

EXECUTIVE SUMMARY AND SCOPE OF THE REPORT

Implementation of partitioning and transmutation (P&T) technology is intended to reduce the inventories of actinides and long-lived fission products in nuclear waste. Implementation of such technology may decrease the hazards associated with pre-disposal waste management and with physical disturbance of a waste repository, e.g. by human intrusion, but it is not seen universally as a short-term alternative to contemporary fuel back-end policies nor as a means of avoiding the need for a deep geologic repository facility for long-lived waste from contemporary operations.

Nevertheless, there is a widely held view that technologies for reducing these inventories may have the potential for alleviating the waste disposal problems of the fuel cycles designed specifically to exploit them. The associated studies would serve, at least, to advance the science of nuclear energy and attract capable young researchers into the field in preparation for a possible swing of global public opinion towards accepting nuclear power. In addition, studies of laser and accelerator technology, for example, may well provide spin-offs for other fields of science and technology.

Much of the current work in this area is at an early stage of development. It is difficult to predict the outcome, or associated timescales, of current research and development (R&D) or of the costs associated with development and operation of technologies under study. The structure of a total system study and the related requirement for information are outlined in this report but the primary focus, for the present, is on review of the potential technologies for partitioning and transmutation together with a critique of development status and prospects for eventual industrial application.

It is the purpose of this report to provide an authoritative analysis of the technical, radiological and, as far as possible, the economic consequences resulting from the proposed partitioning and transmutation operations on the present and future fuel cycle options. The present report is subdivided in a general part for non specialist readers and a technical systems analysis discussing the issues in partitioning, transmutation and long-term waste management:

Part I: General overview

- Introduction on the international programme co-ordinated by the OECD/NEA.
- A general overview of status and expectation of P&T technology and a comparative analysis of the issues associated with the different fuel cycles.
- A critical evaluation which tries to identify the strong points and weaknesses of the partitioning and transmutation strategy.
- General conclusions and recommendations on future priorities in development.

Part II: Technical analysis and systems study

- An analysis of the potentials of different partitioning methods, either as extensions of the PUREX process or as an autonomous pyrochemical fuel processing method.
- An extended technical discussion on the possible transmutation options for minor actinides (MAs) and selected fission products (FPs) by light water reactors (LWRs), fast reactors (FRs) and hybrid transmutation devices (accelerator-driven transmutation systems (ADS)) including:
 - a feasibility assessment of minor actinide fuel and target fabrication technology and safety;
 - a detailed overview of the issues involved in irradiating minor actinide-containing fuels and targets in current pressurised PWRs (homogeneous and heterogeneous recycling);
 - a comprehensive overview of the transmutation potential of minor actinides in fast reactors as mixed oxides (MOX), metal, and inert matrix fuel forms or targets;
 - a preliminary analysis of the accelerator-driven transmutation systems potential, the issues involved and the current projects;
 - a comparative analysis of the transmutation potential for selected long-lived fission products in different reactor types and accelerator-driven transmutation systems.
- A description of the major research and development programmes carried out in Japan and France.
- A comprehensive discussion of the impact of partitioning and transmutation on risk assessment and waste management, especially the long-term impact of currently proposed methods.
- An analysis of the different transuranic element inventories according to the strategic option taken in the evolution of nuclear power plant (NPP) types (light water reactors, fast reactors, accelerator-driven transmutation systems, etc.) in a future nuclear electricity production grid.
- A short overview of the current waste disposal concepts and the possible impact of partitioning and transmutation on the repository structure and characteristics.
- A discussion of the long-term radiological and radiotoxic impact of the partitioning and transmutation operations.

General overview

A general overview is given of the current fuel cycle status and nuclear materials management as it is practiced today. The expectations of new options that partitioning and transmutation might offer, as well as their limitations, are discussed in an introductory chapter. Additional recycling operations of fissile and fertile materials (uranium, plutonium), minor actinides (neptunium, americium, curium) and some selected fission products (^{99}Tc , ^{135}Cs , ^{129}I , etc.) are presented as an introduction to this complex matter for a non-specialist reader.

A comprehensive analysis is given of the three fuel cycle options: the “Once-Through Cycle” (OTC), the “Reprocessing Fuel Cycle” with uranium and plutonium recycling (RFC) and the “Advanced Fuel Cycle” with partitioning and transmutation (AFC) within the framework of a nuclear electricity production grid of, for example, 100 GWe. Particular attention is devoted to the open issues where management decisions have to be taken in order to minimise the radiotoxic legacy resulting from nuclear energy production.

Reference data on irradiated UO₂ and mixed oxide fuels for a typical range of irradiation conditions are summarised in composite overview tables. Extensive nuclear data tables of the radionuclide inventory of spent fuel are given in Annex E of this document. The data provided are mean values from computer programmes accepted throughout the nuclear community as reference figures for the calculation of radiotoxic inventory over extended periods of time.

Critical evaluation and general conclusions

A series of critical evaluations and general conclusions drafted by the expert group have been added to the general part of the report in order to highlight the key issues which may help decision makers to establish the priorities in future research and development work, to underline the potential of the partitioning and transmutation option in waste minimisation and to draw the limits of this strategy in a future nuclear era.

Technical analysis and systems study

Partitioning

Reprocessing of spent light water reactor fuel is considered as a reference scenario which is a necessary preliminary to undertake additional partitioning of minor actinides and selected fission products in the first step of the advanced fuel cycle.

The aqueous separation techniques, compatible with the well known PUREX process, are briefly described and illustrated by their flowsheets. The possible and desirable improvements for transuranic element extraction in the PUREX process to recover ²³⁷Np, and the associated minor actinide extraction processes “TRUEX”, “DIDPA”, “TRPO”, “DIAMEX”, “TPTZ”, “SESAME” and the latest “CYANEX 301™” processes to recover americium and curium, are briefly described with comments on their merits and limitations. Their impact on the conventional waste treatment processes are briefly discussed, especially the secondary waste generation. An extensive bibliography on partitioning is given in Annex D.

The separation of some fission products requires additional operations within the head-end of the PUREX process since there is segregation between insoluble and soluble fission products. The recovery of ¹²⁹I and ¹⁴C from dissolver off-gas are examples of how the impact of reprocessing on the environment can be reduced. For two other long-lived radionuclides, i.e. ⁹⁹Tc and ⁹³Zr, their separation from tributylphosphate (TBP) streams could also reduce their environmental impact of the wastes.

Over the last two decades considerable progress has been made in the selective extraction of minor actinides and fission products but the technical maturity of each of these methods, as summarised in an overview table, is very variable.

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 till 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 till transmutation techniques become available.

Americium could ultimately be separated as individual element and recycled into special LWR-MOX or FR-fuel to be partially transmuted or could be 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 curium isolation 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.

In a perspective of increased burnups and shorter cooling times, the development of non-aqueous pyrochemical partitioning methods deserves special attention. Pyrochemical methods in molten chloride salt (LiCl-KCl or NaCl-KCl) baths have been tested in laboratory and hot pilot scales in order to separate uranium, plutonium and minor actinides from fission products. A combination of electrolytic separation, chemical precipitation and reductive extraction might be used in the future either as pre-separation steps before aqueous reprocessing or perhaps as an autonomous process for dry reprocessing of high burn-up spent fuel with shorter cooling time. Successful achievement of this technology may shorten the time interval for a complete recycle sequence.

Transmutation

The Transmutation chapter covers a number of activities: fuel and target fabrication, transmutation by neutron capture and “incineration” by fission.

Transmutation can be carried out in thermal reactors, in fast neutron reactors and in accelerator-driven subcritical reactors. The proper choice of each of these transmutation facilities derives from strategy studies which have to provide an optimum combination between the many diverse aspects of fuel fabrication and refabrication, availability of reactor types, transmutation/incineration yields, and last but not least, a significant reduction in overall radiotoxic inventory of the nuclear waste. Extensive studies have been undertaken within the European Union and Japan to provide a first strategic approach.

However these strategy studies are subject to political decisions and economic realities which cannot be forecast over the several decades or centuries necessary to achieve a steady state between input and output of fissile and fertile nuclear materials.

Fuel and target fabrication issues

The report treats in a factual way the problems encountered when introducing minor actinides into the nuclear fuel cycle. Fabrication of mixed oxide fuel with low (<2.5%) neptunium and americium loadings is a feasible option which does not require large investments in the light water reactor mixed oxide fuel (LWR-MOX) fabrication plants. However, the introduction of these minor actinides makes subsequent reprocessing and refabrication of fuel or targets more difficult because a large amount of ²³⁸Pu is present in the recycled plutonium stream.

Heterogeneous recycling of minor actinides in light water reactors is highly preferable since the targets can be managed independently of the light water reactor mixed oxide fuel and recycled in dedicated small-scale processing facilities. However the main problem which has not yet received a satisfactory solution is the recycling of ²⁴⁴Cm which is a strong heat and neutron emitter and cannot be handled in regular fuel fabrication facilities. Future work in the field of partitioning is required to provide a workable technology.

Fast reactor fuel has been produced for several decades and experimental fuels with large loadings of americium and neptunium have been fabricated in the frame work of the "SUPERFACT" programme. The irradiation of minor actinides in a fast neutron flux should preferably be achieved in inert matrices (aluminum oxide, magnesium oxide, etc.).

In order to further increase burnups and transmutation yields, new fuel forms are being studied, in particular metal alloy (of minor actinides, uranium, plutonium and zirconium) and nitride fuel. One of the advantages of using metal fuels is the possibility of forming a compact fuel cycle system based on the pyrochemical techniques, although substantial research and development efforts are still needed for its realisation.

The feasibility of nitride fuel is dependent on the availability of nitrogen-15 enrichment facilities and recovery techniques during recycling procedures. The application of pyrochemical processing to the nitride fuels may also facilitate its recycling.

Transmutation of minor actinides

The physics of transmutation/incineration of minor actinides shows that fast neutrons, either in fast reactors or in accelerator-driven systems, are preferable to a thermal one because of the more advantageous ratio between fission and capture cross-section. Moreover, all minor actinides are to a certain extent fissile in a fast neutron spectrum and contribute during irradiation to the overall energy production.

Comparative tables with calculated transmutation/incineration yields are given for homogeneously and heterogeneously loaded minor actinides in light water reactors fuelled with mixed oxide as well as for fast burner reactors and advanced liquid metal reactors. Most of the advanced

reactor concepts are still in the conceptual phase and the calculated yields have yet to be verified in representative conditions.

Minor actinide transmutation in fast reactor cores (liquid metal fast breeder reactors, advanced liquid metal reactors) has no serious drawback in terms of core performance, provided that the ratio of minor actinides to fuel remains small.

The design of fast burner reactors and minor actinide burner reactors (MABRs) needs further research and development because the effects on reactivity coefficients and kinetic parameters cast some doubt on their feasibility with high minor actinide loadings.

Recently hybrid type accelerator-driven sub-critical transmutation systems have been receiving increased attention world-wide as an alternative to conventional nuclear reactors. Accelerators, whether linear accelerators or cyclotrons, produce a proton flux which impinges on a heavy metal target which undergoes spallation. The spallation neutrons impinge in their turn on fissile and fertile nuclear materials, e.g. minor actinides, which undergo fission and capture.

The transmutation of minor actinides cannot be dissociated from the transmutation of plutonium, and any strategy in this connection has to take into account the depletion of minor actinides in targets or fuels and the simultaneous generation of minor actinides from plutonium.

Transmutation of long-lived fission products

Transmutation of long-lived fission products (^{99}Tc , ^{129}I) to short-lived or inactive nuclides has been investigated and led to the conclusion that it is theoretically possible but that higher fuel enrichments are necessary in light water reactors. The transmutation half-life varies from 15-18 years in fast reactors to 40-77 years in light water reactors. Irradiation of selected fission products in accelerator-driven systems or optimised fast reactors would make it possible to decrease the transmutation half-life. Intensive development of target and fuel assembly materials would be required (as in the case of Iodine), as well as refined isotopic separation in the case of some elements (e.g. Se, Zr, Cs, Sn, ...).

Current trends in partitioning and transmutation research

Taking into account the progress of the research and development programmes in Japan and in France, a special section and Annexes B and C deal with the current partitioning and transmutation activities in these countries. These programmes, in which many countries participate directly or indirectly, are undoubtedly the most important in the world.

Impact of partitioning and transmutation on risk assessment and waste management activities

In order to set out very clearly the impact of partitioning and transmutation on long-term radiotoxicity, a special section in the report is devoted to the significance and definition of radiotoxic inventory.

The radiotoxic inventory depends on the physical inventory of radionuclides in the various fuel cycles and on the effective dose coefficients of individual radionuclides. Graphs show the natural

evolution of radiotoxic inventories corresponding to the once-through cycle, reprocessing fuel cycle and advanced fuel cycle.

The role of partitioning and transmutation in waste management depends primarily on the minor actinide inventories to be handled in the advanced fuel cycle facilities. The section discusses the reactor strategy to be implemented in order to achieve a significant reduction of the radiotoxic inventory. Three representative cases are considered with various combinations of light water reactors loaded with uranium dioxide and mixed oxide fuel, and fast reactors loaded with mixed oxide fuel. A substantial proportion of fast reactors is necessary. In each of the cases examined, the radiotoxic inventory in the waste streams is reduced by a factor of about 100. However, the decrease observed in the waste discharges is accompanied by a steady increase of the radiotoxic inventory of the reactor and fuel cycle facilities operated over a long period of time to accomplish this waste minimisation.

Where all minor actinides are recycled including the curium fraction, the radiotoxic inventory reduction factor, compared with once through cycle, ranges from 77 to 100 after 10 000 years, and in the very long term (10^5 and 10^6 years) from 80 to 150.

If the curium fraction is not recycled, the radiotoxic inventory is reduced only by a factor of 7 to 14 at 10 000 years owing to the decay of ^{244}Cm into ^{240}Pu and ^{243}Cm into ^{239}Pu .

When making a global mass balance of the minor actinides involved in a long-term multiple recycling programme, the radiotoxic inventory of the nuclear materials in the reactors and fuel cycle facilities becomes overwhelmingly more important than the annual waste discharges. Various cases leave equilibrium inventories of several hundred tonnes of transuranic elements.

Based on data from different economic assessments, but principally relying on a comprehensive study carried out within the European Union's R&D programme on radioactive waste management and disposal, some very preliminary cost data for the advanced fuel cycle with partitioning and transmutation strategy are given in Annex F.

Waste management concepts

Estimates of the radiological benefit to be expected from partitioning and transmutation must take into account the mobilities of the elements in the geosphere, which may substantially modify the simple picture presented by the reduction in radiotoxic inventory alone. Other benefits, e.g. with respect to intrusion, may however be derived from the possibility of especially secure conditioning and emplacement of the long-lived radionuclides.

In order to fit the partitioning and transmutation option into the context of existing waste management strategies, the direct disposal concepts in granite formations of Spain and Sweden are briefly described as examples. A section discusses the issue of criticality safety in the disposal of spent fuel.

In the case of the reprocessing fuel cycle, the impact of the disposal of high- and medium-level waste as scheduled to be realised in Germany, Switzerland and Belgium, respectively in salt, granite and in clay, are given as illustrations. A number of options are being studied on disposal of waste forms resulting from the advanced fuel cycle incorporating partitioning and transmutation. Nevertheless, the report sheds some light on the technical and operational aspects of such a strategy; in particular,

intermediate storage management, the fate of irradiated targets and the residual reactor cores from a prolonged nuclear energy production.

In addition to the highly radioactive materials resulting from an advanced fuel cycle, attention should be drawn on the large inventories of depleted uranium produced during the enrichment processes. The depleted uranium stocks can in a short time interval be considered as a strategic material to be reused in case the FRs would become an important fraction of the total electricity generating capacity.

If the FRs do not emerge as nuclear electricity producers, depleted uranium will be considered as a waste material whose radiotoxicity will eventually become equal to that of natural uranium ore.