation in order to obtain a stable matrix (as insoluble as possible) in aqueous media.

Separation of ^{99}Tc ($t_{1/2}$ = 213 000 years) is a more complex issue since it occurs as insoluble residue from the dissolver process and as soluble Tc in HNO_3. By scrubbing the TBP stream from the first extraction cycle with high-acidic NO_3, ^{99}Tc could quantitatively be eliminated from the HLW and treated separately, if necessary, in order to combine it with the insoluble fraction and to condition this in a stable metallic or oxide form.

Separation of minor actinides (MA) from HLW

Whereas Np could be separated during conventional aqueous reprocessing, this is not the case for the two main radiotoxic MAs: Am and Cm. Special off-line separation techniques from liquid HLW are necessary. This separation is difficult because rare earths (lanthanides) are generated during the fission process with ten times lager mass than MA and because the chemical behaviour of lanthanides is very similar to that of the MAs. Very high separation factors have to be reached in order to get an acceptable chemical purity of the separated MA fractions.

For about 10 years R&D on this subject has been performed in the US-DOE laboratories (ANL, SRP, LANL), in Japan (JAERI, PNC), France (CEA-FAR & Marcoule), Russia (Saint-Petersburg,) China and within the national nuclear laboratories in Europe and the laboratories of the European Commission (TUI).

TRUEX process

At ANL (USA) a process was developed using a very powerful extractant, CMPO, which allows to extract the MAs together with the lanthanides from strong acid solutions. The method was tested on real HLW solutions at PNC (Japan) and showed to be very efficient. There are some technical problems to be solved but as a whole this technique is a possible route to decrease the radiotoxicity of HLW. The TRUEX process is compatible with the PUREX technology.

The main unresolved question is the separation of lanthanides from MAs and the associated production of secondary waste.

DIDPA process and talspeak process

At JAERI, a new process was developed using DIDPA as extracting agent, this process has been very thoroughly investigated but suffers from the drawback that concentrated liquid HLW is much too acidic and that denitration has to be performed prior to the extraction. This leads to losses of insoluble actinides (Pu+MA) and reduced separation yields. One of the advantages of the DIDPA process is the incorporation of Np into the Am-Cm separation.

The separation of the Am-Cm fraction from the lanthanides is proposed by a so called "TALSPEAK" process used industrially for the separation of individual lanthanides. This process, if slightly adapted, could advantageously be used on the direct extraction of MAs from the more dilute first aqueous-raffinate of the PUREX process.

TRPO Process and cyanex-301 process

Recently a new process was developed in China using TRPO as extractant for the mixture of actinides and lanthanides from 2M HNO_3 waste stream. A straight-forward extraction of Ac-Ln fraction followed by stripping stages to remove impurities has been demonstrated with slightly diluted liquid HLW at the TUI of Karlsruhe.

Very promising performances in the direct separation of Am-Cm from lanthanides have been demonstrated with a special reagent "CYANEX-301". Although it is applied to low-acidic solution, this development is the most important one in this field for many years.

DIAMEX-SESAME Process

New, very effective extraction reagents for MAs, called "DIAMIDES", which are fully incinerable, have been developed by CEA-FAR and Marcoule and tested in hot-cell conditions. The reagent DMDBTDMA is almost as powerful as CMPO and can be used in very acidic media. Additional development work will be necessary to make this process applicable in industrial conditions in order to obtain acceptable purity of An in the product. The electrochemical oxidation process called "SESAME" process is the only one in the world which can, in principle, separate Am/Cm in one process step. The combination of DIAMEX with SESAME offers perspectives to separate the An from Ln and prepare pure chemical fractions of Am.

Conclusions and recommendations

From the above overview it is clear that great progress has been accomplished in the group separation of Ln+An from liquid HLW but that purification and separation of each MA by liquid extraction are still in the exploratory phase. Most of these methods are still on the bench scale or at hot-cell laboratory level. Table 1 shows a synthetic overview of the development state of each method.

In order to be useful on industrial scale, the above listed methods have to be **upscaled in terms of radioactivity level and throughput** of the liquid extraction rigs. Moreover very much attention will have to be devoted to the **secondary waste** issue which may become the bottleneck of their applicability in industrial facilities.

Table 1. **Status of R&D on aqueous separation techniques**

	Phase 1	Phase 2	Phase 3	Remarks
U and Pu separation (PUREX)	-	-	-	achieved industrially
Np separation (PUREX)			❍	95% separation
(PUREX)		❍		> 95% separation
(DIDPA)		❍		
(HDEHP)		❍		
(TRUEX)		❍		
Am + Cm separation:				
* based on An/Ln co-extraction				
(TALSPEAK)			❍	
(DIDPA)		❍		
(TRUEX)		❍		
(TRPO)		❍		
(DIAMEX)		❍		
* based on An selective extraction				
(TPTZ)	❍			
(Picolinamides)	❍			
(CYANEX 301)	❍			SF=5 900
* based on precipitation				
(Ferricyanide)	❍			
Am separation in the oxidised state				
(SESAME)		❍		Am/Cm separation
Tc separation (PUREX)			❍	soluble Tc
(PUREX)	❍			insoluble Tc
Tc - PGM separation				
(Denitration precipitation)		❍		
(Active carbon adsorption)		❍		
I separation (PUREX)			❍	95% separation
Zr separation (PUREX)		❍		
Cs separation (Calixarenes)		❍		
(Zeolite)		❍		
Sr separation (Titanic acid)		❍		
Cs and Sr separation (Dicarbollides)			❍	
Pd (PGM), Se, Ru separation				
(Electrolytic extraction)	❍			soluble Pd, Se, etc.

However it is also important that the more fundamental type of R&D, on the **synthesis of new selective extractants**, on their analytical characteristics and on their radiation stability, would be continued.

Dry reprocessing

In the future, nuclear fuel will reach higher burnups and recycling of spent fuel will have to be performed after short cooling times to be economically and ecologically attractive. Dry reprocessing using pyrochemical methods received for a number of decades the attention of some research institutes: the RIAR (Dimitrovgrad) in Russia; ANL (Argonne (Idaho)) in the USA; CRIEPI in Japan, and some older studies in France, Belgium etc.

Dry reprocessing is a pyro-electrochemical extraction of actinides from chlorinated spent oxide fuel which involves the following high temperature (650°C) processes: chlorination of oxide fuel, pyro-electrolysis of UO_2, selective precipitation, and additional electrolysis for the TRU nuclides. Recycling of Li-KCl, Cd and/or Bi reagents, conditioning of the salt and metal waste forms.

The proposed processes are very complex and necessitate the use of highly controlled atmospheres to avoid hydrolysis and precipitation reactions. However these processes have the inherent advantage in criticality safety considerations and in the compactness of the plant

Except for the pilot-scale demonstration of the pyro-electrolysis at Argonne-West and RIAR, all the other studies are at the laboratory scale. If other fuel types, e.g. nitride fuel or metallic fuels are to be used in multi-recycling processes, pyro-electrochemical methods might become necessary to implement this option.

Very much R&D work will be required in order to upgrade the pyro-electrochemical reprocessing process to the level of the present industrial aqueous reprocessing. But in order to shorten the cooling times which determine the recycle period of highly irradiated fuel (>150 GWd/tHM) the development of alternative recycling processes to the present PUREX process will become mandatory.

Possible scenarios for separated MAs

There are several scenarios which can be considered in the management of separated minor actinides:

- Neptunium resulting either directly from the reprocessing process or extracted from HLW could be separated easily from the other nuclides and recycled into MOX fuel or kept in separated form until advanced transmutation-incineration processes (FR, ADS) become industrially available. Alternatively, the separated neptunium could be conditioned in a thermodynamically stable phase and kept in a retrievable long-term storage. This matrix could be designed to serve either as irradiation matrix or as final disposal waste form.
- The Americium-curium fraction separated from HLW could be treated as a single fraction both in a transmutation strategy and in a disposal option. Both elements behave chemically so similar that very special radiochemical and/or electrochemical techniques have to be used to achieve the americium/curium separation. In a group separation option the americium-curium fraction could be stored as such or conditioned as a ceramic type of matrix and kept in engineered storage until new transmutation techniques become available.
- Americium could ultimately be separated as an individual element and recycled into special LWR-MOX or FR-fuel to be partially transmuted or conditioned in a ceramic form for extended transmutation-incineration processes by multi-recycling and ADS processes.

- The management of separated curium has not yet received convincing answers not only because the americium-curium separation is difficult but also because this very radioactive nuclide (heat and neutron source) cannot be recycled in LWR- or FR-MOX fuel. Storage of the separated nuclide for a century would alleviate the processing problems and make recycling in an advanced fuel cycle possible.

Transmutation of radiotoxic and long-lived nuclides

Since transmutation is the only method to reduce the half life of the separated nuclides, the separated MA and LLFP fractions have to be transformed into irradiation targets and submitted to neutron fluxes. Two options are open; irradiation with thermal neutrons or with fast neutrons.

Irradiation of MAs and LLFPs by thermal neutrons

The most obvious route to be followed is apparently to try to destroy the radiotoxic nuclides in the existing facilities in which they were produced. The existing NPPs could for example be specialised to carry out such job.

Transmutation of Neptunium

Among the MAs, Np is the easiest nuclide to be separated during reprocessing, and without major problems, it can be mixed with MOX fuel and irradiated. The transmutation yield (Table 2) in a single pass varies from 40 to 50% but the main product of the transmutation is ^{238}Pu with 87 years half life. The gain resulting from this operation is the disappearance of ^{237}Np with 2×10^6 years half-life but its replacement by a highly toxic nuclide which would interfere with multi-recycling operations by aqueous reprocessing. Mixing of Np with MOX fuel will increase the radiotoxic inventory of the spent LWR-MOX assemblies, but it will reduce a very long-term risk. The Np reduction factor by such single step irradiation varies from two to three. Transmutation of Americium

Mixing of a pure ^{241,243}Am fraction with MOX fuel requires already an adaptation of the fuel fabrication equipment and technology. The gamma emission of ^{241}Am increases the radiation field around the fabrication units which have to be shielded and remotely operated. But if this adaptation is made ^{241,243}Am fraction can be recycled into fresh LWR-MOX fuel.

During irradiation, ^{241}Am undergoes transmutation into ^{242}Am which partly decays into ^{238}Pu; and ^{243}Am is mainly transmuted into ^{244}Cm, a highly radioactive nuclide which decays into long-lived ^{240}Pu. The annual depletion of Am ranges from 40 to 80% (Table 2). However the main end products of a single-pass irradiation are ^{238}Pu and ^{240}Pu.

Table 2. **Mass balances for homogeneous recycling in thermal reactors**

		PWR-UOX* moderation ratio 2	PWR-MOX** moderation ratio 2	PWR-MOX*** moderation ratio 3
Burn-up (*GWd/t*)		47.5	47.5	47.5
Initial minor actinide content (%) (mass of heavy metal)		1	1	1
Np	Initial mass (*kg*)	1 104	1 104	1 104
	Specific consumption (*kg/TWhe*)	12	10	11
	Depletion (% of initial mass)	53	43	45
Am	Initial mass (*kg*)	1 104	1 104	1 104
	Specific consumption (*kg/TWhe*)	19	5	10
	Depletion (% of initial mass)	83	20	42

* U-235 enrichment of 5.5% to recycle Np, or Am (standard enrichment 4.5%)
** Pu enrichment of 12% to recycle Np and 12.5% for Am (standard enrichment 9%)
*** Pu enrichment of 7.7% to recycle Np, 8.2% for Am (standard enrichment 5.7%)

Transmutation of Curium

Recycling of the ^{243,244,245}Cm fraction into fresh LWR-MOX fuel is a very difficult task which cannot be realised unless dedicated fuel fabrication plants with extensive neutron and gamma shielding are being developed. The irradiation of the Cm fraction yields a range of long-lived nuclides, e.g. ^{246,247}Cm, and decay products ^{239,240}Pu, and ^{241}Am. There is no gain in radiotoxicity and a long term reduction of the Cm inventory would require multi-recycling by very special reprocessing processes, e.g. pyro-electrochemical techniques.

Presently the preferred strategy would be to store separated Cm for ~100 years and to re-inject the resulting Pu nuclides into MOX fuel.

Transmutation of long-lived fission products

Iodine-129

Iodine being separated from dissolver off gases can be transformed into a single compound, e.g. insoluble AgI, $PbIO_4$ or kept as soluble NaI. Transmutation of ^{129}I is a very slow process since the capture cross section is very small for practical transmutation purposes.

Whatever the final product, it is necessary to develop a vented capsule or pin, since ^{129}I transforms during irradiation into gaseous ^{130}Xe. The thermal stability of the target material during irradiation is a questionable issue dedicated reactors would in any case be necessary because of the high iodine inventory.

Technetium-99

If the soluble and insoluble Tc fraction can be combined into a single metallic target, irradiation in LWRs is possible since the transmutation reaction yields a metallic Ru-100 which is inactive. The half-life of this irradiation its inventory reduction with a factor two is about 40 years in LWR-UO_2 and about 80 years in LWR-MOX.

Other LLFP

The other significant LLFP are: ^{93}Zr, ^{135}Cs.

Due to the relatively low mass fraction of the radionuclide (20% ^{93}Zr, 10% ^{135}Cs) in the total fission yield the presence of other isotopes would interfere in the transmutation reaction. Further advance in this field can only be achieved if isotopic separation becomes an economically feasible intermediate step.

Conclusions for R&D activities on transmutation in thermal neutron spectra

- There is a need for more precise determinations of the **MA generation** in spent LWR-MOX fuel by a more elaborate verification of the yields in high burnup LWR-UO_2 and LWR-MOX assemblies.
- Study of the impact of **multi-recycling of MAs** in LWR-MOX as a function of cooling time.
- The **delayed transmutation of Cm** after 100 years storage would have to carefully examined in order to establish the potential benefits of recycling.
- Among the LLFP **only ^{99}Tc** could technically be considered for transmutation in thermal reactors. The core management of this option needs to be carefully established: in core inventory, transmutation yields, residence times etc. as a function of a preset inventory reduction.

Transmutation and incineration in fast reactors

The prime advantage of irradiating nuclides particularly MAs in fast neutron fluxes is the increase of the fission contribution in the total transmutation yield. A secondary effect is the very high fast flux in FRs, which can be partially thermalised if necessary, e.g. by installing solid neutron-moderator materials in the peripheral section of the FR core.

Irradiation of MAs in FRs

There are two recycling modes to be considered: homogeneous and heterogeneous recycling. The advantages of heterogeneous recycling are apparent for the fuel cycle operations (reprocessing, fuel refabrication waste management) and the total transmutation capability is almost identical for homogeneous and heterogeneous recycling. However in standard core calculations homogeneous recycling is used as standard method.

Either specially fabricated fuel assemblies with high MA content (25 to 50%) or pure Np, Am, Cm targets have to be introduced in the FR core. But for reasons of reactor safety the total MA content in a sodium cooled core may never exceed 2.5% because of the positive sodium void coefficient.

By using lead cooling higher MA contents can be used, but the negative reactivity of the MA has to be compensated at BOC by an increase in the fissile isotope (^{239,241}Pu or ^{235}U) content. Due to the formation of fissile nuclides (^{239}Pu, ^{242}Am, ^{245}Cm) from the "fertile" isotopes (^{237}Np, ^{241}Am, ^{244}Cm) the burnup-reactivity swing becomes smaller than in a conventional core loading.

According to computations (Table 3) the depletion yield for ^{237}Np and ^{241,243}Am targets/fuels in FR-MOX reactors amounts to 60% composed of 23% fission and 37% capture. The deletion rate of ^{241,243}Am amounts to 45% partly fissioned and partly transmuted into other actinides.

Table 3. **Mass balances for homogeneous recycling in fast reactors**

		FBR*
Burn-up (*GWd/t*)		120
Initial minor actinide content (%) (mass of heavy metal)		2.5
Np	Initial mass (*kg*)	1 010
	Specific consumption (*kg/TWhe*)	10
	Depletion (% of initial mass)	60
	Fissioned fraction (% of initial mass)	27
	Fissioned mass (*kg*)	273
Am	Initial mass (*kg*)	1 174
	Specific consumption (*kg/TWhe*)	9
	Depletion (% of initial mass)	45
	Fissioned fraction (% of initial mass)	18
	Fissioned mass (*kg*)	211
*EFR type reactor with three enrichment zones (15.32, 18.18 and 22.08%)		

The mixed MA loading method, a combination of the homogeneous and heterogeneous methods was examined as an alternative; Np is uniformly loaded in the core region and a small number of subassemblies containing Am, Cm and RE nuclides is loaded into the blanket region. Parameters were surveyed systematically to investigate the basic characteristics of MA transmutation in a 1 000 *MWe*-class FR core with mixed oxide fuel.

The mixed MA-loading method can transmute a large amount of MAs without serious drawbacks in terms of core performance. The transmuted mass of MAs is ~530 *kg/cycle*, which is almost 16 times the mass produced by an LWR of the same power output.

It was found that a combination of homogeneous and heterogeneous loading methods, has the potential to achieve the maximum transmutation of MAs with no special design consideration.

Transmutation of MAs and LLFPs in Dedicated Systems

Dedicated transmutation systems are being studied at JAERI and CEA and based on a strategy named the double strata fuel cycle concept (or multi-component concept). The double strata concept is to consider a P&T fuel cycle (second stratum) separated completely from the conventional fuel cycle for commercial power reactors.

The P&T cycle could be made very compact by co-locating the entire facilities. This would minimise the transportation of nuclides that are troublesome with respect to waste management, and confine them effectively in the P&T fuel cycle.

Both critical reactors (burner reactors) and sub-critical reactors (hybrid systems or accelerator-driven systems) are potential candidates as dedicated transmutation systems used in the second stratum.

In dedicated “burner” reactors studied by JAERI: lead-cooled MA-burner reactor and He-cooled MA burner reactors with coated particle nitride fuel the transmutation is much higher, but the price to be paid is an initial enrichment of the MA-fuel with 35% ^{235}U.

In this case burner fuel containing 65% MAs has been considered. The major characteristic of this type of reactor core is the very high mean neutron energy which amounts to 720 keV which is close to the fission threshold of MAs. The MA fission in the ABRs ranges from 11 to 13% per cycle and is 190 to 200 Kg/GWth-year.

Accelerator driven transmutation systems

Accelerator-driven systems (ADS, frequently called hybrid systems) combine high-intensity proton accelerators with spallation targets and a subcritical core with or without blanket. The proton accelerator will be either a linear accelerator (linac) or a circular accelerator (cyclotron). The high-intensity continuous-wave (CW) proton beam with an energy around 1 *GeV* and a current of several tens *mA* are injected into a target of heavy metal. This results in spallation reaction that emits neutrons, which enter the subcritical core to induce further nuclear reactions. The subcritical core can, in principal, be operated with either a thermal or a fast neutron spectrum.

ADSs have unique features to burn MAs and possibly FPs, preferably in the double strata option. They operate in a subcritical mode and can more easily address safety issues associated with criticality than in critical systems. They also offer substantial flexibility in overall operation. ADSs can provide more excess neutrons compared to critical reactors. The excess neutrons may be utilised for transmutation, conversion, and breeding purposes. These features may be exploitable to prepare a safe and efficient means of transmuting nuclear waste. Both homogenous and heterogeneous fuel recycling is possible.

Various concepts of ADS have been proposed with different goals and approaches. Relevant R&D programmes are being pursued at CEA, JAERI, LANL, CERN, etc. In recent years, all the system concepts proposed by these groups have converged on a fast neutron spectrum because of its large neutronic advantage over the thermal one, and the reduced production of higher actinides.

Technical issues

High intensity proton accelerators are operational at Los Alamos National Laboratories (USA) and at the Paul Scherrer Institute in Switzerland. The LANSCE is a 1 MW linear accelerator while the PSI accelerator is a 1 MW cyclotron.

In the high-intensity accelerator, beam loss should be minimised to avoid resultant damage and activation of accelerator hardware. For industrial applications, primary issues are the stability, efficiency, reliability, operability and maintainability of the accelerator. Recent reviews of the operation reliability of accelerators have shown that beam trips and proton source failures are very frequent. Substantial improvement is required and R&D priority should therefore be given to that essential part of ADS. Further R&D and operational experience will increase the dependability. Reducing the cost and size of the accelerator should also be included in the design priorities.

Lead and lead-bismuth spallation targets are presently being considered in design studies. At present most of the available data and codes are sufficient for approximate evaluations. But uncertainties in the prediction of spallation products and neutron multiplication factors should be reduced in order to allow detailed technical design studies.

Trips and fluctuations of the incident proton beam are inevitable, causing thermal shocks in the core components. Therefore the design must take this into consideration: power distribution, effective neutron multiplication factors, the neutron flux shape transient response and the size of the system.

The accelerator-driven subcritical system has clear safety advantages for severe reactivity accidents. It can cope with fast ramp rate accidents which could occur too rapidly for scram systems in critical reactors. A margin to accommodate fast reactivity insertions is important to avoid supercriticality accidents.

The consequences of cooling failure for ADSs are similar to critical reactors. A reliable beam shut-off system is, therefore, required for an ADS, just as a reliable scram system is required for a critical reactor. A reliable emergency decay heat removal system is required for both.

Performance assessment studies

Over the past few years a number of different ADS concepts have been developed. The most attractive are those where a fast neutron flux is being used. Two studies are commented in the OECD-NEA report; one by FZK, Karlsruhe and one by JAERI, Tokai.

In the FZK study the sodium cooled, fast spectrum ADS employs a very high power accelerator of up to about 200 *mA* of proton beam with an energy of 1.6 *GeV*. The spallation target is MAO_2 or PuO_2 fuel itself. The proton beam is spread over the entire surface of the target to avoid the hot-spot problem. In this scenario, three different cores are required; the first one is for the incineration of MA from 14 units of LWRs, the second core is loaded with ^{99}Tc and ^{129}I and plutonium produced in 19 LWRs. ^{129}I eventually left over from this core is transmuted in the third core, fuelled by a fraction of Pu coming out of the second core. It was shown that the considerable power swings between the BOC (k_{eff} = 0.95) and the EOC (keff = 0.80) stages could be smoothed out by adjusting the beam current. A technical difficulty of this concept is the use of a spallation target of MAO_2 or PuO_2 and the use of spread proton beam.

JAERI designed two-types of subcritical cores of 820 *MWt* with nitride fuels. One type is a Na-cooled core with a solid tungsten (W) target at the core centre and the other is lead-bismuth target and coolant system. The spallation targets are bombarded by high energy and power protons of 1.5 *GeV* and 45 *mA*. The parametric studies were conducted to obtain the optimal neutronic characteristics of the subcritical core to maximise the MA transmutation rate and to minimise the burnup reactivity swing during irradiation by adjusting the MA and Pu fraction of nitride fuels with ZrN inert matrix as thermal diluent. The coolant void reactivity is negative in the lead-bismuth cooled core, though it is positive in the Na-cooled one. The MA transmutations in both cores are 250 *kg/y* and this amount corresponds to the annual production of MA in about 10 LWRs.

Transmutation of long-lived fission products (LLFP)

The transmutation capacity for long-lived fission products in conventional reactors is very limited, and these neutron-absorbing substances tend to poison the core. The reactor neutron balance makes it conceivable to recycle some but certainly not all of them. The transmutation yield is very low because the cross sections are low, but the presence of other, e.g. inactive isotopes in the target may result in the production of LLFP and interfere with the depletion reactions.

Only the irradiation of ^{99}Tc and ^{129}I in standard PWRs has been considered in calculations with these fission products located in special target pins and inserted into the guide tubes of the PWR assembly. ^{99}Tc was assumed in metallic form, iodine was considered to be in the form of cerium iodide (CeI_3) or sodium-iodide (NaI). The stability of the iodine target during the irradiation is the most important bottle-neck in the search for high transmutation yields and conversely shorter irradiation half-lives.

Transmutation of ^{99}Tc in fast reactors may be accomplished in several ways: in a special moderated sub-assembly loaded at the periphery of the core or in the inner core, or in a non-moderated subassembly loaded in the core. Moderation could be realised with a material like CaH_2. Table 4 gives for ^{99}Tc some calculated annual transmutation rates in LWRs and FRs, together with the radionuclide inventories and the irradiation half -lives.

As a general conclusion it can be postulated that **transmutation of ^{99}Tc or ^{129}I in present reactors is not encouraging** because of the long transmutation half-lives and the huge inventories of fission products required. Special-purpose high flux reactors or ADS systems may perhaps improve the prospects.

Table 4. **Ranking of reactors with respect to ^{99}Tc transmutation capability**

Reactor	Configuration	Inventory ^{99}Tc (*kg*)	Transmutation ^{99}Tc (*kg/y*)	Transmutation ^{99}Tc (*kg/MWe y*)	Irradiation Half-life(*) (*y*)
FR	Moderated S/A in inner core	2 741	122	0.11	15
FR	Non-moderated S/A in inner core	2 662	101	0.09	18
LWR	Pin in guide tube UO_2 fuel	3 633	64	0.07	39
LWR	Pin in guide tube MOX fuel	1 907	17	0.02	77

(*) Irradiation half life $T_{1/2}=\ln 2/\sigma\varphi$

Impact of P&T on risk assessment and waste management

Radiological risk assessment

The radiological risk associated with the long-term waste management is a very complex issue which involves the radiotoxic inventory of the waste or the spent fuel, the confinement conditions of the radionuclides during the disposal period and the expected collective dose to man resulting from the release into the biosphere over a very long time interval.

It is recommended to abandon the term ‘potential radiotoxicity’, in speaking of nuclear wastes, because it gives the illusion of a management scenario for these wastes, whereas it is merely an inventory. It is preferable to express for each radionuclide the inventories in *Bq/t* of fuel (or of heavy metal it contains) or in *Bq/TWhe*.

Radiotoxic inventory of wastes

The general strategy of introducing P&T as an additional waste management option is based on the radiological benefit which is expected from such an option. The ranking of the actinides and long lived fission products can be made on the comparison of their intrinsic hazard (effective dose coefficients, *Sv/Bq*) coupled to their radioactive concentrations in spent fuel or HLW (*Bq/tHM*), as a function of time.

In the case of the OTC all radionuclides contribute to the source term and the long term radiotoxic inventory is mostly due to Pu, MAs and some LLFPs. However, the conditioning operations can provide artificial barriers which are potentially capable of confining the radionuclides within their package for thousands of years. After this time interval nothing can be predicted except that the solubility of the actinides (except Np) is generally very low whereas the long-lived fission products, particularly ^{135}Cs, ^{129}I and in some cases ^{99}Tc, display high mobilities in the geosphere.

In the case of the RFC, the reprocessing operation and recycling of Pu as LWR-MOX significantly reduces the radiotoxic inventory of the HLW. In this case the long-term radiotoxicity is essentially determined by the minor actinides (Np, Am and Cm,) and the long-lived fission products ^{99}Tc, ^{126}Sn, ^{79}Se and ^{135}Cs. An important long-lived fission product ^{129}I does not appear in this list since it is discharged into the ocean as a consequence of the reprocessing operations. Between 100 and 10 000 years the most important radiotoxicity contribution in the HLW comes from Am and Cm isotopes. Beyond that period both isotopes have significantly decayed to ^{239,240}Pu (daughters of ^{243,244}Cm) and ^{237}Np (daughter of ^{241}Am) which becomes predominant.

The radiotoxic inventory of spent MOX fuel is much higher since the Pu content of seven spent UO_2 assemblies is “concentrated” in one MOX fuel element, the “actinide” radiotoxic inventory of a spent MOX fuel element is about eight times that of spent UO_2 fuel elements.

In a long-term perspective of waste management the disposal of spent MOX fuel is a major factor in the overall assessment of the radiotoxic inventory in the RFC fuel cycle. Reprocessing of spent MOX fuel and recycling of recovered plutonium are therefore important issues which determine the long term radiotoxicity.

The main impact of the AFC strategy is a significant reduction in the radiotoxic inventory of the vitrified wastes with conversely a transfer of the long-lived actinides to the fuel cycle facilities and reactor core inventories. This long-term benefit has to be weighed against the short term doses to workers, and the production of additional contaminated wastes, due to the increased complexity of the fuel cycle

P&T facilities for TRU reduction

In order to reduce the overall radiotoxic inventory of a 100 GWe reactor park it is important to emphasise the additional nuclear facilities which have to be made available.

Fuel cycle facilities

If the P&T strategy intends to improve the radiotoxic inventory situation on world-wide scale, additional industrial reprocessing plants totalling 3500 *tHM*/*y* should be built and operated.

However, in a first phase only the necessary chemical facilities for separation of MAs from HLLW have to be designed and constructed around existing reprocessing plants.

A 100 *GWe* park will discharge annually 1.54 *t* Np, 1.56 *t* Am, and last but not least 94-180 *kg* of Cm. The corresponding fuel fabrication outputs are: 60 *tHM* U-Np fuel and 60 *tHM* U-Pu-Am fuel with 2.5% MA concentration. Provisionally a storage capacity of 3 to 5 *t* Cm would be needed to cover, e.g. a 30 year production period. All these facilities would preferably be constructed on the reprocessing site in order to minimise transportation of these highly active concentrates. After conditioning and encapsulation a transfer to a dedicated reactor site for transmutation should in principle be possible. However, if multi-recycling of the targets is required, either the irradiation facilities should be installed near the existing processing units or dedicated (e.g. pyrochemical) reprocessing facilities should be erected near the reactor buildings.

Recycling scenarios in a power reactor grid

Research conducted in recent years has helped to clarify the feasibility of recycling actinides (mainly Am and Np) in the different types of reactor (FR, PWR) and in different recycle modes (homogeneous or heterogeneous).

Plutonium recycling and multi-recycling have been thoroughly studied. However, to reduce the overall radiotoxic inventory, it has been emphasised that any Pu multi-recycling strategy must consider the essential goal of not increasing the production of minor actinides. Theoretically, the advantage of FRs seems obvious.

According to present views and insights, a scenario with simultaneous management of plutonium and MAs seems possible in order to achieve equilibrium between production and consumption of Pu, Am, Np and Cm within the reactors. The radiotoxic inventory of the wastes generated by advanced reprocessing of these reactor fuels would significantly be reduced. The overall losses in the different reprocessing and partitioning operations would represent the minimum radiotoxic inventory of the waste resulting from the nuclear electricity generation. Several reactor park compositions with balanced reactor types can be considered.

The following reactor park compositions have been proposed in this study:

- THERM 100% PWRs, some of which are used for the multi-recycling of Pu and MAs.
- MIX 1 70% PWR-UO_2 and 30% FR capacity.
- MIX 2 70% PWR-UO_2, 10% PWR-MOX, 20% FR capacity.
- FAST 100% FRs of the EFR type.

Since these reactor systems all have the same installed capacity, they are said to be in equilibrium in two respects.

1. The characteristics of these systems (proportions of the different reactors, actinide contents etc.) are determined so that equilibrium exists between actinide production and consumption. In the reactors, the actinide inventory remains unchanged. Only the losses in the different cycle operations (fabrication, reprocessing) represent the ultimate wastes intended for disposal.
2. However, since the PWR MOX and FBuR incinerator reactors cannot consume all the actinides loaded in a single passage, this obviously implies a change in the isotopic composition of the fuel during multi-recycling.

Isotopic equilibrium is reached after about five to six cycles, and considering the inter-cycle times (cooling, reprocessing, fabrication), this corresponds to at least 50-70 years depending on the intercycle cooling time. Nevertheless, it must be emphasised that mass equilibrium between actinide production and consumption is always achieved in the first five cycles and before isotopic equilibrium is reached.

The reactor system consisting exclusively of PWRs (THERM) has the following features:

- The recycling of MAs must be dissociated from that of Pu, with some of the PWR reactors recycling Pu in MOX and others dedicated to recycling the MAs.

The characteristics of the other types of reactor system considered are as follows:

- All the PWRs and FRs have an electrical capacity of 1 450 *MWe*. The burnup of PWR UO_2 is 55 GWd/tHM.
- For the MIX 1 and MIX 2 scenarios, the FR cores correspond to the CAPRA reference oxide, with initial enrichment of 45% and core residence time of 855 EFPD.
- For the FAST scenario, the cores correspond to the reference EFR with zero regeneration gain (configuration with two axial blankets of 5 *cm* and one ring of radial fertile assemblies), with initial enrichment of about 20% and core residence time of 1 700 EFPD.

Neptunium is recycled homogeneously and mixed with the fuel in the core. Americium is recycled heterogeneously in the form of targets placed in the first peripheral ring of the core.

Curium is assumed to be stored for a century to allow the ^{243}Cm and ^{244}Cm to decay to Pu. This Pu is then recycled with the standard Pu flux. The residual curium (essentially ^{245}Cm) is recycled with americium in the targets.

Only the losses during reprocessing and fuel cycle operations are considered with assumptions of 0.1% for Pu and 1% for MAs.

Results

The radiotoxic inventory, expressed in *Sv/TWhe*, going to the wastes, and their changes over time are graphically shown for the different scenarios, and are compared with the radiotoxic inventory resulting from a reactor system with the same capacity consisting exclusively of PWR UO_2 managed in open cycle, i.e. with all the irradiated fuel discharged in the wastes. The reduction of radiotoxic inventory in the wastes are found to be similar for all the scenarios and close to a factor of 100.

Table 5**. Radiotoxic inventory reduction factor due to MA recycling as a function of disposal times in comparison OTC option**

Time (years)	100	1 000	10 000	100 000	1 000 000
MIX 1	74	72	77	130	82
MIX 2	71	75	84	140	80
FAST	110	100	100	150	150

If Curium is not recycled, which is the present proposal, the reduction factors become much lower and range between 7 and 24 during the first 10 000 years. Beyond that period, the daughter nuclide of ^{244}Cm, ^{240}Pu has decayed and the reduction factors become more favourable.

One important result of all the scenarios examined is that the Pu+MA inventory in the cycle can be stabilised. but the actinide mass inventory in the reactors increases steeply from 580 kg/GWe for the THERM case to 3184 kg/GWe for MIX1, 3 289 kg/GWe for MIX2 to reach 6721 kg/GWe for the FAST case.

Risk and hazard assessment over time

While the radiotoxic inventory is a physical-biological concept intrinsically based on the laws of radioactive decay and the radiological damage due to a quantity of radioactivity incorporated in the human body, the risk and hazard concepts on the contrary rely on the extent of conditioning and packaging of waste streams, on the long-term behaviour of waste packages in geological media and on the routes which could be followed by radioactive releases on their return to the biosphere and to mankind.

- In a first phase one has to assess the direct radiological impact of additional fuel cycle operations.
- The second step is the radiological assessment of the waste types created by the AFC operations.
- Finally the hazard assessment over time is closely related to the repository design and location.

Direct radiological impact of nuclear power production

The statistical data on the collective dose to the workers suggest that compared with the RFC, the AFC with P&T would only induce a moderate increase of the dose to man in the fuel cycle activities, and particularly to those in fuel and target fabrication. However, appropriate measures must be taken to reinforce shielding, especially against neutrons, throughout the entire recycling facilities, and this will significantly increase the overall investment cost.

In the OTC scenario, uranium mining and milling constitutes 90% of the total regional dose. In the RFC scenario, the impact of reprocessing on the regional dose amounts to 10% in the UNSCEAR study and 28% in the Cogéma study. If the AFC should become an industrial reality in those countries having a reprocessing infrastructure, the contribution of the MAs and FPs separation and conditioning operations would slightly increase the overall radiological impact on the environment but not modify the overall picture.

Impact of P&T on waste management decisions

The potential impact of P&T is an improvement of the long-term hazard but it requires additional actinide handling facilities and does not eliminate the necessity of geological disposal.

- Without reprocessing, P&T cannot be implemented.
- Partitioning of MAs from HLLW is possibly a first step in the gradual decrease of the radiotoxic inventory of vitrified HLW.
- Long-term storage of partitioned MAs waste fractions and long lived fission products will be necessary since special reactors have to be developed for transmutation.
- Partially "incinerated" or "deactivated" separated actinide or fission product targets will need special preparation and conditioning before disposal.
- The risk of contaminating the geosphere will be decreased if the conditioning of the toxic radionuclides is improved (e.g. by using ceramic matrices or improved glass compositions for the separated MAs).
- The fraction of radionuclides involved in fuel cycle and waste management activities will shift from mainly disposed materials in the OTC to mostly stored inventories as shown in Table 6. Compared to these reactor and facility inventories, the weight of waste discharged annually by a reactor park of 100 GWe-0.8 year (= 700 TWhe) would be drastically decreased in comparison with the OTC as shown in Table 7.

Table 6. **Mass of transuranic elements in reactor park (tHM/100 GWe)**

Elements	UO_2	MIX1	MIX2	FAST
Np	2.9	7.3	7.7	3.3
Pu	52.2	249.2	257	620
Am	2.3	54	56.1	41.6
Cm	0.6	7.9	8.1	6.8
Total	58	318.4	328.9	671.7

Table 7. **Annual discharge of TRU wastes (tHM/100 GWe-0.8 Year)**

Elements	UO_2	MIX 1	MIX 2	FAST
Np	1.155	0.021	0.022	0.0056
Pu	22.78	0.076	0.056	0.111
Am	1.152	0.075	0.0798	0.0354
Cm	0.126	0.042	0.040	0.018
Total/year	25.21	0.214	0.198	0.17
Total/30 year	756	6.42	5.94	5.1

Impact of P&T on waste characteristics and management

In the OTC option with a continuous nuclear power production of 320 *GWe*, additional large repositories will be necessary for a single type of "waste" form. If the maximum content of a repository is 70 000 *tHM* spent fuel like that of Yucca Mountain in USA, then without any reprocessing, every 13 years a new repository of that size or equivalent will have to be constructed. With partial reprocessing (3 900 *tHM/y*), the interval is increased from 13 to 25 years and vitrified HLW together with Medium Level Waste forms will have to be disposed of. The repositories will have to be adapted to the nature of the waste types and the size of the national or the regional nuclear power capacity.

In an AFC strategy the separation of Pu, MAs and LLFP will create a multiplicity of waste forms which could be conditioned according to the required chemical or metallurgical properties. A better conditioning will decrease the long-term risk and will allow in the mean time the development of transmutation technologies which are the only route to long term hazard reduction.

In the MIX 1, MIX 2 and FAST options, the equilibrium TRU inventory in the reactor and nuclear facilities amounts to 98% of the total amount of actinides involved in the fuel cycle, only one percent is discharged as waste. However, this strategic option implies the replacement of 20% or more of the present LWRs by FRs, and a complete recycle of recovered plutonium. Apart from the construction of dedicated transmutation and incineration facilities, this option also implies the construction of specialised fuel fabrication facilities capable of processing 1 490 *tHM* UO_2 fuel, 236 *tHM* LWR-MOX fuel, and last but not least the construction of 20 FRs with an actinide throughput of about 100 *tHM* per year.

Conclusions

- Fundamental R&D for the implementation of P&T needs long lead times and requires large investments in dedicated fast neutron spectrum devices (FRs, ADS which may complement or substitute for operating LWRs), extension of reprocessing plants and construction of remotely manipulated fuel and target fabrication plants.

- Partitioning facilities for MAs and some long-lived fission products could be designed and constructed as extensions to the existing reprocessing plants.

- Partitioning methods for long-lived radiotoxic elements have been developed on a laboratory scale but much work is still to be done on improving and scaling up the partitioning technology in order to make it compatible with industrial reprocessing practices.

- The short-term impact of partitioning would be to reduce long-term radiotoxic inventory of the resulting HLW at the expense of an increase of the operational requirements for the nuclear facilities concerned.

- Fast Neutron-spectrum devices (dedicated FR or ADS facilities) are more efficient than current LWRs for recycling and transmuting long-lived radionuclides. The ADS might play an important role in "incinerating" the actinides and some FPs from the spent LWR-MOX or FR-MOX fuel in order to reduce the long-term radiotoxic inventory of the wastes.

- Recycling of Pu+MAs in a composite reactor park could stabilise the TRU inventory of an electro-nuclear park. Multiple recycling of TRUs is a long-term venture which may take decades to reach equilibrium TRU inventories.

- Conditioning of separated long-lived nuclides in appropriate matrices which are much less soluble than glass in geological media, or which could serve as irradiation matrix in a delayed transmutation option, is a possible outcome for the next decades.

- P&T will not replace the need for appropriate geological disposal of HLW, irradiated TRU concentrates and residual spent fuel loads from a composite reactor park.

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