EXECUTIVE SUMMARY

Partitioning and Transmutation (P&T) has gained interest during the past decade and the OECD Nuclear Energy Agency has included P&T in its programme of work since 1988. The information exchange meetings are a key component of this international P&T-project, aiming at giving experts a forum to present and discuss current developments in the field.

The Fifth Information Exchange Meeting, hosted by the Nuclear Research Centre SCK•CEN and co-organised by the European Commission, was held on 25-27 November 1998 at Mol, Belgium with more than 120 participants from 15 countries and 3 international organisations. The programme included 43 oral presentations and 12 posters covering introductory papers, partitioning, fuel and target fabrication, cross-section data, reactor irradiations, comparisons between reactors and accelerator-driven systems as well as long-term radiological impact evaluations. During this meeting, the report *'Status and Assessment Report on Actinide and Fission Product Partitioning and Transmutation'* was presented as conclusion of the first phase in the OECD-NEA P&T study.

This Fifth Information Exchange Meeting could be characterised by two main directions; first of all, a more integrative view on P&T was observed where consensus on the way to perform P&T was achieved but where questions were raised on the added-value of P&T in the nuclear fuel cycle (and the most appropriate way to achieve it). Secondly, the breakthrough in partitioning of minor actinides was achieved on lab-scale at pre-set performances.

This meeting also highlighted issues for the near future. The comparative assessment of transmutation in fast reactors and accelerator-driven systems, addressing also the impact on the needed reprocessing, is one of the main issues to be studied in the near future to answer questions on performance and needed developments for P&T.

This summary addresses the main items and discussions held during this meeting as a general introduction for the interested reader, referring to the respective papers for a complete and comprehensive view on the mentioned subjects.

Partitioning

Partitioning is the first cornerstone in each strategy of P&T and strongly defines the boundary conditions of the minor actinides (MAs) and/or long-lived fission products (LLFPs) transmutation, e.g. solid fuel or molten salt, batch-processing or on-line processing, remaining fraction into high level waste. This fraction of MAs is still the most important because the MA content in the waste vector is still above the solubility limits of MAs in the surrounding geosphere [p. 463][a]. Further reductions of this MA fraction can be achieved by the interplay of P&T in dedicated processing

---

[a] Reference to papers will be given by the page number in these proceedings.

facilities combined with dedicated reactors or accelerator driven systems (ADSs) and could have a definite impact on the radiotoxicity content of a repository. These considerations will be reflected in upcoming discussions which will be apparent in the coming years.

The discussions also addressed the hydrometallurgical process and the pyrochemical process as both potential partitioning methods with their respective merits and drawbacks. The hydrometallurgical process for the separation of Am, Cm and Lanthanides, is becoming a well-known technology making upscaling to prototype pilot facilities possible. However, the final performance of radiotoxicity reduction after P&T could be strongly limited by the hydrometallurgical process because of cumulative losses in a multiple recycling scheme. Severe reliance on very high efficiencies of separation is therefore required. The pyrometallurgical process, being less mature and facing a renewal of interest, could be more adapted to an in-line, and as such, multirecycling P&T process of highly irradiated and short cooled fuel and targets but this route will impose the burden of radiotoxicity reduction on the transmutation step. The following text gives a summary of the presentations related to partitioning and highlights the results of current research.

Four main programmes or research axes were mentioned within the research on partitioning of MAs.

A *dual solvent extraction process* has been proposed by JNC [p. 113] and is based on the U-Pu-Np separation from dissolver solution by the Advanced PUREX process and subsequently the additional recovery of Am and Cm from the high level waste by the Advanced TRUEX process. Actual R&D have been pursued in hot-lab conditions at the Chemical Processing Facility in Tokai-works using real high-level liquid waste (HLLW). Actinides (An) and Lanthanides (Ln) were successfully separated from the highly active raffinates and application of some sort of ‘soft’ donor ligands, purified CYANEX 301 and ODP, proved excellent Am/Eu separation. Electrolytic extraction tests proved to be very promising to separate several rare metals including some LLFPs.

The *NEWPART-programme* [p. 53, 121, 141], within the 4th European Framework program, focused on partitioning methods for Am and Cm within a PUREX raffinate (Np separation was considered within an improved PUREX process). Within this programme a two-step process has been targeted; the first step (DIAMEX process) aiming at the separation of trivalent actinides and Lanthanides from fission products (FPs) and, in a second step, (CYANEX process) the group separation of An(III)/Ln(III) by selective extraction of An(III) which is the most difficult goal to achieve. The SESAME process [p. 161] could complement the previous steps by separation of Am from Cm. Definite progress has been made within the DIAMEX process [p. 153] in the understanding of the basic chemistry of malonamide extractants in their ability to react with trivalent actinide and lanthanide nitrates and active tests are foreseen in the near future. A real breakthrough has been reported by the independent confirmation by two institutes of the very high separation factors (6000) of Am and Eu using the CYANEX 301 as extractant. Another potential breakthrough has been booked in the An(III)/Ln(III) separation using BTP (pyridine-bis-1,24-triazines) resulting in separation factors of Am over Eu of 140 in high nitric acid concentrations. In conclusion, confidence has increased in that the processes based on the NEWPART programme can be developed to reach the goals required for modern efficient industrial MA partitioning processes.

The *Four Group Partitioning process* [p. 131], developed by JAERI, separates the elements in HLLW into four groups: transuranium elements (TRU) using DIDPA, Tc-platinum group metals (PGM), Sr-Cs and others. Separation efficiencies obtained experimentally with simulated HLLW and the estimated recovery through whole partitioning process attain or even pass the target recoveries to make the potential radiotoxicity of a vitrified HLW after storage for 1 000 years comparable to that for naturally occurring minerals containing 3 wt% of U. As for the separation of Am and Cm from Lanthanides, it is possible to reduce their lanthanide content to less than 25 wt% with 99.99% separation efficiency from actinides. The radiation resistance of DIDPA is about 7 times higher than TBP and its degradation products have no harmful effect on the TRU extraction resulting in less secondary waste generation due to solvent retreatment. The four group partitioning process reduces the volume of waste by a factor of at least 3.3, compared with that of the vitrified HLW, and the volume of cemented waste is only about 20% of that generated in the PUREX process. Moreover, an α-activity concentration of the cemented waste becomes lower than 1 GBq/ton and its shallow land disposal might be a possibility. Demonstration of the four group partitioning process with real HLLW is now in progress.

Research activities in Russia [p. 77] are focused on the separation process of heat emitting nuclides (^{137}Cs and ^{90}Sr) and minor actinides by use of cobalt dicarbollide. The upscaling test of the process has shown to be very successful from the process- and safety points of view. An industrial extraction facility using cobalt dicarbollide has been operated for several months at the Mayak plant and showed excellent results with real HLW. International discussions are underway to restart the process.

The *pyrochemical partitioning processes* have been presented by Japan [p. 169], Russia [p. 179] and the Czech Republic [p. 187]]. In Japan, CRIEPI studies a pyrochemical partitioning process involving denitration, chlorination and multistage extraction. In this process, more than 98% of alkali metals are separated in the denitration step. Almost all of the other elements are converted from oxides into chlorides by a chlorination step. Separation experiments by means of multiple extraction were also carried out in a LiCl-KCl/Bi system, and more than 99% of each TRU was recovered. The chemical compounds used in the process, such as Cd, Bi, Pb, Li chlorine gas can be recycled. As a result, this process is expected to generate less secondary radioactive waste in comparison with the aqueous processes and could require rather relatively compact facilities. However the corrosion and material problems associated with the industrial use of pyrochemical processes needs to be carefully investigated.

In Russia [p. 179], the use of molten salt reactor (MSR) technology was started in the second half of the 70s. This MSR is seen as a second component in a multi-strata fuel cycle where the MSR incinerates part of the plutonium together with MAs, and if needed, some FPs. The results of recent studies have demonstrated that a broad range of MSR-processes with PuF_3 and MAs in liquid 2LiF-BeF_2 is conceptually feasible. Reactor-loop tests of the radiation stability of fluoride molten-salt fuels have indicated that these fluoride salts could be the fuel for high temperature reactors. Experimental studies suggested that neither physical nor process-chemical obstacles have been observed in the way to implement MSR and related fuel cycle technologies.

In the Czech Republic [p. 187], dry reprocessing of fast reactor spent fuel has been based investigated. This method, developed in the former Czechoslovakia in the 70s and 80s, is based on the difference in the volatility of U, Pu and FPs in their fluoride forms. A pilot-plant technological line, called FREGAT-2, was constructed for the fluoride-technology verification through the experiments on reprocessing of spent fuel from BOR-60. At present, the research and industrial organisations in Czech Republic intend to use the experience for a national program on transmutation technology development.

Transmutation

The second cornerstone of P&T consists of the irradiation, and as such, transformation of the MAs and/or LLFPs in other nuclides having shorter life-times or even being stable. This transmutation involves the inclusion of the separated nuclides in a form which is compatible with the irradiation device used. Transmutation can be done in LWRs, in fast reactors (FRs) and in ADSs. The last two options provide a homogeneous, as well as a heterogeneous, mode of introduction of MAs in the system, but also provide the potential to use pyrochemical processes on-line. ADSs could be distinguished by their potential of surplus neutron availability, providing higher transmutation capability for LLFPs, and their potential of higher reactivity losses per cycle and thus higher transmutation rates achievable without hampering core safety characteristics. In general, transmutation studies involve a multitude of aspects (e.g. target fabrication and irradiation tests, reactor physics and nuclear data, safety studies) which will be briefly discussed in the next paragraphs.

Major research and development work is going on world-wide to address the *target fabrication* aspect. The transmutation of Am in existing LWRs and future fast systems has been the primary focus of much of this R&D [p. 197, 207]. The fabrication of such Am targets can be done according to different processes: powder mixing process, sol-gel method and the infiltration by an active solution of inactive pellets. The first two processes have been well developed and implemented in the fabrication of Pu-bearing fuels (MOX). However, when fabrication of highly radioactive materials (like Am, Cm or LLFPs) is considered, dust production and consequent radiation exposure to personnel and minimisation of secondary waste production could limit the applicability of these mechanical processes. Therefore, alternative fabrication routes have been envisaged. One should remark also that the application of current MOX-fabrication techniques for the fabrication of Am-bearing targets will demand adaptation of the fabrication parameters to obtain good quality pellets and taking into account the new wishes of the fuel designers to have the Am-loading to be distributed heterogeneously in the matrix, which is contrary to all present requests for industrial MOX fabrication.

One of these alternative fabrication routes involves the infiltration of radioactive materials (INRAM), a process developed by the Institute of Transuranium Elements (ITU) [p. 197]. This novel technique has been developed for non-nuclear ceramics and for isolation of radioactive wastes, and consists of the dissolution of the active material and its infiltration into a porous (non-active) medium. Such an infiltration process drastically reduces the number of production steps involving handling of highly radioactive materials. The infiltration process relies on the action of capillary forces to draw the solution into the pores of the host material. The INRAM method has been used in ITU to produce two fuel pins containing 11w% Am in spinel for the completed EFTTRA-T4 irradiation in HFR Petten. The Am distribution within the pellets was not as uniform as in the pre-fabrication tests, requiring further investigation to obtain the guaranteed homogeneity. ITU is currently developing a minor actinide laboratory to cover the needs of the ITU and its partners for the fabrication of fuels and targets in the framework of test irradiation programmes and to make tests for future industrial applications.

The *irradiation tests* EFTTRA [p. 207] have shown that extensive swelling (+5.2% in length; +6.7% in diameter; increase in volume about +18%) of the Am bearing fuel (micro-dispersed Am in a spinel-based fuel) took place, probably due to the damage of the matrix by FPs and/or accumulation of He resulting from α-decay. Next to this Am transmutation, EFTTRA also focuses on the Tc and I transmutation. As reported in previous conferences, no technical limitations to the use of metallic Tc as a target for transmutation are identified. A comparable programme on transmutation of Tc is being undertaken by JAERI [p. 223] focusing on the formation of a database of the Tc-metal target based on out-of-reactor experimental studies.

Nitride fuel is a promising candidate for transmutation of MAs because of its excellent thermal and neutronic properties. Recent progress of the research on the nitride fuel, which is the reference system for transmutation of MAs in JAERI [p. 213], includes the establishment of a database of the properties on nitride solid solutions of the UN-NpN-PuN system including thermal conductivity and evaporation behaviour. JAERI also introduced a new concept, “LINEX”, where actinide elements in spent nitride fuel are selectively dissolved as chlorides into the molten salt by electrolysis or oxidation, and where the actinide nitrides are synthesised from the chlorides by adding Li_3N, which is formed by the reaction of Li with $^{15}N_2$ gas released at the dissolution step of the spent nitride fuel. Experimental results show that 95% of the nitrogen could be recovered (difficult in the Purex process) which reduces the required price of $^{15}N_2$ enrichment to about $100/g which might be commercially produced using the present enrichment technology.

From the irradiation tests of (U,Pu)N fuel in JMTR, the nitride fuel shows excellent performance; the fission gas release was found to be very low, around a few percent, and there was no significant chemical interaction between fuel and cladding, at least up to the burnup of 5.5% FIMA. These results indicate that the ‘cold’ fuel concept indeed shows the advantages especially in lowering the fission gas release.

Basic and fundamental research remains one of the cornerstones of science and technology, and *cross-section measurements* and evaluations are key of many transmutation uncertainty assessment studies as the MAs are among the main contributors to reactivity and the nuclear data for these MAs are not so well known as for the main actinides (U,Pu). The ongoing measurement-campaigns, guided by the OECD-NEA High Priority Data Request List, focus especially on those nuclides of primary importance; e.g. ^{237}Np, ^{242m}Am, ^{99}Tc and ^{129}I, to clarify inconsistencies between cross-section libraries and refinement of resonance cross-section data. Reported work involved the measurement of the ^{242m}Am thermal neutron capture cross section at ILL-Grenoble [p. 231], indicating agreement with the ENDF-B/VI value of 252 barns, and as such, leaving open the option to transmute ^{241}Am by an intense flux of thermal neutrons. Very precise cross-section measurements for ^{237}Np and ^{99}Tc were performed at the GELINA installation [p. 239]. These measurements focused the total cross section and Doppler broadening for ^{237}Np resulting in a preliminary list of new resonances under 120 eV. Comparable measurements on ^{99}Tc for radiative capture and total cross section in the resonance region [p. 269] resulted into more precise and consistent information concerning this resonance region of ^{99}Tc. The use of the adiabatic resonance crossing method was confirmed by experimental measurements in the TARC experiment in CERN [p. 249] and brought another approach to the evaluation of ^{99}Tc cross sections. Fission cross section ratios of MA nuclides (^{237}Np, ^{241}Am and ^{243}Am) relative to ^{235}U in a fast neutron energy regions have also been measured by JNC, Kyoto University and measurements of keV-neutron capture cross sections of rare earth nuclides have been performed at the Tokyo Institute of Technology [p. 323].

The transmutation of MA is coupled on a proper use of Pu. The Pu being an energy carrier and its recycling in a FR with U allows use of almost all the initial U and increases the energetic gain of U inventories by several decades. Today, the *studies for an efficient transmutation* of MAs are linked by different ways of utilising Pu (in LWR, in FR). Based on scenarios of Pu recycling, the transmutation of MAs was studied and different possibilities compared [p. 283, 289, 303, 315]: the first one using a dilution in LWR-fuel; secondly, the use of Am and Cm in a ‘once-through’ irradiation in FR; and thirdly, the multirecycling of those minor actinides in a FR. Other schemes are possible as well, but mostly rely on previous basic schemes.

By using the dilution (homogeneous or heterogeneous) scheme and by following a multirecycling strategy of the (Pu+MA)-fuel in LWRs, one can conclude that all mass flow rates in the fuel cycle have to be increased, in particular, the Cm masses generated are three times higher. However, the fabrication of the needed targets (homogeneous or heterogeneous) is not feasible in current fuel fabrication facilities and dedicated facilities, favouring the heterogeneous recycling mode, should be envisaged. Another approach could be the delay in (Am+Cm)-target fabrication using the Cm decay to Pu, and as such, facilitate the fabrication (anyway in automated cells).

The use of specific MA burners, called *dedicated reactors* [p. 315], with important MA loads (ideally 100% of the reactor fuel) would allow the reduction of specific fuel-cycle operations (fabrication, handling, storage and reprocessing) and limit safety countermeasures to a small fraction of the reactor fleet with respect to the homogeneous option. Those dedicated burner reactors have to take account of the core characteristics by increasing the MA-fraction, e.g. smaller Doppler and void reactivity coefficients. Therefore, the use of small cores, increasing the leakage effects, and the use of ‘cold’ fuel, e.g. nitride fuel, are main axes if one considers the use of solid fuel type burner reactors. Based on the lower reactivity losses and as such possible longer fuel residence times in combination with multirecycling, this scheme could well increase the transmutation potential.

Comparable studies have been performed by JNC [p. 323] and it was found that the mixed hybrid MA and FP loading method (where Np nuclide is dispersed uniformly in the core and target assemblies containing Am, Cm, rare earth nuclides and FP are loaded into the radial shield region of a fast reactor) have a great potential to achieve effective transmutation of both MA and FP without serious drawbacks in terms of core performance.

Metal fuel Fast Neutron Reactors show suitable performances for the consumption of MAs, and their pyro-reprocessing is quite compatible with the recycling of MAs [p. 335]. In this case, the MAs content in the feed is limited to as low as a few weight percent to keep the core safety parameters acceptable and facilitating a simple casting fuel fabrication method. The results of the studies show that the Pu and MAs from LWRs can be consistently consumed without further accumulation by establishing the same scale of FR power generation and its fuel cycle

The above evaluations of dedicated FRs for MA transmutation have indicated that contradictory trends have to be combined. The transmutation of LLFPs in those reactors, placed in the blanket region, further reduces the degree of freedom in the design of such dedicated FRs. The use of an external neutron source, e.g. by the use of spallation neutrons, relaxes in some respect this degree of freedom and is a valuable means to alleviate these contradictory trends. Research on ADS is currently conducted world-wide and is mostly focused on system design studies.

In the past years a number of proposals for ADS applications have been presented, varying from an energy amplifier with lumped Th/^{233}U fuel and fast spectrum with lead coolant, to fission product and actinide transmuters with fast and thermal spectra. The main argument used for these ADS concepts is usually improved safety compared to critical reactor systems, but this argument is merely of secondary nature. The main arguments for ADS rely on neutron economy and fuel flexibility. A comparison [p. 343, 355, 361] of similar ADS and critical reactor system designs shows an improvement of the safety characteristics of an ADS, caused by the lower required fissile enrichments. However, it is not clear that if in a fast spectrum ADS, recriticality can be avoided in all circumstances during core damaging accidents. In relation to kinetic behaviour of ADS compared to critical reactor systems, a transient over power without scram or a source shutdown accident results in a six times faster and 50% higher power jump in critical systems than in an ADS. It shows, in particular, that the ADS gives a much slower response to any type of feedback over the reactivity. However, in case of loss of flow, the ADS shows a less favourable behaviour because the resulting reactivity variations are smaller in comparison with the subcriticality.

The ADS concepts are mostly specially designed for transmutation purposes as dedicated transmuters to be deployed in the P&T-cycle of the double-strata fuel cycle. Most of the presented design studies focuses on three categories; sodium cooled solid fuel fast reactor technology, liquid metal cooled options favouring harder neutron spectra and reducing the effects of positive void reactivity coefficients and technological limits, and finally, the concepts relying on molten salt technologies. Accelerator-driven Transmutations Systems (ADTS) are being investigated by JAERI [p. 383] according to the above mentioned system categories. Conceptual designs are currently being studied [p. 401] and are based on fundamental studies in reactor physics [p. 393, 435, 453], materials, basic nuclear data and accelerator physics [p. 375] to be used eventually in detailed design considerations. The molten salt option has been investigated by different countries, including for instance, the Czech Republic [p. 443]. The molten fluoride salt fuel material is in the form of a fluoride salt Ac-Fluoride dissolved in a molten salt carrier whose composition is a mixture of ^{7}LiF and ^{9}BeF$_2$.

Using Pu instead of ^{233}U as fissile material in an ADS, e.g. as a burner of multirecycled Pu with high contents of ^{240}Pu and ^{242}Pu, results in a more complicated burnup behaviour where a strong increase of reactivity with burnup is observed. These results indicate problems with subcriticality during burnup in such a Th/Pu system [p. 413] and this may lead to a need for active control devices in this ADS, e.g. control rod systems.

A very important and distinctive part in all ADS-designs relates to the accelerator reliability. The requirements for the accelerator [p. 375] are new for accelerator physicists. Not only the beam energy and intensity are rather high-end, but the reliability of the beam, especially reduction of the troubling beam trips, are key issues for future developments. A reduction with at least a factor of 100 of those beam trips should be obtained. Material and technological problems related to the beam window are very important issues for the future. Future R&D should be focused on reducing the need of overdesign of these.

Conclusions

The five successive OECD-NEA information exchange meetings which took place from 1990 until today have led to the establishment of a very effective information exchange mechanism which has contributed to a companionship of scientific institutes world-wide.

The international co-operations among the countries inside Europe has been catalysed by the EURATOM program during the Fourth Framework Programme. The European Commission foresees to amplify the research activities in the field of P&T under the Fifth Framework Programme (1998-2002).

The International Atomic Energy Agency (IAEA) conducts a more basic program aiming at the coverage of P&T activities from non-OECD-NEA countries and focuses on the safety, environmental and non-proliferation aspects of P&T.

The OECD-NEA P&T status and assessment report is currently the most extensive data base for P&T and comprises a critical analysis of the impact of P&T on the nuclear fuel cycle and waste management. P&T can conceptually reduce the long-term radiotoxicity but it will require the development, construction and operation of new fuel cycle plants (fabrication and reprocessing) and new dedicated reactors (FRs and/or ADS devices), and it will amplify the specialised intermediate storage plants. Eventually geologic disposal structures for high active wastes remain inevitable.

Future R&D in the field of P&T will address especially this interaction or comparison of FRs and ADSs and their respective roles in the future nuclear fuel cycle. The developments reported from different countries will result in new insights and maturity in P&T. OECD-NEA will contribute to these developments and is starting, in the beginning of 1999, the second phase of the project on P&T.

This second phase will address the comparative assessment of ADSs and FRs and will focus on the added value in future advanced fuel cycles, for instance the ‘double-strata’ fuel cycle. This study will especially address the requirements, feasibility and reliability of those ADSs as new transmutation devices and will focus on the needs of development, lead time for introduction in the fuel cycle, and a cost/benefit evaluation in comparison with FRs. This phase is planned to last two years and will result in the publication of an expert group report which will be presented during the sixth information exchange meeting, to be held in the year 2000.